Moisture adsorption characteristics of freeze-dried composite tropical fruit purees and application to shelf life prediction in flexible packages

#### **Abstract**

The moisture adsorption characteristics of freeze-dried watermelon, orange, and mango composite fruit purees were determined using the standard gravimetric method at 20, 25, 30, 40 and 50°C respectively over a water activity range of 0.11-0.95 and the data analyzed using the BET, GAB and Oswin sorption models. The data obtained and moisture permeability characteristics of commercial aluminium pouches, low density polyethylene (LDPE),Melinex 813, and Propafilm Cflexible materialswere used for shelf life prediction at various ambient storage temperatures. Sorption data exhibited type III isotherms, typical of foods rich in soluble components. The GAB model provided the best fit for the experimental data compared to the other tested models. BET monolayer moisture contents (4.72 – 8.80 g  $\rm H_2O/100g$  solids) decreased with increasing temperature indicating reduced water binding sites at higher temperatures. Net isosteric heats (0.00012 to 0.0052 kJ/mol) decreased while the entropy of adsorption (-0.1343 to -0.0312kJ/mol). C) increased with moisture content. The predicted shelf life was highest for Aluminum pouch followed by, Propafilm C, LDPE andMelinex 813 in that order. This study provides valuable insights for optimizing storage conditions and packaging selection for freeze-dried composite fruit products, with practical implications for commercial applications in the food industry.

**Keywords:**Watermelon, orange, mango, fruits, freeze-dried purees,moisture adsorption isotherms, BET, GAB, Oswin models, flexible packaging, shelf life prediction,

#### 1.0 Introduction

The global market for convenience foods has witnessed remarkable growth, with particular emphasis on preserved fruit products that maintain their nutritional and sensory qualities. Among various preservation techniques, freeze-drying (lyophilization) has emerged as a premium dehydration method that vields highquality products with excellent rehydration properties and minimal thermal damage (Radojčin et al., 2021). Tropical fruits, particularly watermelon, orange, and mango, represent significant economic value in both fresh and processed forms due to their distinctive organoleptic properties and rich nutritional profiles, including vitamins, minerals, antioxidants, and bioactive compounds (Vicente et al., 2022). The freeze-drying process involves the removal of water through sublimation under vacuum conditions, resulting in a highly porous structure that preserves the original shape and volume of the product (Nowak & Jakubczyk, 2020). While this process yields superior-quality products, the resulting porous structure makes freeze-dried materials particularly susceptible to moisture absorption, which can lead to various degradation phenomena including loss of crispness, structural collapse, enzymatic reactions, and microbial growth (Ngamwonglumlert & Devahastin, 2018). Understanding moisture sorption characteristics is fundamental to predicting and controlling the stability of dehydrated food products during storage. Moisture sorption isotherms (MSI) describe the relationship between water activity (aw) and equilibrium moisture content at constant temperature, providing crucial information for process design, packaging selection, and shelf life prediction (Al-Muhtaseb et al., 2002; Yaptenco et al., 2017). In composite food systems, such as mixed fruit purees, the complexity of moisture sorption behavior increases dueto multiple component interactions and varying chemical compositions, including different sugar profiles, organic acids, and structural polymers (Shahidi & Pan, 2022). The mathematical modeling of moisture sorption isotherms has received considerable attention, with numerous theoretical, semi-empirical, and empirical models proposed to describe the relationship between equilibrium moisture content and water activity (Barbosa-Cánovas et al., 2020; Peleg, 2019). These models, including GAB (Guggenheim-Anderson-de Boer), BET (Brunauer-Emmett-Teller), and various modified equations, serve as valuable tools for predicting moisture transfer phenomena and product stability under varying environmental conditions (Aviara, 2020). Packaging plays a crucial role in protecting freeze-dried products from moisture ingress and maintaining their quality throughout the distribution chain. The selection of appropriate packaging materials involves consideration of multiple factors, including water vapor transmission rate (WVTR), oxygen permeability, mechanical properties, and cost-effectiveness (Wu et al., 2021). Flexible packaging materials such as aluminum pouches, low-density polyethylene (LDPE), and high-barrier films offer varying degrees of protection against moisture and oxygen transmission, with their effectiveness depending on environmental conditions and required shelf life (Wu et al., 2021). Recent advances in packaging technology have introduced various high-barrier materials and modified atmosphere packaging systems, yet the optimal selection of packaging materials requires a thorough understanding of product-package interactions and environmental factors (Lechevalier, 2016). The water vapor permeability of packaging materials, combined with the moisture sorption characteristics of the product, determines the rate of moisture uptake and consequently, the product's shelf life (Edrisi Sormoli & Langrish, 2015). Despite extensive research on individual freeze-dried fruits and their storage stability, there remains a significant knowledge gap regarding the moisture sorption behavior of composite tropical fruit purees and their interaction with different flexible packaging materials. The complex nature of mixed fruit systems, combined with the variety of available packaging options, necessitates a comprehensive study to optimize storage stability and predict shelf life accurately. This understanding becomes particularly critical given the increasing market demand for convenient, shelf-stable fruit products and the economic importance of minimizing quality deterioration during storage and distribution.

