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Moisture sorption characteristics and modelling of babool (*Acacia nilotica*) gum

Shristi Shefali Saraugi¹, Shadanan Patel¹, Chandrahas Sahu²*, Prashant S. Pisalkar¹, Pooja Sahu¹ and Dharmendra Khokhar¹

Abstract

Background: The study of sorption isotherm is the most important for prediction of shelf life, selection of packaging materials, drying characteristics, etc., of any food and agricultural materials. Gum is hygroscopic in nature, so finding of moisture sorption is crucial. Equilibrium moisture content of babool gum at 30, 40, 50 and 60 °C temperatures under relative humidity ranging from 11 to 95% was determined.

Results: Maximum equilibrium moisture content (EMC) of babool gum (33.6%, db) was recorded with the set of highest relative humidity (92.3%) at lowest temperature (30 °C), and minimum (2.8%, db) was obtained with the set of lowest relative humidity (10.9%) at highest temperature (60 °C) under the experimental conditions. The EMC data were employed in five well-identified sorption models, namely GAB, Iglesias and Chirife, Caurie, Halsey and BET model. GAB model best interpreted the data in a reasonable way as per statistical parameters of goodness of fit. The monolayer moisture content M_0 was found to be 8.46 and 7.49 g/100 g at 30 and 60 °C, respectively.

Conclusion: The sorption behaviours of babool gum were classified as type-II curves. It was obviously perceived that the isosteric heat decreases with the boost in moisture content.

Keywords: Babool gum, Equilibrium moisture content, Isosteric heat, Water activity, Relative humidity

Background

A natural gum, in general, is a water-soluble polysaccharide which is extractable from natural sources, especially from plants. Plant gums may be broadly classified into exudate gums, which are obtained as exudates of trees and shrubs and seed gums, which are obtained from the endosperms of seeds (Sao 2012). Chemically, natural gums are bio-polymeric materials composed of complex hetero-polysaccharides and proteinaceous materials, in addition to some mineral elements (Palipane and Driscoll 1993). Acacia nilotica (Babool tree) is one of the chief gum-yielding acacia species originated in the Indian subcontinent (Muniappan et al. 2009). Acacia nilotica

(Indian gum Arabic) belongs to the family *Leguminosae* and subfamily *Mimosaceae*. It is indigenously known as 'Babool' or 'Kikar.' *Acacia nilotica* has been acknowledged worldwide as a multipurpose tree (National Academy of Sciences 1980). The physical and chemical properties of acacia gum make it unique from other natural gums and that is why it is very importantly used in the food industry. The gum contains L-arabinose, L-rhamnose, galactose and four aldobiouronic acids. Acacia gum is also utilized in non-food industries for instance in novel pharmacy where it is frequently utilized as a binder, demulcent, emulsifier or for film-forming. Acacia gum is a complex composition of glycoproteins and polysaccharides predominantly consisting of arabinose and galactose.

The awareness of moisture adsorption isotherms for food products is of immense significance in designing and improving the processing operations like drying, dehydration, design of drying equipments, forecasting

² Department of Dairy Engineering, College of Dairy Science and Food Technology, CGKV, 492 012, Raipur, India Full list of author information is available at the end of the article



^{*}Correspondence: ercsahu2003@yahoo.com

the shelf life of products, selection of packaging materials, modelling for drying of products, designing in changes of moisture during storage, suitability of storage conditions, etc. A moisture sorption isotherm represents the affiliation between the equilibrium moisture content and water activity (a_w) for food products at a particular temperature and pressure (Kaymak-Ertekin and Gedik 2004). Some of the gums have been commonly studied for sorption characteristics by the previous researchers (Gabas et al., 2007; Vishwakarma et al. 2011). The hygroscopic behaviour of gums is an essential factor on moisture, textural properties and qualities (Brown 2007). The relationship between equilibrium moisture content of any food commodities and atmosphere is primarily important for determination of the appropriate processing conditions due to physico-chemical properties, microbial activities and other important characteristics which mainly depend on water activity (Rizvi 1986). It is worth mentioning that the water sorption characteristics are important in relation to stability and acceptance of food products. Water sorption isotherms are key thermodynamic tools for forecasting the interactions between food components and water. Interpretation of sorption data with suitable sorption model could be used as a tool for achieving these designs. According to literature the food commodities normally exhibit type-II and type-III water sorption isotherm as per the BET classification (Brunauer et al. 1940). In addition to above, the classic application of modelling of sorption isotherm data is to determine the optimum condition of storage and packaging systems to the knowledge of monolayer moisture content (Bell and Labuza 2000).

