Neuro Fuzzy Modeling Approach for Prediction of Equilibrium Moisture Characteristics and Shelf-life of Corn Flour

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Abstract: Corn flour was studied to determine the physical properties such as equilibrium moisture Content, Equilibrium Relative Humidity (ERH), initial moisture content and critical moisture content. These were experimentally determined using the static gravimetric method at 4 temperatures, ranging from 20-50°C and at various ERH values ranging from 0.113-0.9812. Two mathematical models- Neuro-fuzzy and Gab were used to fit experimental isotherm data. The 2 models were used to predict shelf life of the corn flour. Comparisons were made to determine the validity and suitability of the 2 models. Within the range of temperatures investigated, GAB and Neuro-fuzzy models better describes the experimental data for corn flour for adsorption isotherm. Neuro-fuzzy model not only accommodated temperature and water activity parameter but is also better than GAB model in the prediction of corn flour shelf-life.

Key words: Modeling, neuro-fuzzy, gab, moisture content, relative humidity, sorption isotherm

INTRODUCTION

Moisture content of food is very important with respect to the properties of food systems. The presence of water in food influences physical or textural characteristics of the product as well as its chemical stability (Bell and Labuza, 2000). This stability is as a result of the relationship between the Equilibrium Moisture Content (EMC) of the food material, and its associated water activity (equilibrium relative humidity of the surrounding environment at constant temperature) at a given temperature. Moisture Sorption Isotherm (MSI), which is a plot of Equilibrium Moisture Content (EMC) of product against water activity (a,,), is unique for individual food materials and has a direct use in solving food processing design problems, in predicting energy requirements determining proper storage conditions (Myara et al., 1996). Many empirical and semi-empirical equations which describe the sorption characteristics of foods have been proposed in the literature. This is attributed to the water association with the food matrix by different mechanisms in different water activity regions (Labuza, 1975). There are several categories of Models available in the literature which have been used to describe MSI, among these are kinetic models based on a mono-layer (Mod-BET model), kinetic models based on a

multi-layer and condensed film (GAB model), semiempirical (Ferro-Fontan, Henderson and Halsey models) and empirical models (Smith and Oswin models). Iglesias and Chirife (1995) reported that the Ferro-Fontan equation accurately represented the sorption isotherm of 92 different food products. Peleg (1993) proposed a four parameter model and noted that the model can be used for both sigmoid and non-sigmoid isotherms, and that it fitted as well as, or better than, the GAB model. Smith (1947) model is useful in describing the sorption isotherm of biological materials such as starch and cellulose. Henderson (1952) proposed a semi-empirical model for the equilibrium moisture content of cereal grains. Chirife and Iglesias (1978) found that Halsey and Oswin models are also versatile, they reviewed 23 equations existing in the literature for fitting moisture sorption isotherms of foods and food products. Later, Boquet et al. (1978) evaluated eight equations for 39 different foods. Van den Berg and Bruin (1981) collected and classified 77 such equations. In all these studies, the researchers reported equations which gave the best fit to food isotherms. Among the sorption models, the Guggen-heim-Ander-son de Boer (GAB) equation has been applied successfully to various foods (Van den Berg, 1985) and has been considered and adopted by a group of European food researchers COST 90 (Bizot, 1983; Vanden Berg and Bruin, 1981) as the most versatile sorption model available in the literature. The three parameter GAB model for multilayer adsorption which is based on Brunauer Emmett Teller (BET) theory was reported to fit sorption data over a wider range of aw than the widely used BET equation. Temperature variation is not included in the original GAB model effort made to accommodate temperature variation results to a model that is complex to use in one hand and at the other hand its activity range is reduced for some food products (Jayas and Mazza, 1993; Menkov, 2000).

The objective of this research is to develop a model of water sorption isotherms for Corn flour using Neuro-fuzzy approach. The model will accommodate temperature and should be able to predict the equilibrium moisture content of the data at given $a_{\rm w}$ and temperature. The performance of the Neuro-fuzzy model will be then compared to the empirical GAB model. Further the two models will be used to predict shelf-life of the product. However, in our present research, we are incorporating water activity and temperature data into a single model. Comparison will then be made between GAB model and our proposed model to see which one give better prediction.