#### 2.0 Materials and Methods

### 2.1 Sources of raw materials and preliminary handling

About 20 kg each of fresh fully matured watermelon, oranges and mango frits were obtained from a popular local fruit market in Makurdi, Benue State, Nigeria and were thoroughly washed with tap water and kept in the refrigerator (10°C) prior to use within 6 h for composite puree production.

#### 2.2 Fruit Purees Production

### 2.2.1. Watermelon fruit puree production

After washing and sorting, the fruits were peeled manually using stainless steel knives, followed by slicing, removal of the seeds, and blending of pulps in a household electric blender (Kenwood Electricals, UK) at speed number 3 for 15s into smooth pastes, which were pasteurized at 70°C for 15s in 250 mL glass beakers with aluminum foil coverings. After cooling, the watermelon purees were kept in a refrigerator before use for composite puree formulation.

### 2.2.2 Production of orange fruits puree

Essentially, the fruits were sorted, washed, peeled and sliced using stainless steel knives. After the removal of the seeds, the slices were blended into a smooth paste using the house hold electric blender. The orange puree was then pasteurized at 70 °C for 15s in 250 ml glass beakers with aluminium foil covers. The pasteurized orange puree was rapidly cooled in an ice bath and promptly stored in a refrigerator prior to use for mixed puree formulation.

# 2.2.3 Production of mango fruits puree

The mango fruits were sorted, washed and blanched by immersion in a boiling hot water bath maintained at 98°C for 5min. The blanched mango fruits were then cooled in running tap water, peeled using stainless steel knives and the fleshy mesocarp sliced to obtain pieces which were

blended in the Kenwood mixer in the presence of 0.2 M citric acid buffer (pH 5.2) into a smooth slurry. The slurry was then stored in the freezer compartment of a household refrigerator prior to use for composite puree formulation.

### 2.3 Composite fruit purees formulation

The composite fruit puree compositions are shown in Table 1 Each puree type was treated with commercial maltodextrin as a carrier agent respectively to obtain a dextrose equivalent (DE) of 30 for each group. The composite purees together with the malodextrins were each blended into smoothies and subjected to preliminary sensory evaluation which indicated that the composite puree sample comprising 50% watermelon, 30% orange and 20% mango composite puree (code: 618) was the most acceptable smoothie and hence was used for the freeze drying experiment.

# 2.4. Freeze drying operations

Freeze drying of the composite fruits puree was as described by Varghese et al. (2014) using a pilot scale freeze dryer (model: Labconco Free Zone Triad, Freeze Dry System, Freeze Dry Ltd., Warwickshire, UK). The freeze-drying machine (with a Capacity of 18 <a href="https://liters.python.org/liters">https://liters.python.org/liters.python.org/liters</a> of ice condensing capacity) was

preheated to 50°C with an initial pressure of 0.030 mbar according to the manufacturer's instructions. The mixed fruit puree was poured into 250g capacity freezer bags, sealed and placed on freezer trays prior to loading in the freeze-drying chamber of the freeze dryer. The initial freezing was done to ~45 °C while during drying the temperature was increased up to 60 °C. A vacuum of 100 mmHg was maintained during freeze drying. The process was regularly monitored to ensure proper drying. The freeze-dried composite fruit puree powder containing 2–3 % moisture was allowed to cool to room temperature. The freeze dried puffy material was then blended in a household Kenwood dry mixer for 2min and the powder promptlypackaged in air-tight aluminum pouches which were then stored on dry shelves in glass desiccators containing activated silica.

