International Journal of Farming and Allied Sciences

Available online at www.ijfas.com

©2012 IJFAS Journal-2012-1-3/72-76

ISSN 0000-0000 ©2012 IJFAS

Evaluating Monolayer Moisture Content of Rubber Seed using BET and GAB Sorption Equations

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ABSTRACT: In this investigation, the monolayer moisture content of rubber seed was evaluated from Brunauer, Emmett and Teller (BET) and Guggenheim-Anderson-de Boer (GAB) sorption equations at 25^0C for assessment of its storage stability and accessibility. The equilibrium moisture contents of rubber seed were found to vary from 10.01 to 17.37% (d.b.) after two months exposure on bare floor at 25^0C. The monolayer moisture contents were found to be 16.47 % (d.b.) and 11.75 % (d.b.) using BET and GAB sorption equations in the range of water applicability of 0.2-0.4 and 0.3-0.9, respectively. All the equilibrium moisture contents, with the exception of the initial, evaluated from the GAB equation were slightly higher than the monolayer value. The level of deterioration of the stored seeds, as indicated by mould growth, increased with increase in moisture content. Hence, it is better to store rubber seeds at moisture content slightly above their corresponding monolayer value for extending their shelf lives.

***Keywords*:** Evaluating, storage, Sorption equations, Shelf life, Monolayer, Rubber seed

INTRODUCTION

Oilseed is an agricultural commodity that plays an important role in Nigerian economy. Rubber seed is one such commodity, obtained as ancillary product of the rubber tree (*Hevea brasiliensis*). Rubber seed oil has many areas of potential applications such as its use as biodiesel in renewable energy industry and as additive for use in paint manufacturing industry among many others (Fadeyibi and Osunde, 2011). Rubber seed contained high level of moisture (29.7%) at the time of harvest (between August and September) in Niger Delter Nigeria (Oteide and Begho, 1986; George and Kuruvilla, 2000). Oteide and Begho (1986) reported a moisture drop of 8.4% of the seed after two months exposure on bare flow. Therefore, drying and storage conditions are very important to protect the quality of the seed. Drying is essential immediately after harvest to prevent deterioration and seed spoilage before safe storage can be effected (Giner and Gely, 2005).

Knowledge of the sorption characteristics of agricultural seeds is necessary in understanding the stability and acceptability of the plant materials in storage. More so, it will help in modelling the drying process, designing and optimizing the drying equipment, aeration, calculating moisture content changes occurring in storage, and in selecting appropriate packaging materials (Samapundo et al., 2006, Corina *et al.*, 2009). Ajisegiri and Chukwu (2004) reported post-harvest stability of most agricultural seeds to depend partly on their moisture contents at harvest and partly on their moisture-sorption behaviours. Hence, storage stability of the seeds is greatly affected by moisture depending on the chemical composition and physical structure of the plant material.

The moisture-sorption of most seeds and grains and their associated models have been investigated in literature (Khalilihqdam *et al.*, 2012). The ability of some of these models to predict moisture content during storage under a variety of conditions is important for reducing cost and cycle time of process development. Therefore, the objective of this study was to evaluate the monolayer moisture content of rubber seed using BET and GAB sorption equations at 25^{0}C for assessment of its storage stability and accessibility.

MATERIAL AND METHODS

Sample preparation

The rubber seeds used in this investigation were obtained from the plantation of the Rubber Research Institute of Nigeria. The seeds were left exposed on bare floor for a period of two months to remove residual latex (Figure 1). The initial moisture content of the sample was determined, in triplicates, in an air circulated oven dryer for 24h at 105^{0}C and the average value was found to be 10.01% (d.b.). The final moisture contents were also determined as 12.74, 14.16 and 17.37 % (d.b.) in triplicates, by adding calculated amount of distilled water and storing in an ambient environment.

Each sample of different moisture content was stored in a standard room for thirty days. The severity of the damage caused by mould growth on the seeds in storage was noted at the various moisture contents. Also, Brunauer, Emmett and Teller (BET) and Guggenheim-Anderson-de Boer (GAB) sorption equations were used to evaluate the moisture sorption data of the seeds at 25^{0}C and at their corresponding range of water applicability.

Sorption Isotherm

The water sorption isotherm is the relation between the equilibrium moisture content of a material (expressed as mass of water per unit mass of dry matter) and water activity, at a given temperature. Such relationships are the key to understanding the water sorption properties of agricultural produce, being of particular value when selecting suitable packaging materials and predicting stability and moisture changes during storage. Since water activity is temperature dependent, it follows that temperature has a significant effect on sorption isotherms. So, when a material is subjected to an upward temperature shift, at any constant moisture content, water activity increases with increasing temperature (Andrade *et al.*, 2011).