Neuro-fuzzy methodology: In the last decade, neuro-fuzzy systems found a wide success due to their hybrid nature that allows the acquisition of accurate knowledge from numerical data via neural learning, with the ability of representing it in a fuzzy rule-based structure so as to be classified as human readable. The first study that used both NN and fuzzy as keywords was written by the Lee brothers in 1974. They proposed a neuron model of multiinputs and multi-outputs (Lee and Lee, 1974, 1975). They generalized the McCulloch and Pitts neuron model whose output characteristic is a binary step function to deal with intermediate values. Neuro concepts have found wide application in many food and agricultural applications such as psychometrics (Sreekanth et al., 1998), drying (Huang and Mujumdar, 1993), thermal processing (Sablani et al., 1995) rheology and sensory science (Park et al., 1994).

Neuro-fuzzy systems approximate non-linearity by combining membership functions, sigmoidal functions, or other base functions. The neuro-fuzzy approach is based on a multistep strategy: once the feature selection process has been performed, the fuzzy inference model is automatically produced starting from input/output data, then it is subsequently modified to improve its accuracy. Before the application of the proposed method, the fuzzy model has to be initialied. This is based on the expert's knowledge or based on measured input-output data. The Least-Squares Error method (LSE) can efficiently be used

to find the initial consequent singleton fuzzy sets that minimize the Mean Square output Error (MSE), while the antecedent membership functions are equidistantly distributed over the input universes of discourse. A supervised learning process is then applied for the consequent enhancement of the fuzzy rule base:

$$\begin{array}{lll} R_k: \text{IF } x_1 \ A^k \ \text{AND....AND} \ x_n \ \text{is} \\ A^k_n \ \text{THEN y is} b_k \ k=1,...K \end{array} \tag{1}$$

Where, $x = (x_1, \dots x_n)$ are the input variables and y is the output variable, A_i^k are fuzzy sets defined on the input variables and b_k is a fuzzy singleton defined on the output variable (the example above refers to a generic kth rule). Fuzzy sets A_i^k are defined by Gaussian membership functions:

$$\mu_{ik}(x_i) = \exp(-\frac{(x_i - c_{ik})^2}{\sigma_{ik}^2})$$
 (2)

Where, c_{ik} σ_{ik} are the center and the width of the Gaussian function. When an input vector x is presented to the prediction model, the activation.

Strength of the k rule is firstly evaluated as:

$$\mu_k(x) = \prod_{i=1}^n \mu_{ik}(x_i), k = 1,..., K$$
 (3)

Then the output y is obtained by:

$$y = \frac{\sum_{k=1}^{K} \mu_{k}(x)b_{k}}{\sum_{k}^{K} \mu_{k}(x)}$$
 (4)

The described inference system is translated into a particular neural network, the neuro-fuzzy network, which reflects in its topology the structure of the fuzzy rule base. In particular, the neuro-fuzzy network consists of 4 layers performing:

- The introduction of the input values.
- The evaluation of the Gaussian membership functions.
- The computation of the fulfillment degree for each rule.
- The production of the final output.

The described neuro-fuzzy network is depicted in Fig. 1.

The tuning of the parameters used in fuzzy inference systems will be achieved by using numerical optimization procedure. The procedure is based on Gradient-Descent (GD) adaptation method which permits accurate learning of all parameters of the fuzzy model. The GD adaptation

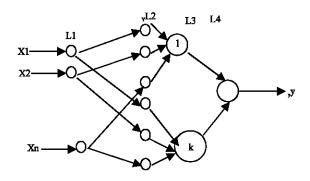


Fig. 1: The neuro-fuzzy network

rules are probably the best known supervised learning rules, producing a technique ideally suited to on-line instantaneous learning (Guely and Siarry, 1993; Jang, 1993). It is simple, it has low memory requirements and low computational cost, therefore it allows to built real-time learning algorithms. The GD method seeks to decrease the value of the quadratic objective function based on the instantaneous model output error:

$$E(k) = \frac{1}{2} (y_{m}(k) - y(k))^{2}$$
 (5)

Where y(k) is the reference for the modelled output $y_m(k)$. The parameter set, $\theta(k)$, of the fuzzy model is changed via the following iterative learning rule:

$$\theta(k+1) = \theta(k) + \Delta\theta(k) - \alpha \cdot \frac{\delta E(k)}{\delta\theta(k)}$$
 (6)

Where, α is learning parameter, which controls how much the parameters are altered at each iteration.