Table 1: Composite purees formulation

| Sample | Puree composition (%) |        |       |  |  |  |
|--------|-----------------------|--------|-------|--|--|--|
| Code   | Watermelon            | Orange | Mango |  |  |  |
| 573    | 30                    | 50     | 20    |  |  |  |
| 618    | 50                    | 30     | 20*   |  |  |  |
| 335    | 20                    | 50     | 30    |  |  |  |
| 804    | 50                    | 20     | 30    |  |  |  |
| 732    | 20                    | 30     | 50    |  |  |  |
| 408    | 30                    | 20     | 50    |  |  |  |

<sup>\*</sup>Most acceptable sample upon which quality analyses was carried out

# 2.5. Measurement of Equilibrium Moisture Content and Water Activity

Equilibrium moisture content was determined gravimetrically by exposing the samples to atmospheres of known relative humidities following the method described by (Ariahu et al., 2005). Sulphuric acid (H<sub>2</sub>SO<sub>4</sub>) solutions of 10, 20, 30, 40, 50 and 60 % were used to provide water activities ranging from 0.15 to 0.96. A thermostatically controlled biochemistry incubator (Model: SPX-80-II, Searchtech Instruments) and 500 mL plastic containers were used for temperature and humidity controls respectively. The solutions made from the acid (200mL each) were carefully introduced into the plastic containers. A screen made of wire gauze was arranged in the plastic containers above the acid solutions to provide a platform for the samples to rest. The experiment was carried out at four temperatures (20, 25, 30, 40 and 50 °C respectively). The equilibrium moisture contents were determined by material balance from the initial moisture content.

### 2.6. Moisture sorption models

The equilibrium moisture data was fitted using the Brunauer–Emmett–Teller (BET), GAB and Oswin models. These models were chosen for their reported fit for starchy foods. They are the most widely used model that gives a good fit to data for a variety of foods over the  $a_w$  region of 0.05 to 0.45 (Chirife& Iglesias, 1978; Rizvi, 1995) in (Ocheme et al., 2013). The models are;

BET Model: 
$$\frac{a_w}{(1-a_w)M} = \frac{1}{M_0C} + \frac{(C-1)a_w}{M_0C}$$
 (1)

GAB Model: 
$$M = \frac{M_0 G k a_w}{(1 - k a_w)(1 - k a_w + G k a_w)}$$
 (2)

Oswin Model: 
$$M = A \left[ \frac{a_w}{1 - a_w} \right]^B$$
 (3)

where; M = equilibrium moisture content,  $M_0 =$  monolayer moisture content and A, B, G, K and C are constants related to heat of sorption, aw = water activity

The isotherm model parameters were obtained using OriginLab 2024 through linear and polynomial regression methods as the case may be.

The goodness of fit of different models used was evaluated using percent root mean square of error (%RMS) between experimental ( $M_{obs}$ ) and predicted ( $M_{est}$ ) moisture contents as described by Sengev et al. (2016).

$$\%RMS = \frac{\sqrt{\sum \left[\frac{M_{obs} - M_{est}}{M_{obs}}\right]^2}}{2} \times 100 \tag{4}$$

The net isosteric heats of sorption was calculated by applying Clausius-Clapeyron equation to the isosteres obtained at constant moisture content following the procedure reported by Ariahu et al. (2005).

$$\ln a_w = C_{st} - \frac{\Delta H_{st}}{R} \frac{1}{T} \tag{5}$$

By plotting In (aw) versus inverse absolute temperature (1/T) for a specific moisture content,  $\Delta H_{st}$  was evaluated from the slope (- $\Delta H_{st}/R$ ) with R = universal gas constant ( 0.008314 kJ/mol.deg). The differential entropies of sorption ( $\Delta S^{\circ}$ ) were obtained from the intercept, C, coefficient ( $\Delta S^{\circ}/R$ ) of the same plot. Applying this at different moisture content allowed the dependence of  $\Delta H_{st}$  and  $\Delta S$ 0 with moisture to be determined.