The moisture sorption isotherm may be measured by addition of water to a dry sample (adsorption) or by removal of water from a wet sample (desorption). It has been established that in hygroscopic high pectin and sugar-containing materials the sorption-desorption isotherms are sigmoidal in shape and show marked hysteresis. Typically, this means that for values of water activity below 0.40 the water content of the agricultural produce will be greater during desorption than during adsorption. Several explanations have been proposed for the hysteresis phenomenon. One theory is based on the availability of active polar sites for the bonding of water molecules. Under this theory, in the original wet condition, the polar sites in the molecular structure of the material are almost entirely satisfied by adsorbed water. Upon drying and shrinkage, the molecules and their water-holding sites are drawn closely enough together to satisfy each other. This reduces the water holding capacity of the material upon subsequent adsorption. Another theory is related to the glass transition concept. Upon drying high sugar-containing materials the sugars may be converted to an amorphous form. At low water contents the glass transition may occur at about room temperature, being the transition between glassy and rubbery states one of the main reasons for hysteresis. In the rubbery state the diffusion of water to the product will be enhanced (Andrade *et al.,* 2011).

The sorption isotherm describes the interaction between water and the agricultural product. Equations for fitting these data are of special interest in many aspects of seed preservation by dehydration. Numerous mathematical equations have been reported in literature for describing water sorption isotherms of agricultural materials. They vary a lot in terms of origin (empirical, semi-empirical or theoretical) and range of applicability (Andrade *et al.*, 2011).

The high number of sorption equations developed suggests the difficulty of having a unique mathematical model for describing the sorption data in the whole range of water activity for different products (Andrade *et al.*, 2011). This is due to the following reasons:

1. The depression of water activity in seed is due to a combination of factors each of them being predominant in a given range of water activity. In general, the insoluble macromolecules dominate the sorption behaviour at low water activity and the soluble components exert their effects largely through their colligative properties over the high water range.
2. Moisture sorption isotherms of seeds represent the integrated hygroscopic properties of numerous constituents whose sorption properties may change as a consequence of physical and/or chemical interactions induced by heating or other pre-treatments.
3. As a seed sorbs water, it usually undergoes changes of constitution, dimensions and other properties; water sorption also leads to phase transformations of the sugars contained in the seed. When a sugar containing product is freeze dried, the sugars can be converted to an amorphous form. When adsorption takes place, the sugars will slowly reconvert to the crystalline form.

The choice of an equation to fit the sorption data takes into account different factors:

1. The agreement between the sorption data and the model
2. The range of applicability
3. The theoretical basis of the parameters

4. The simplicity
5. The desired objectives.

Evaluating Model parameters of rubber seed using BET Equation

Brunauer, Emmett and Teller (BET) equation was originally developed in 1938 on the thermodynamic principles for the physical adsorption of non-polar gases on homogenous metal surfaces. The equation is useful for determining the optimum moisture conditions for good storage stability of agricultural material. The BET equation represents a basis in the interpretation of isothermal sorption multilayers and it has been applied in gas adsorption and porous steam in surfaces and solids, as well as in water, especially in steam absorption, by homogenous polymers and other materials. Nevertheless, the considerable success of the isotherm is rather qualitative than quantitative (Andrade *et al.*, 2011). The model is shown in Equation (1).

$$\frac{a_w}{(1-a_w)M} = \frac{1}{M_0C} + \frac{(C-1)a_w}{M_0C} \quad (1)$$

Where: M = Moisture content in % (d.b.), M_0 = monolayer moisture content in % (d.b.), C = Energy constant related to net heat of sorption, a_w = water activity in the range 0.1-0.5

Applying this equation at 0.2 and 0.4 a_w of rubber seed (chosen from range of applicability of the BET equation), and taking M_0 as the initial moisture content of rubber seed, the following equations were obtained and solved simultaneously.

$$M_0C = 32 + 8C \quad (2)$$

$$M_0C = 9 - 6C \quad (3)$$

The procedure was repeated for the three other moisture values, each time evaluating the moisture sorption data of rubber seed.

Evaluating Model parameters of rubber seed using GAB Equation

Guggenheim-Anderson-de Boer (GAB) equation was developed in 1966. The equation postulates that the state of sorbate molecules in the second layer is identical to the one in superior layers, but is different from those of the liquid state. GAB equation has been used due to its theoretical bases as it described the sorption behaviour in a wide range of water activity (0 - 0.9). Thus, it was found to be suitable for analysing most agricultural materials (Andrade *et al.*, 2011). Equation 4 expressed the GAB equation.

$$M = \frac{M_0CKa_w}{[(1-Ka_w)(1-Ka_w+CKa_w)]} \quad (4)$$

Where: C and K are sorption constants of the GAB model

Applying this equation at 0.3, 0.6 and 0.9 a_w of rubber seed (chosen from range of applicability of the GAB equation), and taking M_0 as the initial moisture content of rubber seed, the GAB equation was reduced to the following.

$$M_0CK = 33.33(1-0.3K)(1-0.3K+0.3CK) \quad (5)$$

$$M_0CK = 16.67(1-0.6K)(1-0.6K+0.6CK) \quad (6)$$

$$M_0CK = 11.11(1-0.9K)(1-0.9K+0.9CK) \quad (7)$$

Equations (5), (6) and (7) were reduced to the following equations which were then solved simultaneously.

$$K^2(1+C) = 5.56 \quad (8)$$

$$K^2(C-1) = -1.85 \quad (9)$$

The procedure was repeated for the three other moisture values, each time evaluating the model parameters of rubber seed.

