INVESTIGATION OF MOISTURE SORPTION BEHAVIOR OF AN INDIAN SWEET 'SON-PAPDI

Suni Bajpai[1] *and Pradeep Tiwari*[2]

Address(es): Suni Bajpai
[1] Polymer Research Laboratory ,Govt. Model Science College Jabalpur (M.P.) – 482001, India, Phone : + 91 9993220651.
[2] Vindhya Institute of Technology and Science, Jabalpur (M.P.) - 482001, India.

Corresponding author: sunil.mnlbpi@gmail.com

ABSTRACT

Water vapors sorption isotherms of *Sonpapdi,* an Indian traditional sweet, were studied over a selected temperature range of 25 to 37°C using gravimetric method. Various conventional isotherm models, namely GAB, Oswin, Henderson, Caurie and Halsey, were applied on equilibrium moisture uptake data and it was found that GAB isotherm model described adequately the moisture absorption behavior of *Sonpapdi*.The monolayer moisture content M_0 and GAB constant C showed negative temperature dependence for the increase in temperature from 25 to 30°C. For the moisture uptake range of 0.02 to 0.07 kg kg^{-1}db, isosteric heat of sorption, q_{st} and differential entropy of sorption, S_d were evaluated. Finally the uptake data was used to determine isokinetic temperature T_β (405K) and harmonic mean temperature, T_{hm}. (392K). As $T_\beta > T_{hm}$, the overall sorption was found to be enthalpy driven. Properties of sorbed water were also determined.

Keywords: Sonpapdi, isotherm, isosteric heat of sorption

INTRODUCTION

Sonpapdi, a traditional Indian sweet, is very popular in asian countries due to its sweetness, aroma and crispiness. The basic ingredients of this product are gram flour (20%), semolina (a by-product of wheat flour during milling) (20%), sugar (35%), edible oil (20%) and a trace amount of coconut powder, dry fruits. The ingradients gram flour, semolina and edible oil are hand mixed thoroughly and the resulting dought is put in the electric oven at nearly 80°C for baking. A slurry of sugar is prepared in hot water (60-70°C) and is thoroughly mixed with the baked product and then is placed on big trays and allowed to cool to set at room temperature. Finally, the product is cut into small rectangular pieces and sold in the market (See Fig. 1).

Figure 1 Indian sweet dish "Son-Papdi"

Because of high content of proteins and sugar, this product is quite hydrophilic in nature and after moisture absorption it becomes hard and there is great detoriation in its test as well. This, in fact, limits its production and consumption in rainy season. Although, there is no official information available, but according to local manufactures, its consumption is reduced by almost forty to fifty percent in this season. In this respect, this product resembles fairly with *Basbusa,* a traditional Arabian confectionary (**Ahmad *et al.,* 2004**) and Indian sweet *Sandesh* (**Sahu *et al.,* 2008**). It has also been observed that prolonged

storage under ambient conditions adversely affects consumers acceptability due to microbial changes in the flavor profile such as taste, smell, texture and color.

Relative humidity or water activity (a_w) plays an important role in food product development, storage and packaging. It is well documented that water activity has significant role in product safety and stability with respect to microbial growth, chemical/ biochemical reaction rates and physical properties (**Fontana, 2000**). The quantitative relationships between water activity and moisture content of a food at constant temperature are described by moisture sorption isotherms (MSI). They give an insight into the moisture-binding characteristic of a food. Therefore, knowledge of the moisture sorption behavior of a food is important for prediction of its quality, stability, and shelf-life during its packaging and storage (**Mathlouthi *et al.,* 2001; Cervenka *et al.,* 2008**).

Moisture sorption characteristics of various foods have been reviewed by **Al-Muhtaseb *et al.* (2002)**. Moisture sorption isotherms of various types of Indian sweets including *Coconut Burfi* (**Gupta *et al.,* 2010**), *Khoa* (**Singh *et al.,* 2008**), *Sandesh* (**Sahu *et al.,* 2001**), *Kheer* (**Kumar *et al.,* 2005**), *Dudh Churpti* (**Hossain *et al.,* 2002**), *Chhana Poda* (**Rao *et al.,* 2006**) *Milk Barfi* (**Ramkrishnan *et al.,* 2005**), etc. have already been reported. However, studies on water sorption isotherms of *Sonpapdi* have never been reported. The present work describes moisture sorption characterstics of *Sonpapdi* at various temperatures to evaluate the applicability of various existing models for their ability to fit the sorption data and to calculate the properties of sorbed water.

MATERIAL AND METHODS

Materials

Sonpapdi samples were purchased from a local departmental store and used for the study. Different salts employed for preparation of saturated salt solutions to create desired relative humidity (RH) or water activity (a_w), were purchased from E.Merck India Ltd. and were analytical grade. The double distilled water was used throughout the investigations.