Efforts have been made in the past to study the sorption behaviour of xanthan gum by Basu et al. (2007), guar gum by Vishwakarma et al. (2011) and pectin by Panchev et al. (2010). However, the information on the water sorption characteristics of babool gum is not available in the literature. Therefore, the present investigation was initiated with the main aim of establishing the moisture sorption characteristics of babool gum in the range of temperatures 30–60 °C and relative humidity (10–95%). To contrast the sorption data, five popular mathematical sorption isotherm models were used to establish the most appropriate model describing the isotherm of babool gum. Determination of monolayer moisture content and net isosteric heat of sorption was also accomplished in this study.

Methods

Collection of plant exudate

The exudate babool (*Acacia nilotica*) gum samples were procured from the Network Project on Harvesting, Processing and Value Addition of Natural Resin and Gums operational at IGKV, Raipur (Chhattisgarh).

Preparation of sample

Gum samples were dried using sun drying and cleaned properly. The cleaned gum samples were pulverized and converted into uniform fine grits. To achieve this, the gum samples were grounded with the help of a laboratory crusher and passed through a sieve BSS-16 (width of aperture = 1 mm) and retained on BSS-22 (width of aperture = 0.71 mm). The weight of the grit samples obtained from the sieve was measured by an electronic balance (least count 0.001 g) and stored in air-tight box until further use.

Experimental site

The work was carried out in the Department of Agricultural Processing and Food Engineering, Swami Vivekananda College of Agricultural Engineering and Technology & Research Station, and Plant Physiology, Agricultural Bio-Chemistry, Medicinal and Aromatic Plant Department, Indira Gandhi Krishi Vishwavidyalaya, Raipur (Chhattisgarh).

Determination procedure and equipment for sorption analysis

Moisture sorption quality of babool gum sample was determined by the application of standard static gravimetric technique. Water activity $(a_{\rm w})$ of saturated salt solutions was adopted from the data provided by Greenspan (1977) and Labuza (1984). Saturated salt solutions made from laboratory grade salts were used in the study to maintain different level of relative humidity (11–95%) under four temperatures as shown in Table 1. The desiccators loaded with the solution of saturated salt were conditioned in equilibrium at experimental temperatures using BOD incubator up to four days before inserting the test samples for authentic study.

The exactly measured quantity of gum samples (about 2.5 ± 0.5 g) was placed inside the different preprepared desiccators for different levels of relative humidity. Desiccators were placed carefully inside the pre-set temperature-controlled chamber (BOD incubator). Samples of three set in the desiccators were kept throughout the test, and their mean was used to reduce the investigational fault. The reading of the samples was documented within the period of 3–4 days until it reached into equilibrium. The samples were permitted to equilibrate until there is no perceptible weight change (±0.001 g).

The equilibrium moisture contents of samples were expressed as g of water/100 g solids and determined by drying in a hot air oven at 102 ± 5 °C for 4-5 h (Koc et al. 2010). The plot between equilibrium moisture content

Table1 Saturated salt solutions and equilibrium relative humidity at different temperatures

S. no.	Saturated salt solution	Equilibrium relative humidity (%)					
		30 °C	40 °C	50 ℃	60 °C		
1	Lithium chloride (LiCl)	11.28	11.21	11.40	10.95		
2	Potassium fluoride (KF)	-	22.68	20.80	20.77		
3	Potassium acetate (CH ₃ CO ₂ K)	21.61	-	31.40	_		
4	Magnesium chloride (MgCl ₂)	32.44	32.70	-	29.26		
5	Potassium carbonate (K ₂ CO ₃)	43.17	-	-	37.70		
6	Magnesium nitrate (Mg (NO ₃) ₂)	51.40	48.42	46.30	47.70		
7	Cobalt chloride (COCl ₂)	-	55.48	50.01	-		
8	Sodium nitrite (NaNO ₂)	63.00	_	_	-		
9	Potassium iodide (KI)	-	66.09	64.49	63.11		
10	Sodium nitrate (NaNO ₃)	-	_	_	67.35		
11	Sodium chloride (NaCl)	75.00	74.68	74.70	-		
12	Potassium chloride (KCI)	83.62	82.32	-	80.25		
13	Potassium nitrate (KNO ₃)	92.31	89.03	85.00	-		
14	Potassium sulphate (K ₂ SO ₄)	_	-	95.80	95.82		

(EMC) and water activity (a_w) at particular temperature was indicated as the sorption isotherm graph.