# 2.7 Application of moisture sorption to shelf life prediction

The relationship between effective diffusivity and storage temperature at 85 % RH was evaluated. From the parameters obtained from the packaging films (Aluminum Pouch, LDPE, Melinex 813 and Propafilm C) and sorption data, the shelf life of the instant *fufu* flour samples can be predicted following the expression for shelf life predictions by Karel (1975) as cited in (Offiah et al., 2016).

$$\ln \frac{M_e - M_i}{M_e - M_c} = \left(\frac{B}{x}\right) \left(\frac{A}{W_c}\right) \left(\frac{P_0}{b}\right) t_s \tag{6}$$

where;  $M_e$ =equilibrium moisture content at outside relative humidity (obtained from the BET sorption isotherms at various storage temperatures),  $M_i$ =Initial moisture content of packaged food,  $M_c$ =Critical moisture content of packaged food (from sorption isotherm), B=moisture permeability of packaging materials, b=slope of sorption isotherm,  $W_s$ =weight of solid food, A=package surface area, x=thickness of the packaging material,  $P_o$ =vapor pressure of pure water at ambient temperature,  $t_s$ =shelf life.

A working isotherm straight line equation derived from a given sorption isotherm is given as;  $M = Y + ba_w$  (7)

where b=slope, Y=intercept at a<sub>w</sub>=0 (representing initial moisture content, M<sub>i</sub>), M=moisture content (gH<sub>2</sub>O/100 g solids) at given a<sub>w</sub>. The water vapour permeability of the different packaging films at 85 % RH are given as follows; Aluminum pouch (0.098 gH<sub>2</sub>O/m<sup>2</sup>.day.mmHg), low-density polyethylene (0.8169gH<sub>2</sub>O/m<sup>2</sup>.day.mmHg), melinex 813 (0.88 gH<sub>2</sub>O/m<sup>2</sup>.day.mmHg), propafilm C (0.122 gH<sub>2</sub>O/m<sup>2</sup>.day.mmHg). The surface area of each packaging material was taken as 0.25 m<sup>2</sup> with the weight of the sample to be packaged being 500 g.

### 3.0 Results and Discussions

### 3.1. Effect of temperature and water activity on moisture sorption isotherms

The moisture sorption isotherms of the freeze-dried tropical fruit purees are presented in Figure 1. At all temperatures, the equilibrium moisture content was observed to be increasing as the water activity increased; as opposed to a decrease in the moisture content with an increase in temperature. The sorption isotherms were of type III (J-shaped), also known as the Flory-Huggins isotherms, which have been reported to be relatively rare in occurrence. This shape of the curves is characteristic of foods with high sugar contents, which sorbs small amounts of water at low water activity and large amounts of water at high relative humidity(Ocheme et al., 2013). The J-shaped exhibited by the freeze-dried fruit puree composites could be due to the high sugar content in the composites as a result of the sugar composition in the individual purees(Vicente et al., 2022). The observed differences in moisture sorption capacity can be explained by the differences in bulk porosity and the pore size of the dried materials. Freeze-drying resulted in a highly porous product with small pores, which sorbed more water (Tsami et al., 1998). The explanation for the nature of the isotherms may be that at low water activities, physical sorption on strongly active sites occurs since water can be sorbed only to surface - OH sites of crystalline sugar. The decrease in equilibrium moisture content with an increase in temperature is consistent with other researchers who observed similar trends (Ocheme et al., 2013; Pedro et al., 2010; Sengev et al., 2016). This could be due to the reduced affinity of water molecules for the product. This trend is related to the Formatted: Left, Line spacing: Multiple 1.08 li, Tab stops: Not at 2.42"

excitation state of molecules reported by Diosady et al in (Ariahu et al., 2005). The attractive forces between molecules are lower at high temperatures due to an increase in the kinetic energy of water molecules, allowing the connection between moisture and sorption sites to be broken, which reduces the moisture content of the product (Argyropoulos & Müller, 2014; Jebitta et al., 2022). Dalgiç et al. (2012) have also reported similar results for freeze-dried mint leaves. Though there was an observance

of an increase in moisture content with an increase in water activity, the increase was more profound at water activity in the range of 0.5 to 1.0. This would imply that at water activity >0.5, microbial growth, enzymatic reactions and lipid oxidation will occur much faster leading to rapid spoilage of the freeze-dried fruit puree composites.

# 3.2 Model regression parameters and derivatives

The different model regression parameters are presented in Tables 2, 3 and 4 for the GAB, BET and Oswin models respectively. It was observed that the monolayer moisture contents decreased with increase in temperature. Mo values are particularly important for determining the storage and preserving conditions above which deteriorative changes may occur. It represents the minimum amount of water bound to active sites to give a monolayer coverage and maximum stability of storage foods (Ariahu et al., 1999).