Equilibrium moisture sorption studies

For moisture sorption studies, the following salt slurries were used :lithium chloride, potassium acetate, magnesium chloride, potassium carbonate, magnesium nitrate, cobalt chloride, stroncium chloride, sodium chloride, ammonium sulphate, potassium chloride and barium chloride. Pre-weighed samples (1.0g each, in triplicate) were placed in small Petridishes and put in glass desiccators containing different saturated salt solutions, thus providing a constant

relative humidity environment ranging from 5 to 97 percent as described elsewhere (**Arabhosseini et al., 2010**). The samples in the desiccators were weighed periodically until constant weight was obtained. The total time for removal, weighing and replacing the samples was less than 15s. This minimized atmospheric moisture sorption during weighing. Equilibrium was considered to have reached when the difference between the two successive measurements was less than 0.001 g. The moisture content was determined by drying the *Sonpapdi* samples in an vacuum oven (<100 mm Hg) at $60 \pm 2^{o}C$ until constant weight (**Fabra et al., 2009**). The equilibrium moisture contents (EMC) were calculated on a dry basis using following expression:

$$EMC = \frac{Hydrated\ mass - Dry\ mass}{Dry\ mass}\ kg\ kg^{-1} dry\ basis$$

$$......(1)$$

Modeling of isotherm models

The equilibrium moisture uptake data of *Sonpapdi* samples, at three temperatures, namely 25, 30 and 37°C, were fitted to various isotherm models (Table 1).

Table 1 Sorption isotherm models used in this study

Name of model	Equation	Reference
GAB (Guggenheim-Anderson-de Boer)	$M = \dfrac{M_0 C k.a_w}{(1 - ka_w)(1 - ka_w + cka_w)}$	(**Anderson, 1946**)
Halsey (Linearized)	$a_w = \exp\left(\dfrac{-A1}{M^{A2}}\right)$ $In\ M = InA_1 + A_2\ In\ (-In\ a_w)$	(**Halsey, 1948**)
Oswin (linearized)	$M = A\left(\dfrac{a_w}{1 - a_w}\right)^{B}$ $In\ M = In\ A + B\ In\ \dfrac{a_w}{1 - a_w}$	(**Oswin, 1946**)
Henderson (Linearized)	$Log\ [-In\ (1 - a_w)] = n\ log\ M_c + log\ K$	(**Henderson, 1952**)
Caurie	$\ln \dfrac{1}{m} = -\ln(CM_0) + \dfrac{2C}{M_0}\ln\dfrac{1 - a_w}{a_w}$	(**Caurie, 1970**)

Abbreviations : M = equilibrium moisture content (kg kg⁻¹ dry solid), a_w = water activity, Mo = monolayer moisture content (kg kg⁻¹ dry solid), C and K = GAB constant, A_1 and A_2 = Halsey constants, A and B = Oswin constants, n and k are Henderson constants, C is caurie constant.

The parameters of the GAB sorption model were calculated using non-linear regression analysis whereas other models were applied in linearized forms to evaluate associated parameters. Two criteria that were mean relative deviation modulus (P) and the standard error of estimate (SE) were used to evaluate the well fitting of sorption models to the experimental data.

$$P = \frac{100}{N} \sum_{i=1}^{N} \frac{|M_{ex} - M_{pr}|}{M_{ex}} \quad ....(2)$$

$$SE = \sqrt{\frac{\sum_{i=1}^{N} (M_{ex} - M_{pr})^2}{N - n}} \quad ....(3)$$

where M_{ex} and M_{pr} were the experimental and predicted moisture content values, respectively; N and n were the number of observations and the number of constants in each model respectively. A model is considered acceptable if it has a P value less than 10 percent (**Lomauro et al., 1985**). The model with lowest SE and P and highest regression coefficient (r^2) was supposed to be the most fitted one.

In the GAB isotherm model, M_o is the monolayer moisture content, C is a constant related to the first layer heat of sorption and K is a factor related to the heat of sorption of the multilayer. The effect of temperature on GAB parameters C and K can be expressed with an Arrhenius form equation (**Goula et al., 2009**).

$$C = C_0 \exp\left[\frac{\Delta H_c}{RT}\right] ....(4)$$

$$K = K_0 \exp\left[\frac{\Delta H_k}{RT}\right] ....(5)$$

where C_o and K_o are pre-exponential factors; ΔH_c is the difference in enthalpies between monolayer and multilayer sorption in kJmol⁻¹; ΔH_k is the difference between the heat of condensation of water and the heat of sorption of multi-layer in kJ mol⁻¹; R is the universal gas constant in kJ mol⁻¹ K⁻¹; and T is the temperature in K.

Using the GAB model, monolayer moisture content M_o was calculated. Once the monolayer moisture content was realized, the solid surface area of the samples could be determined (**Cassini et al., 2006**).

$$S = \frac{M_o N_A Am}{M_{H_2O}} ....(6)$$

where S is the solid surface area (m²g⁻¹ dry solids); M_o is the monolayer moisture content in kg kg⁻¹ dry basis; M_{H2O} is the molecular weight of water (18 g mol⁻¹); N_A is Avogadro's number (6.02x10²³ molecules mol⁻¹); and Am is the area of water molecule (1.06 x 10⁻¹⁹ m²).

The number of adsorbed monolayer was obtained by the formula

$$S = \frac{2}{N} ....(7)$$

where S is cauries slope. Density of bound water is represented by C in Cauries equation and percent bound water or non freezable water is the product of monolayer moisture content M_0 and number of adsorbed monolayers N (**Jayendra Kumar et al., 2005**)

Isosteric heat of sorption and entropy of sorption

The net isosteric heat of sorption (q_{st}) is the amount of energy above the heat of vaporization of water associated with the sorption process and is calculated from the experimental data using Clausius-Clapeyron equation :

$$q_{st} = -R\left[\frac{d \ln a_w}{d(1/T)}\right]_x ....(8)$$

The isosteric heat of sorption is calculated from equation (8) by plotting the sorption isotherm as the natural logarithm of water activity versus 1/T for specific moisture content and determining the slope, which equals – q_{st}/R. The application of method requires the measurement of sorption isotherms at three or more temperatures.