MATERIALS AND METHODS

Corn flour used was supplied by Smithklime Beecham Pharmaceutical Nigeria Plc. The powdery form of the corn flour starch has compositions of (0.38% protein, 0.11% ash, 0.11% fat). Prior to the beginning of the experiment, the initial moisture content was determined. A total of 3-4 g of the powder was added to 3 weighed aluminum dishes. The samples were oven dried between 95-100°C for 6 h to constant mass.

The samples were re-weighed after cooling in desiccators. Initial moisture content was estimated using the following equation

$$MC_{i} = \frac{M_{i} - M_{f}}{M_{e}} \tag{7}$$

Where:

Mci = Initial moisture content (dry basis) %.

Mi = Initial mass of sample (g).

Mf = Final mass of product after drying (g).

The equilibrium moisture contents of corn starch determined by gravimetric technique recommended by the COST 90 project (Wolf et al., 1985). In this technique, the weight will be monitored discontinuously within a standard static system of thermally stabilized desiccators. The entire sample to be used was first bone dried gravimetrically in a convectional oven at 105°C for 8-10 h (AOAC, 1980). 2.5±0.001 g sample of the bone dry powder was placed in an aluminium dish and kept at upper compartment of Glass desiccators while different chemical salt solutions were placed at the lower compartment of the desiccators in order to establish environment with different water activity. The saturated salt solution method suggested by Iglesias and Chirifie (1982) were used to give storage environment with different water activity. The salt solutions used and corresponding relative humidity at different temperatures are given in Table 1 (Young, 1967; Greenspan, 1977; Palipane and Driscoll, 1992). Each experiment was carried out in triplicate. The prepared desiccators were then placed in temperature controlled cabinets maintained at 20, 30, 40 and 50±1°C. Sample weight was taken at 24 h interval; this was allowed to continue for a period of 12 days until constant weight was obtained (±0.001 g). The total time required for removal, weighing and replacing the samples in the desiccators was not more than 30 s. This is to minimize the degree of atmospheric moisture adsorption during weighing.

To obtain the equilibrium moisture content of the product corresponding to different water activity, the following equation was made use of

$$EMC = \frac{M_e}{M_i} (IMC + 1) - 1 \tag{8}$$

Where:

EMC = Equilibrium Moisture Content of the product (dry basis) (g).

Mi = Initial mass of the sample (g).

Me = Final mass of the sample at equilibrium (g).

Determination of critical moisture content: The quality of the flour as hygroscopic materials is strongly affected by water activity (Menkov and Durakora). Microbiological and sensory properties of wheat flour and corn flour were studied at several temperature and moisture levels by Abdullah *et al.* (2000). They carried out investigation on

Table 1: Different salt solutions and their corresponding water activity for

	Water acitivity (%)				
Salt solutions					
Temperature (°C)	20	30	40	50	
Lithium chloride	11.3	11.3	11.2	11.1	
Potassium acetate	23.1	21.6	20.8	20.4	
Magnesium chloride	33.1	32.4	31.4	30.5	
Potassium carbonate	43.2	43.2	40.0	38.5	
Magnesium nitrate	54.4	51.4	48.4	45.4	
Potassium iodide	69.9	67.9	66.1	64.5	
Sodium chloride	75.5	75.1	74.7	74.7	
Ammonium sulphate	81.3	80.6	79.9	79.2	
Potassium chloride	86.7	84.3	82.3	82.0	
$H_2SO_4(sol.)$	98.0	98.1	98.3	98.5	

the fungal spoilage of rice flour and wheat flour and recommended that starch based food should be stored at water activity around 0.65. Also, in the works of Nasir et al. (2003), it was observed that the strong effect of moisture on the shelf life of wheat flour has serious effect on crude protein, crude fat, mould, growth and insect-infestation. Critical moisture content is an important factor and its determination will make it possible to calculate the shelf life moisture of sensitive food product. Based on the recommendation of Abdullah et al. (2000) that suitable condition for storing starch based food is at water activity of 0.65. Critical moisture content of starch food can be determined at this value. This can be read out from the moisture sorption isotherm curve of a specific product.