Modelling of sorption isotherm

Numerous two and three parametric mathematical models for isotherm of food and food materials are available in order to model the experimental data obtained from the sorption experiment. The five different sorption isotherm mathematical models were analysed for sorption data (Table 2).

Statistical investigation

The present experiment was conducted in triplicate to minimize the experimental error, and average data were utilised for statistical analysis. The accuracy or goodness of fit for each of the sorption model was obtained by applying various statistical coefficients like adjusted R^2 , percentage relative deviation modulus (P), root-mean-square percentage error (%RMSE) and standard errors (SE). The most

excellent model was chosen on the basis of highest adjusted R^2 and least errors (%RMSE and P). The equations used to compute the accuracy of fit as recommended by Sahu et al. (2021) are as follows

Adjusted
$$R^2 = 1 - \frac{(N-1)\sum_{1}^{N} (WY - Y')^2}{(N-M)\sum_{1}^{N} (WY - Y'')^2}$$
 (1)

$$P(\%) = \frac{100}{N} \left[\sum_{1}^{N} \left(\frac{Y - Y'}{Y} \right) \right] \tag{2}$$

$$\%RMSE = \sqrt{\frac{1}{N} \left[\sum_{1}^{N} \left(\frac{Y - Y'}{Y} \right)^{2} \right]} \times 100$$
 (3)

$$SE = \sqrt{\frac{\sum_{i=1}^{N} (Y - Y')^2}{N - M}}$$
 (4)

where Y is experimental moisture content, %; Y' is predicted moisture content, %; Y'' is mean of EMC, %; W is weightage applied to each data point (W=1); N is number of inspections; M is number of coefficients.

Isosteric heat determination

The net isosteric heat of sorption is the major of thermodynamic indicators used to measure the requisite energy of the forces between the molecules of water vapour and material. The determination of net isosteric heat of sorption was done by using the general form of Clausius–Clapeyron equation (Eq. 5) suggested by Labuza (1984). The plot between natural log of water activity ($\ln a_{\rm w}$) and opposite of absolute temperature (1/T) exhibits a linear line called isoster. For any specified moisture content, the inclination of the line gives the value of $\left(\frac{q_{\rm st}}{R}\right)$ and net isosteric heat is calculated.

$$\ln\left(\frac{a_{w_2}}{a_{w_1}}\right) = \frac{q_{\rm st}}{R} \left(\frac{1}{T_1} - \frac{1}{T_2}\right) \tag{5}$$

Table 2 Isotherm models used to describe the sorption behaviour of babool gum

Isotherm model	Mathematical expression	References
GAB (Guggenheim–Anderson–de Boer)	$M = \frac{M_{o}CKa_{W}}{(1-Ka_{W})(1-Ka_{W}+CKa_{W})}$	Quirijns et al. (2005)
Hasley	$a_{\rm W} = \exp\left(\frac{-a}{M^b}\right)$	Hasley (1948)
Iglesias and Chirife	$M = a + b \left(\frac{a_{w}}{1 - a_{w}} \right)$	Iglesias and Chirife (1982)
Caurie	$M = \exp\left(a + ba_{\rm w}\right)$	Caurie (1970)
BET (Brunauer–Emmett–Teller)	$M = \frac{CM_{oa_{w}}}{[(1-a_{w})+(C-1)(1-a_{w})a_{w}]}$	Brunauer et al. (1938)

M-EMC, % db, a_w water activity, a and b constants, M_o monolayer moisture, % db, C Guggenheim constant, and k constant related to total heat of sorption of multilayer

where $q_{\rm st}$ is net isosteric heat of sorption, kJ mol⁻¹; R is universal gas constant (8.314 J K⁻¹ mol⁻¹); $a_{\rm w1}$ is water activity at temperature T_1 ; $a_{\rm w2}$ is water activity at temperature T_2 .

Results

Figure 1 shows the effect of change in relative humidity in the storage atmosphere on the colour of babool gum samples when kept at a temperature of 50 °C. Five models of isotherm were considered in this study to fit the sorption data as presented in Table 2. Moisture sorption behaviour of babool gum under different experimental temperatures is presented in Fig. 2a. The sorption