Differential entropy of sorption (S_d) is proportional to number of available sorption sites at a specific energy level. For a thermodynamic system, q_{st} and S_d are related as–

$$\ln a_w = -\frac{q_{st}}{RT} + \frac{S_d}{R} ....(9)$$

The intercept of linear plot of In a_w versus I/T yields S_d (**Aguerre et al., 1986**). In the present study, GAB isotherms, obtained at 25, 30 and 37°C were used to determine q_{st} and S_d.

Enthalpy – entropy compensation theory

The existence of a linear relationship between net isosteric heat of sorption (or enthalpy of sorption) and differential entropy of sorption has been confirmed by several authors (**Thys et al., 2010** ; **Fasina et al., 2006**). The compensation theory proposes a linear relationship between q_{st} and S_d as shown below

$$q_{st} = T_\beta. S_d + \Delta G_p \quad ... (10)$$

where T_β (K) is the isokinetic temperature and ΔG_β (Jmol⁻¹) is the free energy at T_β. A statistical analysis test (**Krug et al., 1976**) can be used to corroborate the

compensation theory by comparing T_β with the harmonic mean temperature T_{hm}, defined as

$$T_{hm} = \frac{n}{\Sigma(1/T)} \quad ....(11)$$

where n is total number of isotherms used.

RESULTS AND DISCUSSION

The equilibrium moisture sorption data for *Sonpapdi* at 25, 30 and 37°C are shown in Fig. 2.
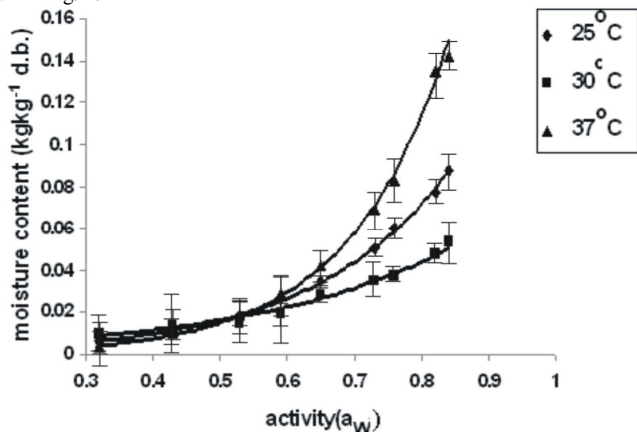
Figure 2 Moisture uptake as a function of water uptake at different temperatures

The equilibrium moisture content (EMC) at each water activity represents the mean value of three replications. The sorption isotherms demonstrate an increase in EMC with increasing water activity at a constant temperature. These curves represent sigmoidal type-II isotherms according to BET classification (**Brunauer *et al.*, 1938**) and are characterstic of amorphous materials rich in hydrophilic components (**Goula *et al.*, 2008**). A close look at Fig.2 reveals some interesting facts. The isotherms, obtained at 25 and 37°C, show similar trends, i.e. they show small increase in EMC in the region of low and intermediate water activity range whereas they exhibit appreciable rise in moisture content at high water activity levels, the so-called capillary condensation region. Moreover, a negative temperature dependence is observed, i.e. for a given water activity the equilibrium moisture content is more at 25°C as compared to that at 30°C. However, the isotherm, obtained at 37°C, shows almost different behavior. There is cross-over observed at 37°C. The observed behavior of isotherms at different temperatures may be explained as follows : In *Sonpapdi*, apart from sugar, the product contains proteins due to presence of gram flour and wheat flour as one of the major constituents (almost 20%). Since proteins are preferred sorption sites at low water activity (**Caurie *et al.*, 1970**), the sorption in low water activity range may be attributable to polar groups that are present along the macromolecular chains (Singh et al.2001). Later on, water sorption spreads to peptide linkage and then leads to multilayer formation at higher relative humidities. The enhanced moisture uptake at 25°C, as compared to that at 30°C, could be attributed to the fact that increased temperature might reduce number of active sites for water binding as a result of physical/chemical changes in the product (**Goula *et al.*, 2008**). However, a more convincing argument is that at increased temperature water molecules get activated to higher energy levels and break away from water binding sites of substrate (**Palipane, 1992**). Such negative temperature effect on equilibrium water content have been observed in many foods of high sugar content (**Ertugay *et al.*, 2000; Kumar *et al.*, 2005**). The observed cross-over of isotherm at 37°C occurred at a water activity value of 0.52. This crossover behavior has also been reported by several investigators for high sugar foods (**Hassan, 2002; Sharma *et al.*, 2009**). It has been suggested that this inversion of the usual dependence of moisture sorption isotherms may be due to endothermic dissolution of the sugar at higher water activities (**Saravacos *et al.*, 1986**). According to an observation(**Jayendra Kumar *et al.*, 2005**), polymers (protein and starch) sorb more water than sugar at low water activities while the soluble component sugar sorbs more water at higher water activity thereby overcoming the negative temperature effect due to an increase in solubility of sugars in water.

Fitting of isotherm models to moisture uptake data

The various parameters, associated with different isotherm models, namely GAB, Oswin, Halsey,Caurie and Handerson are given along with the standard errors of estimate (SE), the mean relative percentage deviation modules (P) and the coefficient of determination (r^2)(Table 2).