Determination of shelf life: Shelf life of a food product is the time frame for which it deteriorates to an unacceptable level under specific storage, processing and packaging conditions. NAFDAC and other international food control agencies like FDA and USDA mandate food manufacturers to indicate clearly the expiration date of their products on the containers or packages to know its shelf life. Among the factors that affect the shelf life of food product are product composition, storage condition, processing condition, distribution condition packaging of initial quality. Storage conditions and packaging are very vital before it finally get to consumers. For the purpose of this research the two strong factors considered for shelf life determination are packaging and storage conditions. LDPE film with thickness mm and average water vapour permeability coefficient of 0.1682 at a room temperature was used as packaging material (Xiong, 2002). The shelf-life is determined when the moisture content reaches critical values. To obtain permeability coefficient at any temperature, the following function which is related by Arrhenius's equation is used:

$$\operatorname{In}\left(\frac{P_{2}}{P_{1}}\right) = \frac{-E_{a}}{R}\left(\frac{1}{T_{2}} - \frac{1}{T_{1}}\right) \tag{9}$$

Where:

P = Permeability coefficient, kg.m/m²/sec/Pa

Ea = Activation energy, kg mol⁻¹.

 $R = Gas constant, 8.314 J mol k^{-1}$

T = Temperature in K

To find P at different temperatures, we need to know the activation energy Ea, (Xiong, 2002) gives this to be 22.859 KJ mol⁻¹.

To determine shelf life experimentally, about 15 g of sample was kept inside 10×10 mm LDPE bag (thickness 1.25 mm). The bag containing the product was measured at regular interval until critical moisture content was reached. The critical moisture content was determined from sorption isotherm at 0.65 water activity. Potassium acetate solution at different temperatures to give different water activities was used to create different storage conditions. Shelf life can be analytically determined by using this equation (Xiong, 2002):

$$\frac{d}{d_i}P = P.\frac{A}{L}(P_o - P_i) \tag{10}$$

Where.

P = Partial pressure of water vapour, Pa.

P₀ = Partial pressure of water vapour of storage environment. Pa.

 P_{i} = Partial pressure of water vapour inside the packaging, Pa.

t = Storage time, sec.

P = Permeability coefficient of polymeric material, Kg. m/m²/sec/Pa.

Expressing Eq. 3 in term of water activity will give

$$dt = \frac{Lw_{D}}{P_{s}.AP} \int_{f(a_{w})}^{f(a_{w})} \frac{f(a_{w})}{a_{sw0} - a_{wo}} da_{w}$$
 (11)

Where,

P_s = Saturated water vapour pressure at a specific temperature, mmHg.

a_{wo} = Water activity (relative humidity) of storage condition.

 a_{w1} = Initial water activity.

 a_{w2} = Critical water activity.

 $f(a_w)$ = is function which represents either GAB model or Neuro-fuzzy model.

 $f'(a_w) = Derivative of f(a_w)$.

RESULTS AND DISCUSSION

Experimental moisture sorption data for corn flour at different temperatures are shown in Fig. 2. The equilibrium moisture content at each $a_{\rm w}$ is the average value of 3

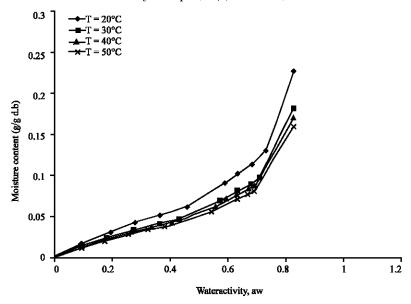


Fig. 2: Experimental moisture sorption curves for corn flour at different temperatures

replicates. Apart from the obvious trend, it was observed that the isotherm curves for temperatures of 30, 40 and 50°C, meet at water activity a_w, of 0.84 which correspond to a moisture content of 0.11(g g⁻¹ d.b). At above water activity of 0.8, slopes of the isotherm curves for all temperatures become steeper, this implies that the flour becomes less hygroscopic as temperature is increasing. This is due to the fact that at high temperature, kinetic energy of the water molecules increases which corresponds to an increase in the absorbance capacity of the material for a given water activity (Pradyuman and Mishra, 2005) with some of the water molecules leaving the active centres of the adsorbent.