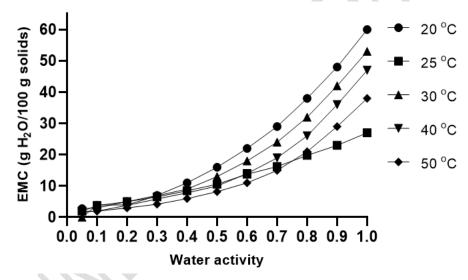


Figure 1: Moisture sorption isotherms of freeze-dried tropical fruit composite purees

The BET and the GAB monolayers were observed to be in the range of 5.80-11.27 gH $_2$ O/100 g solids and 4.27-88 gH $_2$ O/100 g solids respectively with the BET monolayers being significantly lower than the GAB monolayer values. These values were higher than those reported for mint leaves (3.64-7.15 gH $_2$ O/100 g solids) by Dalgıç et al. (2012). This reduction in monolayer values could be explained by the reduction in the total number of active sites for water binding as a result of physical and/or chemical changes in the product induced by temperature during the freezing process. Several researchers have observed similar scenarios (Rakshit et al., 2014; Sengev et al., 2018; Xu et al., 2011).

### 3.3. Fitting of sorption models

Sorption experimental data obtained for the 5 temperatures studied were fitted to 3 models namely BET, GAB and Oswin models over the theoretically appropriate ranges at 20 °C, 25 °C.30 °C and 40 °C and 50 °C. The quality of the fitting for each proposed model was evaluated through the correlation coefficient (r²)

and the values are shown in Tables 2, 3 and 4 respectively for BET, GAB, and Oswin models. The  $r^2$ indicates how well a given model explains the variability of the predicted values indicating that if the correlation coefficient is close to one the model is suitable (Gichau et al., 2020). The  $r^2$ values were 0.83-0.94 for GAB model, 0.55-0.77 for BET with 0.61-0.87 for Oswin respectively. Earlier reports e.g. Ariahu et al (2005) indicated that the BET model is valid at aw  $\leq 0.5$  and hence gave the least fitacross the entire aw range studied. However, the BET isotherm is useful for estimating monolayer moisture contents and surface areas of sorption within the valid range. This is consistent with studies by Pedro et al. 2010) who evaluated the sorption isotherms of passion fruit pulp and alsoVillegas-Santiago et al. (2020) for the drying process of coffee extract.

Table 2: GAB Adsorption Regression Parameters for freeze-dried composite tropical fruit purees

| Temperature | K    | G    | M <sub>o</sub> | r²   |
|-------------|------|------|----------------|------|
| 20          | 0.87 | 3.72 | 11.27          | 0.94 |
| 25          | 0.71 | 5.95 | 8.97           | 0.87 |
| 30          | 0.92 | 9.10 | 7.32           | 0.90 |
| 40          | 0.89 | 3.97 | 7.20           | 0.83 |
| 50          | 0.89 | 4.00 | 5.80           | 0.83 |

r<sup>2</sup>=correlation coefficient, M₀=monolayer moisture content (g H₂O/ 100g solids), G and K are GAB constants

Table 3: BET Adsorption Regression Parameters for freeze-dried composite tropical fruit purees

| Temperature | С     | Mo   | So     | r <sup>2</sup> |
|-------------|-------|------|--------|----------------|
| 20          | 4.36  | 8.80 | 308.97 | 0.65           |
| 25          | 7.70  | 6.01 | 211.24 | 0.63           |
| 30          | 10.80 | 6.22 | 218.33 | 0.77           |
| 40          | 4.85  | 5.73 | 201.32 | 0.64           |
| 50          | 4.68  | 4.72 | 165.79 | 0.55           |

 $\vec{f}$ =correlation coefficient,  $M_o$ =monolayer moisture content (g  $H_2O$ / 100g solid),  $S_o$ =surface area of sorption ( $m^2$  /100 g solids), C= BET constant

Table 4: Oswin Adsorption Regression Parameters for freeze-dried composite tropical fruit purees

|             |      | VIII | "00000"        |
|-------------|------|------|----------------|
| Temperature | Α    | В    | r <sup>2</sup> |
| 20          | 0.29 | 6.80 | 0.87           |
| 25          | 0.21 | 5.52 | 0.70           |
| 30          | 0.31 | 5.16 | 0.72           |
| 40          | 0.30 | 4.50 | 0.61           |
| 50          | 0.30 | 3.63 | 0.61           |

A and B are Oswin constants,  $r^2$ =correlation coefficient.