Table 2 Various isotherm parameters evaluated

Model	Constants	Temperature (°C)		
		25	30	37
GAB	Mo	0.0158	0.0109	0.0042
	C	52.658	29.885	18.143
	K	0.8981	0.8957	1.0827
	SE	0.5213	0.4187	0.3217
	P	3.4812	4.6321	5.1280
	r^2	0.9892	0.9791	0.9921
Halsey	A1	0.0213	0.0203	0.2116
	A2	-0.7222	-0.4180	-0.2339
	SE	1.6857	0.9928	2.3168
	P	8.6219	9.3890	11.6321
	r^2	0.9146	0.9348	0.8996
Oswin	A	0.0328	0.0190	0.0191
	B	0.3771	0.4737	0.6680
	SE	3.6110	2.1890	1.7348
	P	7.4365	6.6129	5.2318
	r^2	0.9652	0.9543	0.9012
Henderson	N	0.9944	1.0088	0.5927
	K	4.0686	4.9516	2.6332
	SE	0.8826	1.2595	1.6304
	P	10.6239	8.1214	9.2357
	r^2	0.9176	0.8996	0.8268
Cauri	M_o	0.2999	0.3675	0.1750
	C	0.0926	0.0531	0.0590
	SE	2.3216	3.1886	1.9879
	P	9.2368	7.4680	8.3216
	r^2	0.9218	0.9346	0.9421

The goodness of the fit was provided for the higher values of r^2 and lowest values of SE and P. It can clearly be seen in Table-II that GAB isotherm model provides best fit to the experimental data over almost the whole range of water activity and temperature for *Sonpapdi*. It has been reported that GAB isotherm model has theoretical background whereas other models are empirical or semi-empirical in nature (**Yao-Element *et al.*, 2008**). Therefore, it has been adopted by the European Project cost 90 on physical properties of foods (**Farahnaky *et al.*, 2009**). The additional advantage of GAB model is that it describes the sorption behavior of nearly all foods from 0 to 0.9 a_w (**Rizvi, 2005**).

The monolayer moisture content M_o is recognized as the moisture content affording the longest time period with minimum quality loss at a given temperature. Below this value, the rates of deteriorative reactions are minimal. Therefore, at a given temperature the safest water activity level is that corresponding to M_o or lower (**Goula *et al.*, 2008**). This is because water is strongly bound to the food below M_o and is not involved in deteriorative reactions. In the present work, value of M_o (kg kg^{-1} solid dry basis) are found to be 0.0158, 0.01092 and 0.00425 at 25, 30 and 37°C respectively. The observed decrease in M_o values with temperature could be due to enhanced kinetic energy of water vapor molecules which discourages their sorption onto active sites of material. This is a commonly observed phenomenon and reported by a number of workers. The monolayer moisture contents, when transformed into percent basis, give values of 1.58, 1.09 and 0.42 at 25, 30 and 37°C respectively. These values can now be compared with monolayer moisture contents of some other Indian sweets as has been reported elsewhere (**Sahu, 2008**) in several reports, e.g. 5.89 to 5.20% for *Sandesh* in 20-30°C range of temperature; 5.56 to 4.83% for *Chhana Poda* in 5-35°C temperature range. 8.73 percent for *Peda* at 30°C; 3.06 to 2.91 percent for *Kheer* in the 25-40°C temperature range. A comparison of these values of M_o with those obtained for *Sonpapdi* clearly reveals that monolayer moisture contents for *Sonpapdi* are very low as compared to other sweets.. The M_0 values,displayed in Table 2, revel some significant information about the packaging and shelf life of this Sweet Dish. In the temperature range of 20 to 37°C,the M_0 value are found to be 0.0158 to 0.0042 kg/kg db. These values correspond to water activities level of nearly 0.37 to 0.32 . This indicates that sweet dish spoilage by bacterial contamination or other processes may start if the relative humidity level of environment during the packaging and storage is above 37 percent at the room temperature i.e. 25°C .As stated in the section Introduction , manufacture of this product is very common , frequent and is done by a large population as small scale industry all over the country, its production and storage in the rainy season (when humidity is almost above 60-70%) shall result in spoilage of this sweet dish and so it is advisable to avoid to prepare and store this when there is fairly higher humidity.. However ,if the product is packaged and stored at 37°C then about 32 percent humidity is the upper safest level. So in summers when there is fairly as higher temperature as 37°C or more, its preparation packaging and storage can be done as there is always very low humidity(never as high as 32 %).

The another GAB isotherm model constant C has been reported to depend upon temperature by Arrhenius type equation (**Gabas et al., 2007**):

$$C = C_G \exp(\Delta Hc / RT) \ldots\ldots\ldots\ldots(12)$$

where ΔH_c is the function of the heat of sorption of water; $\Delta H_c = H_m - H_n$ where H_m and H_n as the heats of sorption of the monolayer and multilayer of water respectively. Moreover, C_G is the adjusted constant for the temperature effect. In a previous work (**Blahovec, 2008**), it was shown that parameter C should fulfill the following conditions; For C≥2 the GAB equation gives a sigmodial shape curve with point of inflection (type II of **Brunauer's,(1943)** classification); however for 0<C<2, the isotherm is of the type of III only (isotherm without point of inflection).

The values of C, obtained at 25, 30 and 37⁰C, are not only grater than 2, but they also show negative temperature effect. The value of C decreases from 52.65 to 18.14 as the temperature is increased from 25 to 37⁰C.This is in theoretical agreement with the eq.(12) showing C as a inverse function of temperature. This may simply be attributed to the fact that the adsorbent - adsorbate interaction decreases with the increase in temperature thus resulting in decrease in value of C. However, it was reported that out of 30 different foods investigated , the parameter C for nearly 74% of them did not decrease with increase in temperature(**Iglesias and Chirife, 1982**) probably due to some irreversible changes associated with increase in temperature like enzymatic reactions and protein de – naturation.In the present work,as the values are greater than 2, they support the sigmoidal type II nature of isotherms.