The GAB model to be correlated is given as:

$$\frac{\mathbf{a}_{w}}{\mathbf{w}} = \frac{1 - 2.\mathbf{k}_{w} + C.\mathbf{k}.\mathbf{a}_{w} + \mathbf{k}^{2}.\mathbf{a}_{w}^{2} - C.\mathbf{k}^{2}.\mathbf{a}_{w}^{2}}{C.\mathbf{k}_{w}}$$
(12)

Where, $a_{\rm w}$ denotes the water activity, w is the water content adsorbed on 1 g of sample, $w_{\rm m}$ is the water content adsorbed in the monolayer and C and k are the sorption constants.

The parameters for the GAB sorption model at different temperatures for corn flour are shown in Table 1, together with the Mean Relative Deviation modules (MRD). A non-linear least square regression using *minerr* function implemented in Mathcad 2000 was used to fit the model to the experimental data. The goodness-of-fit of the model was determined by using mean relative percentage deviation modulus. A modulus value not greater than 10% is an indication of a good fit for most

practical analysis (Lomauro *et al.*, 1985). Its value is in the range 5.284-5.9347% and decreases with temperature. Also shown in the Table 2, is Correlation coefficient which increases with temperature.

Table 1 shows the fitting constant coefficients of the GAB model, the correlation coefficients and the mean. The coefficient for GAB model along with correlation coefficient and mean relative percentage deviation modulus are given in Table 2.

The GAB parameters are useful in telling the structural stability and thermodynamic properties of the food items. W, the monolayer moisture content defines the physical and chemical stability of foods. It has effect on the lipid oxidation, enzymes activity, non-enzymatic browning, flavour preservation and product structure (Menkov, 2001). As shown in the Table 2, W₀ is decreasing with increase in temperature. These values are in the range of acceptability because the maximum monolayer moisture content should not be more than 10% (d.b) for food products (Labuza et al., 1985). As pointed out by Geankoplis (1993) the observed decrease in W_o with increasing temperature is due to decrease in number of active sites for water binding which may be caused by change in physical/or chemical structures in the food products as a result of change in temperature. The trends are in line for highly carbohydrate foods as reported by (Labuza, 1968).

Another parameter in the GAB model which proves useful in the determination of heat of sorption Qs parameter is denoted by C. It is related to Qs by the following equation.

Table 2: Estimated values of coefficients and mean relative deviation module obtained for GAB sorption model applied to experimental adsorption

T(°C)	\mathbf{w}_{m}	C	K	Qs(kJ mol ⁻¹)	R2	MRD (%)
20	0.049	8.991	0.855	5.349	0.974	5.935
30	0.039	8.972	0.857	5.528	0.983	5.821
40	0.037	8.798	0.858	5.575	0.985	5.497
50	0.035	8.534	0.861	5.757	0.987	5.284

Table 3: Correlation coefficients and mean relative percentage deviation

Temperature (°C)	Correlation coefficient, R2	MRD(%)	
20	0.9952	2.353	
30	0.9975	2.136	
40	0.9987	2.124	
50	0.9991	2.015	

$$Qs = RT \ln C \tag{13}$$

Where,

 $R = 8.314 \,\mathrm{J} \,\mathrm{mol}^{-1}$

T = Absolute temperature.

C = Parameter constant for the GAB isotherm equation.

 Q_s is a thermodynamic property which measures the quantity of free water in the foodstuff. As observed from the Table 3, the Q_s increases with increasing temperature. This observation is in contrast to the ones reported for corn meal and fish flour by Labuza *et al.* (1985). However, the values indicated that sorption type corresponded to physical adsorption because the values were lower than 41.67 kJ mol⁻¹ (Lomauro *et al.*, 1985).