# 3.4. Thermodynamic properties of freeze-dried composite tropical fruit purees

Net isosteric heat of sorption: The net isosteric heats and entropies of sorption produced from the slopes and intercepts of the isosteres are shown in Figure 2, respectively, using the relationship between Clausius-Clapeyron and water activity. The net isosteric heat ( $\Delta H_{st}$ ) of sorption generally decreased with an increase in EMCs. The net isosteric heats of adsorption decreased from  $5.2\times10^{-4}$  kJ/mol to  $1.2\times10^{-4}$  kJ/mol. The high correlation coefficients ( $r^2 \ge 0.842$ ) confirmed the validity and usefulness of the Clausius-Clapeyron equation in describing the sorption energetics of the freeze-dried tropical fruit puree composites (Table 5). The heat of sorption decreased with an increase in moisture content until a moisture content of about 6 g  $H_2O/100$  g solids, when the rate of decrease fell and approached an asymptotic value. This trend is consistent with that of fish flour (Dalgıç et al., 2012). Net heat of sorption was considerably high at lower moisture contents and it decreased; approaching the latent heat of vaporization of pure water ( $\Delta H_{si}=0$ ) as moisture content increased. This means as moisture content increases, the heat of sorption tends to that of pure water; an indication of the moisture existing in the free form. This highlights the strong dependence of isosteric heat on moisture content; with the energy required for sorption (in excess of the latent heat) increasing at low moisture content (Yazdani et al., 2006). This also means that the heat of adsorption of the first few layers of water molecules was more

than the heat of condensation of pure water. This could be due to differing strengths of water binding; initial occupation of highly active polar sites (Pedro et al., 2010)on the surface (with greatest activation energy), followed by the progressivefilling of the less available sites with lower bonding activation sites (Argyropoulos & Müller, 2014; Udomkun et al., 2015). The heat of sorption values at specific moisture contents provides insight into the state of the sorbed water and hence measures the physical, chemical and microbiological stability of the food material under given storage conditions. This variation in heats of sorption with moisture content has been important in providing valuable data for energy consumption calculations and subsequent drying equipment, and a knowledge of the extent of water-water vs water-solid interactions. Water sorption in foods is complex; given the main water-sorbing constituents of foods including proteins, starch, cellulose, sugars etc each having different polar groups which provide different energetically preferential sites for water(Labuza & Altunakar, 2020).

Table 5: Regression parameters for Clausius–Clapeyron equation relationship between Inawand 1/T for adsorption isotherms

| Regression      | Moisture content (gH₂O/100 g solids) |         |         |         |         |         |         |         |
|-----------------|--------------------------------------|---------|---------|---------|---------|---------|---------|---------|
| parameter       | 2                                    | 3       | 4       | 5       | 6       | 8       | 10      | 12      |
| N               | 5                                    | 5       | 5       | 5       | 5       | 5       | 5       | 5       |
| Α               | -0.062                               | -0.030  | -0.028  | -0.025  | -0.022  | -0.019  | -0.016  | -0.014  |
| В               | 16.15                                | 6.899   | 6.562   | 6.044   | 5.42    | 4.82    | 4.248   | 3.751   |
| r <sup>2</sup>  | 0.842                                | 0.981   | 0.996   | 0.997   | 0.980   | 0.992   | 0.942   | 0.860   |
| $\Delta H_{st}$ | 0.00052                              | 0.00025 | 0.00023 | 0.00021 | 0.00018 | 0.00016 | 0.00014 | 0.00012 |
| ΔS°             | -0.1343                              | -0.0574 | -0.0546 | -0.0502 | -0.0451 | -0.0401 | -0.0353 | -0.0312 |

n = no. of entries; r' = correlation coefficient; b = intercept coefficient; a = slope;  $\Delta H_{st} = net$  isosteric heat(kJ/mol);  $\Delta S^{\circ} = net$  entropy of sorption (kJ/mol.°C).