The value of K provides a measure of the interactions between molecules in the multilayers with the adsorbent and tends to fall between the energy value of the molecule in the monolayer and that of bulk liquid. If K is equal to 1, the multilayer's have properties of bulk water (**Perez-Alonso et al., 2006**) and the sorption behavior could be modeled by the BET. It can be seen from the values of K, displayed in Table- II that at 37 ⁰C, the GAB constant K aquires the maximum value of unity (i.e.K = 1). This indicates that multilayer has bulk liquid properties .Almost similar type of results have also been reported elsewhere(**Yao Clement and Tano, 2008**).

Properties of Sorbed Water

The physical state of water adsorbed by foods actually determines the actual spoilage (**Van den Berg and Bruin, 1981**).It is therefore essential to generate information related to various aspects of bound water viz its density, its relation to surface area of adsorbent ,number of adsorbed monolayers etc. All the parameters describing the property of sorbed water are shown (Table 3) .

Table 3 Properties of sorbed water of *Sonpapdi* at different temperatures

Temperature (⁰C)	Monolayer moisture content (g/100g dry solid) M_0	Cauri slope S	No.of adsorbed monolayer N	Density of sorbed water C	Bound water %	Surface Area m^2g^{-1}
25	0.932	0.6178	3.23	0.0926	3.01	55.443
30	0.624	0.4792	4.17	0.0648	2.60	38.226
37	0.359	0.6749	2.96	0.0590	1.06	14.875

The data reveals that density of water, percent of bound or unfreezable water, monolayer moisture content and specific surface area of sorbent decrease with increase in temperature. Almost similar results have also been reported elsewhere (**Cervenka et al., 2008**) for *Gingerbread*, a traditional bakery product in Czech Reublic.With the temperature increase from 20 to 30⁰C, the density of sorbed water, surface area, number of adsorbed monolayers and monolayer moisture content decreased from 1.882 to 1.854, 78.73 to 70.55, 2.89 to 2.59 and 7.31 to 6.38 respectively. Similarly for Indian sweet dish *Sandesh*, these values were found to decrease from 2.205 to 1.999, 72.77 to 70.89, 2.67 to 2.60 and 5.892 to 5.205 respectively. (**Sahu and Jha, 2008**). The results, in the case of rice-based milk product *Kheer*. (**Jayendra Kumar et al., 2005**), the surface area and number of adsorbed monolayer showed an increasing trend when temperature was raised from 10 to 25⁰C, but an opposite trend was observed when the temperature was further raised to 40⁰C. Similarly for ready- to- use *Basundi mix* (**Sharma et al., 2009**), the density of sorbed or unfrozen water increased from 0.1522 to 0.2110 for temperature rise from 5 to 45⁰C, while other parameters showed a decreasing trend. In the present study, the observed decrease in surface area with temperature might be due to reduction in number of active sites due to physical and chemical changes induced by temperature (**Rizvi and Banado, 1983**). Here it is also to be noted that specific surface areas, as reported in Table-III, are in the range of 55 to 14 m^2g^{-1} for the temperature range of 25 to 37⁰C. These values appear to be a little lower as compared to values reported by other workers for the products like *Kheer* (79.11 to 83.27 m^2g^{-1}), *Sandesh* (70 to 72 m^2g^{-1}), *Basundi mix* (92 to 141 m^2g^{-1}), *Gingerbread* (70 to 78 m^2g^{-1}), etc. This clearly indicates that the product *Sonpapdi* may not be porous in nature.

Net isosteric heat of sorption (q_{st})

The net isosteric heat of sorption , also known as binding energy of sorption, may be considered as indicative of intermolecular attractive forces between the sorption sites and water vapor molecules.In order to determine the values of isosteric heat of sorption, values of water activity were evaluated graphically at 25,30 and 37⁰C ,for a definite range of moisture content of 0.02 to 0.07 kg kg⁻¹ solid basis.The activity values, thus obtainted, were plotted as ln a_w verses 1/T to obtain linear plots as shown in fig.3.The values of q_{st},obtained using slopes of linear plots were 67.44,14.34,13.39,13.25,and 11.71 kJ/kg for the moisture content of 0.02,0.03,0.04,0.05,0.06,kg kg⁻¹ dry solid respectively.
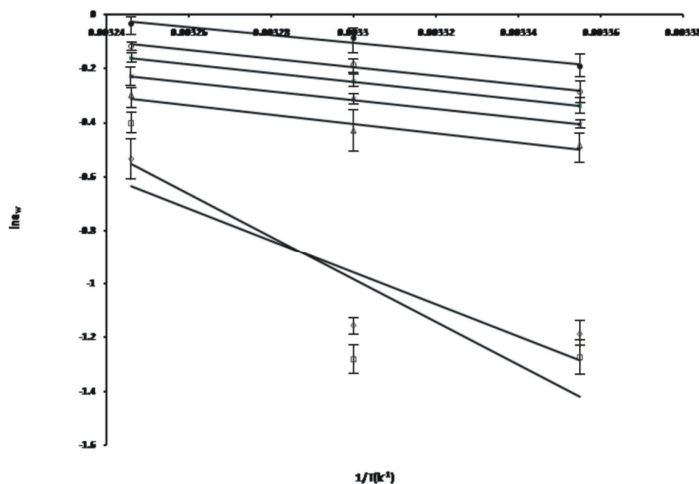