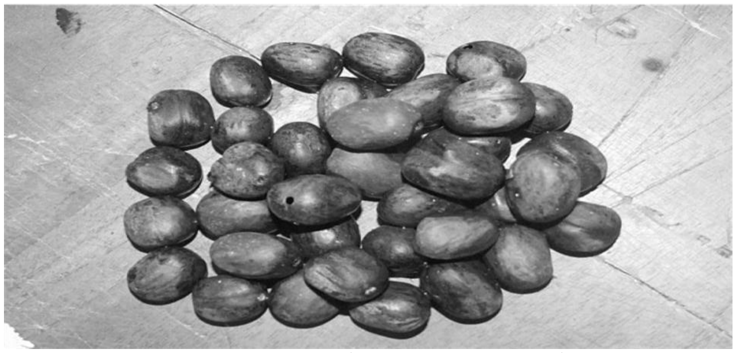

Figure 1. Rubber Seed after two months on bare-floor

RESULTS AND DISCUSSION

Table 1 showed the moisture sorption data of rubber seed evaluated between 0.2 to 0.4 and 0.3 to 0.9 water activities from BET and GAB equations, respectively. The data evaluated from the GAB equation were the same for all equilibrium moisture content of the seed. The level of deterioration, as indicated by the presence of mould on the seeds, increases with increase in moisture content. This was because the equilibrium moisture contents of rubber seed were higher than the monolayer moisture content evaluated from the GAB equation. However, BET equation showed monolayer moisture content to generally decrease from 16.49 to 9.06% (d.b.) with increase in equilibrium moisture contents from 10.01 to 17.37 % (d.b.). Kumar and Balasubrahmanyam (1986) reported moisture level between the monolayer value and that corresponding to the water activity as safest for good storage stability of agricultural material. They further stressed that when drying to low water levels, especially below the monolayer value, it is necessary to supply large amount of heat in addition to the heat of vaporization. Hence, it is preferable to have the moisture content at or slightly above the monolayer value for extending the shelf-life of rubber seed in storage.

The relation between equilibrium moisture content of rubber seed and water activity was shown, according to GAB model, in the following equations:

$$M = \frac{10.869a_w}{[(1-1.85a_w)(1-1.85a_w+0.925a_w)]} \quad (10)$$

Kumar and Balasubrahmanyam (1986) reported high water activity as the main influential factor in product deterioration occurring in form of non-enzymatic browning, lipid oxidation, enzymic activity, loss of colouring pigments, as well as texture and other qualities. From the GAB equation, it was seen that the growth of mould on stored rubber seeds was present in the range of 10.01-17.37% moisture content corresponding to 0.3- 0.9 water activity. Pixton (1982) in a similar study reported that various species of drought-resistant fungi develop in cereal grains and pulses at moisture levels of 13-16% corresponding to 0.6-0.75 water activity.

Table 1. Moisture sorption Data of Rubber seed

	Equilibrium Moisture Content, % (d.b.)			
Sorption Data	10.01	12.74	14.16	17.37
BET Equation	0.2-0.4 Water Applicability			
M_0	16.49	8.6	15.27	9.06
C	-1.643	11.5	11.5	-11.5
GAB Equation	0.3-0.9 Water Applicability			
M_0	11.75	11.75	11.75	11.75
C	0.5	0.5	0.5	0.5
K	1.84	1.84	1.84	1.84
Mould growth	Less Severe	Less Severe	Less Severe	Severe

CONCLUSION

The monolayer moisture content of rubber seed was evaluated using BET and GAB sorption equation at 25^{0}C and the following conclusions were made from the study:

1. The equilibrium moisture contents of rubber seed varied from 10.01 to 17.37% (d.b.) after two months exposure on bare floor at 25^{0}C. The monolayer moisture contents were 16.47 % (d.b.)

and 11.75 % (d.b.) from BET and GAB sorption equations in the range of water applicability of 0.2-0.4 and 0.3-0.9, respectively.

2. The evaluated monolayer moisture content from GAB equation was a better estimation because it was constant at all equilibrium moisture content. However, the inability of the BET equation to predict constant monolayer value at all equilibrium moisture content renders it unsuitable.
3. All the equilibrium moisture contents, with the exception of the initial, evaluated from the GAB equation were slightly higher than the monolayer value. The level of deterioration of the stored seeds, as indicated by mould growth, increased with increase in moisture content. Hence, it is better to store rubber seeds at moisture content slightly higher than their corresponding monolayer value for extending their shelf lives.

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