Entropy of sorption: The entropy of sorption  $\Delta S^{\circ}$  values were evaluated from the intercept coefficients derived from least square linear regression of the relationship between  $\ln a_w$  and 1/T at constant moisture contents. From the Clausius-Clapeyron equation,  $\Delta S^{\circ}$  was calculated from  $C_{st}$ , where  $C_{st} = \frac{\Delta S^{\circ}}{R}$ . The relationship between the entropies of sorption ( $\Delta S^{\circ}$ ) and equilibrium moisture contents (EMC) are shown in Figure 2. Negative values which decreased with increase in moisture content were obtained. This relates to the thermodynamic compensation between the heat and entropy of moisture sorption by the sample. At lower moisture content values, the water molecules are tightly bound on the sorbent surfaces and therefore have low degree of freedom resulting in low entropy of sorption. At higher moisture contents, the water molecules are sorbed on multilayer on the top of the tightly bound first layer. The multilayer water molecules have greater degree of freedom and hence higher entropy of sorption (Li et al., 2011; Yang et al., 2016).

The entropy of sorption of the freeze-dried tropical fruit purees increased up to 6% moisture content and became asymptotic as the moisture content increased. It could be speculated that the increase in moisture content and availability of hydrophilic sites balanced each other, thereby resisting marked increase or decrease in entropy. The asymptotic tendencies of various products have been reported elsewhere in literatures (Ariahu et al., 2005; Li et al., 2011; Sengev et al., 2018).

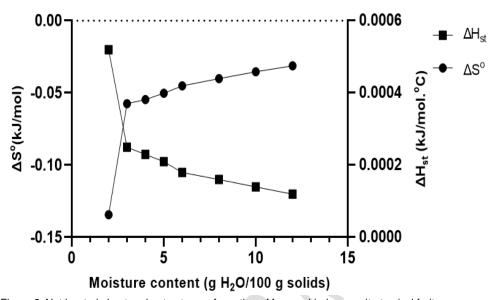


Figure 2: Net isosteric heat and net entropy of sorption of freeze-driedcomposite tropical fruit purees

## 3.4 Shelf life Prediction of freeze-dried composite Tropical fruit purees

The predicted Shelf life (months) of freeze-dried watermelon, orange and mango puree powders at 85 % Relative Humidity is shown in Figure 3. As expected, the product shelf-life decreased with an increase in storage temperature in all the packages. Storage conditions influence the shelf life stability of products especially high ambient temperature and relative humidity is a challenge for storage of dried products (Anandito et al., 2017). Storing at higher temperature is predicted to reduce shelf life due to increase transmission rate of water vapour. Irrespective of the temperature, Aluminum pouch was predicted to give a longer shelf life due to its low vapour transmission rate (0.122 gH<sub>2</sub>O/m<sup>2</sup>.day.mmHg). At 20 °C, for example and as predicted (Figure 3), a shelf life of 53.34 months can be attained for aluminium pouch; which is highest as opposed to the other packaging films with higher water vapor permeability. This estimation of the shelf life focused on the permeability of the packages to water vapour but permeability dynamics to gases; i.e. oxygen and carbon dioxide which may cause lipid oxidation need to be investigated(Gichau et al., 2020).

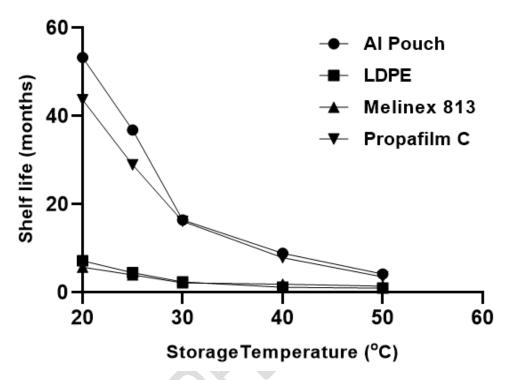


Figure 3: Shelf life Prediction of freeze-dried composite tropical fruit purees

### 4.0 Conclusion

The moisture sorption characteristics and packaging stability of freeze-dried composite tropical fruit purees were systematically investigated in this study. The sorption isotherms exhibited Type III behavior, characteristic of food products rich in soluble components, indicating strong solute-solvent interactions in the system. The temperature dependence of the sorption process was evidenced by the decrease in monolayer values with increasing temperature, suggesting reduced availability of active sites for water binding at elevated temperatures. The GAB model demonstrated superior fitting to the experimental data compared to other mathematical models, providing a reliable tool for predicting moisture adsorption behavior in these composite systems. Aluminum pouch exhibited superior performance in terms of moisture barrier properties compared to Propafilm C, LDPE, and Melinex 813 for shelf life extension of the freeze dried fruit puree.

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