Figure 3 Plot between lna_w and 1/T for evaluation of q_{st}

The values ,thus obtained, are very low as compared to those reported others (**Jayendra Kumar et al., 2005**) for tradition Indian milk product *Kheer*(4100to700kJ/kg), by Sahu et al.(2008) for indian sweet *Sandesh* (6000to2500 kJ/kg). The observed low values of q_{st} in the present study could be attributed to weak interaction between vapor molecules and active site due to the presence of almost 20 percent of edible oil as one of the constituents which reduce the overall hydrophilic nature of the sweet dish. However, these values could be compared to those reported elsewhere (**Ahmed et al., 2004**) for Arabian sweet *Basbusa*(3 to 119 kJ/mol) for the percent moisture content range of 25 to 5 .The values of net isosteric heat q_{st} of *Sonpapdi* as a function of equilibrium moisture contents are shown in fig.4.
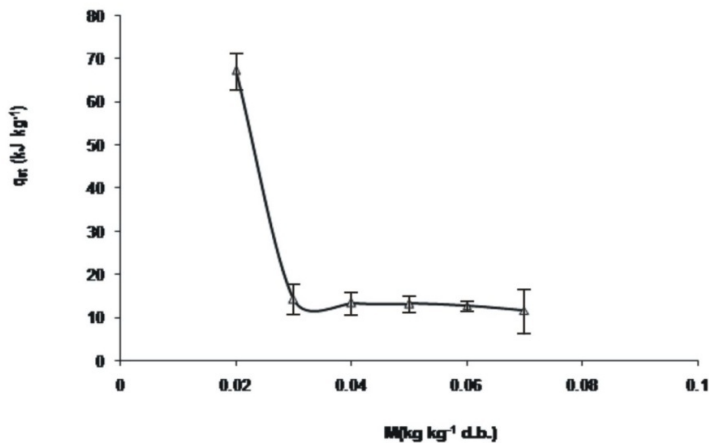
Figure 4 Plot between q_{st} and moisture content

It is clear that q_{st} values show a decreasing trend with moisture content. The relatively higher isosteric heat values at low moisture contents is an indication of strong water - surface interaction in *Sonpapdi*. At low moisture contents, water is sorbed at the strongest binding sites on the external surface of the solid. As the moisture content increases, the number of available binding sites for water sorption decreases which results in lower values of net isosteric heat of sorption. Similar observations have been reported by a number of workers. The defferential entropy of sorption, S_d was also evaluated using the intercepts of $\ln a_w$ versus $1/T$ linear plots(see fig.4). As suggested **(Aguerre et al., 1986)**, it was assumed that at a specific moisture content, q_{st} and S_d did not very with temperature. The results are shown in fig.5. which presents the differential entropy as a function of the moisture contents. This figure also shows that S_d shows a decreasing trend with moisture content and this negative dependence is pronounced at low moisture content values. Similar results have also been reported elsewhere **(Thys et al., 2010)**.
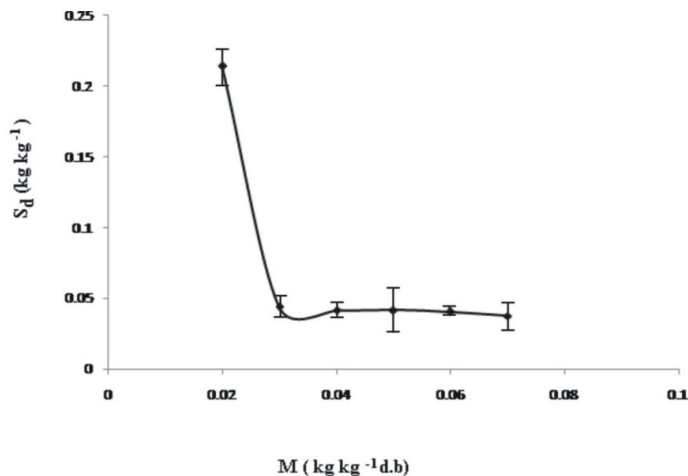

Figure 5 Plot between differential absorption entropy and moisture content

Enthalpy –entropy compensation theory

Entropy-enthalpy compensation theory (or isokinetic theory) is used to evaluate physical and chemical phenomena such as sorption reactions. It has been suggested that there exists linear relationship between enthalpy and entropy for water sorption in some foods, which has been confirmed by several authors **(Ferro-Fontan et al., 1982)**.

The plot of q_{st} versus S_d shows a fair linear relationship as depicted in fig.6 ($r^2 = 0.9998$), thus indicating that a fair compensation exists between q_{st} and S_d. The value of ΔG obtained (1624 jmol^{-1}) suggests that water sorption process is non-spontaneous (ΔG>0). Similarly, there have also been reported positive values of 1.0 kJ/mol for ready-to-use *Basundi mix* and 0.339 kJ/mol for *Pinhao Flour* **(Sharma et. al., 2009; Cladera-Olivwra et al., 2009).**
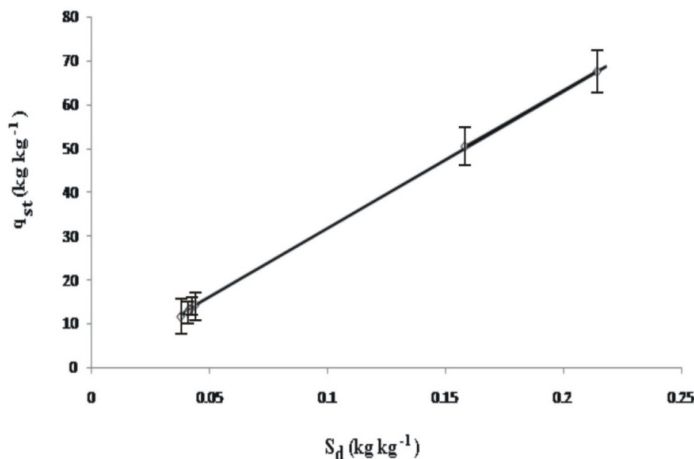