In this research, we attempted to use neuro-fuzzy technique to describe isotherm behaviour of corn flour. We have water activity and temperature as the inputs and the equilibrium moisture content of the foodstuff as output to train the model. Neuro-fuzzy technique provides a procedure to learn information about a data set and to allow computation of the membership function parameter that best allow the associated fuzzy inference system to track the given input/output data. Six gaussian membership functions for fuzzification of each input data set and 6 linear membership function for output defuzzification were used (Takagi-sugeno-Kang method). The tuning of the membership parameters used in fuzzy inference systems was achieved by using numerical optimization procedure based on gradient-descent.

Using this algorithm in conjunction with a least squares method allowed the fuzzy system to learn from the data. The computation of these data was facilitated by a gradient vector. These parameters were adjusted until minimum error was obtained. Presented in Table 4 are correlation coefficients and mean relative percentage deviation modulus for different temperatures. The Mean Square Error (MSE) for the training was obtained as 0.0000456. The R² values obtained for neuro-fuzzy model are greater than that for GAB model and MRD values are

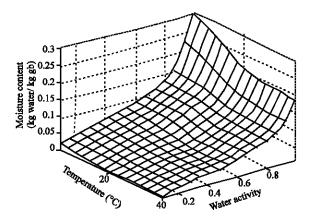


Fig. 3: Surface plot of neuro-fuzzy isotherm model

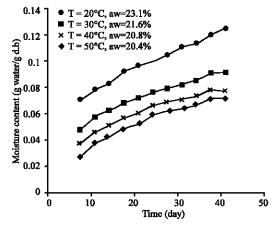


Fig.4: Change in moisture content with time

smaller, these indicate that neuro-fuzzy is more accurate in describing the isotherm sorption behaviours of corn flour. The surface plot of neuro-fuzzy isotherm prediction is shown in Fig. 3. The surface plot is able to show that EMC is decreasing with increase in temperature and increasing with decrease in water activity.

The changes in moisture content with time for given temperature and water activity are displayed in Fig. 4. As observed from the Fig. 4, the moisture content is increasing with number of days but decreasing with increase in temperature and decrease in water activity.

Based on the work of Abdullahi et al. (2000) it has been recommended that the starch based food must be stored at water activity levels not higher than 0.65. The value of moisture content corresponding to this value was taken as critical moisture content the sample. This varied

Table 4: Critical moisture content, experimental self life and predicted self life according to GAB model neuro-fuzzy model

		Critical moisture	Experimental Shelf	Shelf life according to	Shelf life according to
Temperature(°C)	Water activity (%)	content (g water/g d.b)	life(days)	GAB model(days)	neuro-fuzzy model(days)
20	23.1	0.091	14.417	10.346	14.50
30	21.6	0.073	21.277	15.625	21.975
40	20.8	0.068	28.956	22.165	29.051
50	20.4	0.063	35.262	29.245	36.531

for different storage temperature. The critical moisture content at different temperature as obtained from the isotherm curves are given in Table 4. The table indicate that the critical moisture content is decreasing with increase in temperature. Referring to the same table, the shelf life is increasing with increase in temperature. This shows that with decrease in critical moisture content there is increase in shelf life of corn flour.

The shelf-life of corn flour was computed from Eq.11 using adaptive Simpson's rule. Where f'(x) in Eq.11 represents isotherm model, which can be either GAB or neuro-fuzzy models. From Table 4, predicted shelf life for GAB model at different temperatures are far below experimental values, while neuro fuzzy predictions are very close to experimental results. This implies that by using fuzzy model to predict the shelf life of corn flower, the result obtained compares very well with experimental values.

CONCLUSION

A new sorption model of corn flour based on neuro-fuzzy approach was developed. Analysis showed that neuro-fuzzy model not only accommodated temperature and water activity parameter, but also is more accurate than the GAB model. The mean relative deviation of sorption neuro-fuzzy model were 2.353, 2.136, 2.124 and 2.015, respectively for 20, 30, 40 and 50°C. The GAB model is considered to be the most versatile sorption model available in the literature; however the mean relative deviation of GAB model were 5.935, 5.821, 5.497 and 5.284 at 20, 30, 40 and 50°C, respectively. Similar result was observed in the prediction of shelf-life. We conclude that neuro-fuzzy model is more versatile than the widely accepted GAB model to describe sorption isotherm of corn flour.

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