Figure 6 Plot between S_d and q_{st} to test the compensation theory

For Oatmeal biscuits and oat flakes, the ΔG values of 0.280 and 0.325 kJ mol^{-1} were reported respectively **(Mcminn et al., 2007)** . However non-spontaneous moisture sorption (i.e. ΔG<0) has also been reported by Thus et al. (2010) for sorption at *Pinhao* (araucaria angustifolia seeds) starch with ΔG value of -1.354 kJ mol^{-1} . Finally, the isokinetic temperature T_β and harmonic mean temperature T_{hm} were also evaluated using slope of linear-regression data (Fig.6) and equation (10) respectively. The values of T_β and T_{hm} were found to be 315.61 and 294.69 K respectively, thus showing an appreciable difference between the two values (i.e. $T_\beta > T_{hm}$). It has been suggested that , if $T_\beta < T_{hm}$ the process is enthalpy-driven, while if $T_\beta > T_{hm}$ the process is entropy controlled **(Leffler, 1995).** Since the former condition is satisfied in this study, the moisture sorption by *Sonpapdi* is enthalpy driven process. Similar results have also been reported by other workers **(Thus et al., 2010 ; Sharma et al., 2009 ; Mcminn et al., 2008)** .

CONCLUSION

It may be concluded that *Sonpapdi* has very low monolayer moisture content value, thus indicating that its preparation, packaging and consumption need careful attention, particulary in the environment of high relative humidity. The water vapor absortion by *Sonpapdi* is enthalpy driven process.

REFERENCES

AGUERRE, R.J., SUAREZ, C., VIOLLAZ, P.E. 1986. Enthalpy entropy compensation in the sorption phenomena. Application to the prediction of the effect of temperature on food isotherms. *Journal of Food Science,* 51(6), 1547-1549.
AHMAD, J., KHAN, A.R., HANAN, A.S. 2004. Moisture adsorption of an Arabian sweet (Basbusa) at different temperatures. *Journal of Food Engineering,* 64, 187-192.
AHMAD, N., SINGH, R.R.B., SINGH, A.K., PATEL, A.A., PATIL, G.R.P. 2008. Effect of maltodextrin addition on moisture sorption properties of Khoa. *International Journal of Dairy Technology,* 61(4), 403-410.
ANDERSON, R.B. 1946. Modification of Brunauer, Emmett and Teller Equation 1946. *Journal of American Chemical Society,* 68, 686-691.
ARABHOSSEINI, A., HUISMAN, W., MULLER, J. 2010. Modeling of the equilibrium moisture content (EMC) of Miscanthus (Miscanthus x Giganteus). *Biomass and Bioenergy,* 34, 411-416.
BLAHOVEC, J., YANNIOTIS, S. 2008 GAB generalized equation for sorption phenomena. *Food Bioprocess Technology,* 1, 82-90,
BRUNAUER, S., EMMETT, P.H., TELLER, E. 1938, Adsorption of gases in multimolecular layers. *Journal of American Chemical Society,* 60, 309-315.
RAMKRISHNAN, C., SRINIVASA, P.C., REDDY, S.R.Y. 2005. Moisture sorption characteristics of milk Burfi: a traditional Indian sweet using sugar substitutes. *European Food Research and Technology,* 220(2), 136-141.
CASSIN, A.S., MARCZAK, L.D.F., NORENA, C.P.Z. 2006. Water adsorption isotherms of texturized Soy protein. *Journal of Food Engineering,* 77, 194-199.
CAURIE, M. 1970. A new model equation for predicting safe storage moisture levels for optimum stability of dehydrated foods. *Food Technology,* 5, 301-307.
CERVENKA, L., REZKOVA, S., KRALOVSKY, J. 2008. Moisture adsorption characteristics of Gingerbread, a traditional bakery product in Parlubice. *Journal of Food Engineering,* 84, 601-607.

ERTUGAY, M.F., CERTE, M. 2000. Moisture sorption isotherms of cereals at different temperatures. *Nahrung,* 44(2), 107-109.

FABRA, M.J., TALENS, P., MORAGA, G., MARTINEZ-NAVARETE, N. 2009. Sorption isotherm and state diagram of grape fruit as a tool to improve product processing and stability. *Journal of Food Engineering,* 93, 52-58.

FARAHNAKY, A., ANSARI, S., MAJZOOBI, M. 2009. Effect of glycerol on the moisture sorption isotherms of Figs. *Journal of Food Engineering,* 93, 468-473.

FASINA, O.O. 2006. Thermodynamic properties of sweet potato. *Journal of Food Engineering,* 75, 149-155.

FONTANA, A.J. 2000. Understanding the importance of water activity in foods. *Cereals Food World,* 45(1), 7-10.

GABAS, A.L., TELIS, V.R.N., SOBRAL, P.J.A., TELIS, R.J. 2007. Effect of maltodextrin and Arabic gum in water vapor sorption thermodynamic properties of vacuum dried pineapple pulp powder. *Journal of Food Engineering,* 82, 246 - 252.

GOULA, A.M., KARAPANTSIOS, T.D., ACHILIAS, D.S., ADAMOPOULOS, K.G. 2008. Water sorption isotherms and glass transition temperature of spray dried tomato pulp. *Journal of Food Engineering,* 85, 73-83.

GUPTA, V., VIJAYLAXMI, N.S., ASHWANI, B., ANBARSU, K., VIJAYLAXMI, G., PRAKASH, M., INDIRAMMA, A.R., RAMESH, B.S., RANGARA, G.C.P. 2010. Shelli enhancement of coconut Burfi. An Indian traditional sweet. *Journal of Food Quality*, 33(3), 329-349.

HALSEY, G. 1948. Physical adsorption on non-uniform surfaces. *Journal of Chemistry and Physics*, 16, 931-937.

HASSAN, B. H. 2002. Moisture sorption characteristics of dried date powder. *Agricultural Science*, l(2), 105-112,

HENDERSON, M. 1952. A basic concept of equilibrium moisture. *Agricultural Engineering,* 33, 29-32.

HOSSAIN, S.A., PAL, P.K., SARKAR, P.K., PATIL, G.R. 2002. Moisture Sorption Charactertics of Dudh Churpi, a traditional milk product in India. *Nahrung*, 46(3), 136-140.

IGLESIAS, H.A., CHIRIFE, J. 1982. Handbook of food isotherms, New York: Academic press.

JAYENDRA KUMAR, A., SINGH, R.R.B., PATIL, G.R., PATEL, A.A. 2005. Effect of temperature on moisture desorption isotherms of Kheer. *Lebensmittel-Wissenchaft and Technology--Food Science and Technol*ogy, 38, 303-310.

KRUG, R.R., HUNTER, W.G., GRIEGER, R. A. 1976. Enthalpy entropy compensation .2.separation of the chemical from the statistical effect. *Journal of Physical Chemistry,* 80, 2335-2342.

KUMAR, A.J., SINGH, R.R.B., PATIL, G.R., PATEL, A.A. 2005. Effect of temperature on moisture desorption isotherms for Kheer. *Lebensmittel-Wissenchaft and Technology*, 38, 303-310.

LOMAURO, C.J., BAKSHI, A.S., LABUZA, T.P. 1985. Evaluation of food moisture sorption isotherm equations part II. Milk coffee, tea nuts, oilseed and starchy food. *Lebensmittel-Wissenchaft and Technology,* 18, 118-124.

MATHLOUTHI, M. 2001. Water content, water activity, water structure and the stability of food stuffs. *Food Control,* 12, 409-417.

OSWIN, C.R. 1946. The kinetics of package life III isotherm. *Journal Society of Chemical Industry*, 65, 419-421.

PEREZ-ALONSO, C., BERISTAIN, C.I., LOBATO-CALERAS, C., RODRIGUZ-HUEZO, M.E., VERNON-CARTER, E. J.2006. Thermodynamic analysis of the sorption isotherms of pure and blended carbohydrate polymers. *Journal of Food Engineering,* 77(4), 753-760.

RAO, K.J., DHAS, P.H.A., EMERALD, F.M.E., GHOSH, B.C., BALASUBRAMANYAM, B.V., KULKARNI, S. 2006. Moisture sorption characteristics of Chhana Poda at 5^0 C and 35^0C. *Journal of Food Engineering ,* 76, 453-459.

RIZVI, S.S.H., BENADO, A.L. 1983. Thermodynamics analysis of drying food. *Food Technology*, 2, 471-502.

RIZV, S.S.H. Thermodynamic properties of foods in dehydration. In Rao, A., Rizvi, S.S.H., Datta, A. K. (Eds.), Engineering properties of Foods. CRC, Florida USA.

SAHU, J.K., JHA, A. 2008. Storage stability of Sandesh an Indian milk sweet. In *Agricultural Engineering International,* 10, 1-12.

SARAVACOS, G.D., TSIOURVAS, D.A., TSAMI, E. 1986. Effect of temperature on the water adsorption isotherm of Sultana Raisins. *Journal of Food Science*, 51, 381-383.

SHARMA, P., SINGH, R.R.B., SINGH, A.K., PATEL, A.A., PATIL, G.R. 2009. Sorption isotherm and thermodynamics of water sorption of ready –to-use Basundi mix. *Lebensmittel-Wissenchaft Food Science and Technology*, 42, 441-445.

SINGH, R.R.B., RAO, K.H., ANJANEYULU, A.S.R., PATIL, G.R. 2001. Moisture sorption properties of smoked chicken sausages from spent hen meat. *Food Research International,* 34, 143-148.

THYS, R.C.S., NARENA, C.P.Z., MARCZAK, L.D.F., AIRES, A.G., CLADERA-OLIVERA, F. 2010. Adsorption isotherms of pinhao (Araucaria angustifolia seeds) starch and thermodynamic analysis. *Journal of Food Engineering,* 100, 468-473.

VANDEN BERG, C., BRUIN S. Water activity and its estimation in food systems: Theoretical aspects, In L.B. Rockland and G. F. Stewert (Eds) Water activity. Influence on food quality. (pp 1-62). New York : Academic press.

YAO, C.Y.B., TANO, K. 2008. Experimental determination of "Beta" lactose, new water absorbent and sodium bicarbonate. *Journal of Food Technology,* 6(4), 152-157.