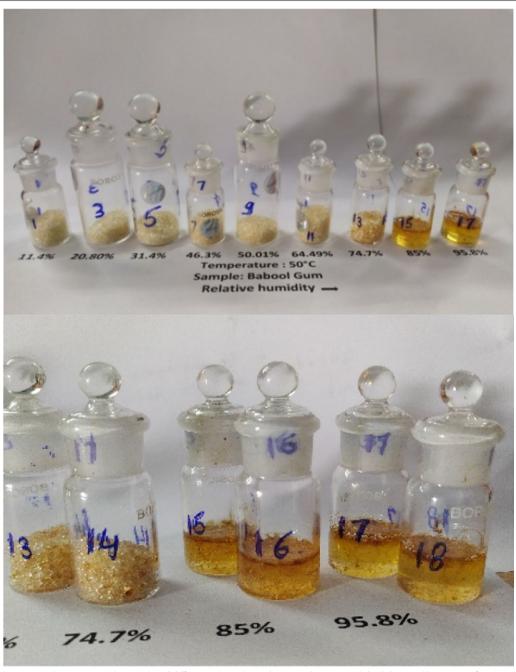


Fig. 1 Babool gum samples maintained at 50 °C and different relative humidity (samples accomplished the equilibrium, i.e. after 3 weeks)

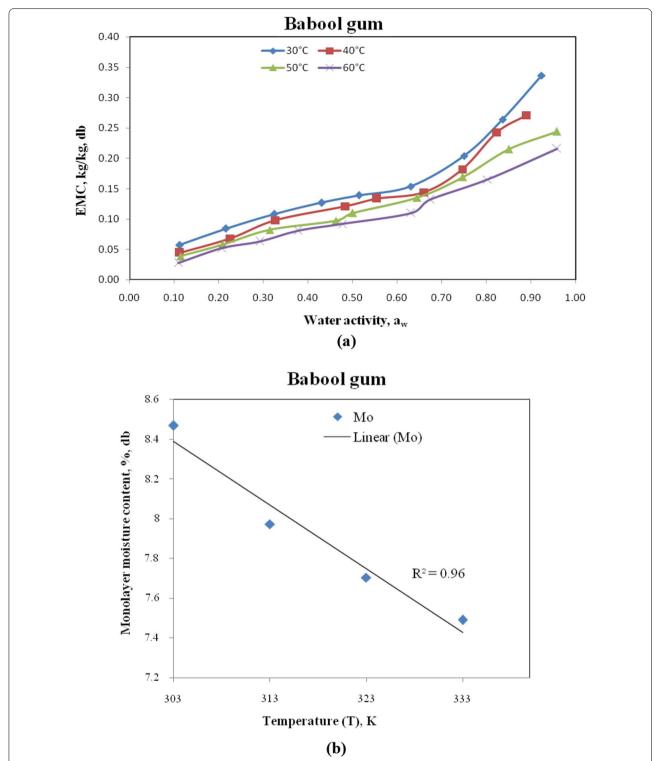


Fig. 2 a Sorption isotherms of babool gum at four different temperatures. **b** Variation of monolayer moisture content (M_0) with temperature (K) of babool gum

Table 3 Sorption models constant and accuracy values for experimental temperatures for babool gum

Model	Temperature (°C)	RMSE (%)	P (%)	Adj R ²	SE	M _o	С	К	а	b
GAB	30	6.42	0.239	0.983	0.997	8.467	0.007	0.806		
	40	7.73	0.019	0.965	1.307	7.972	0.014	0.810		
	50	3.252	0.041	0.995	0.462	7.703	0.018	0.779		
	60	8.541	0.038	0.971	1.203	7.490	0.028	0.815		
Iglesias and Chirife	30	9.13	0.961	0.984	1.047				5.477	11.033
	40	9.68	0.838	0.974	1.223				3.609	11.406
	50	16.619	4.209	0.97	1.315				4.673	8.239
	60	29.048	9.174	0.916	2.213				4.075	8.26
Caurie	30	25.352	3.444	0.503	0.059				- 1.405	532.12
	40	24.788	10.71	0.175	0.051				- 1.48	473.19
	50	25.75	11.31	0.179	0.05				- 1.532	445.24
	60	28.172	12.55	0.197	0.051				- 1.566	401.9
Halsey	30	10.026	0.525	0.954	0.017				3.531	1.963
	40	11.166	0.696	0.966	0.013				5.557	1.627
	50	18.873	1.729	0.773	0.036				1.811	2.026
	60	27.761	3.587	0.549	0.051				2.492	1.806
BET	30	12.649	7.285	0.854	0.011	7.32	27.32			
	40	7.338	0.075	0.932	0.007	7.17	9.31			
	50	33.665	31.78	0.865	0.034	5.9	13.41			
	60	28.652	22.43	0.182	0.025	4.18	6.13			

 M_o monolayer moisture content; C Guggenheim constant; K, a and b constants

isotherm model parameters and statistical goodness of fit constants were found out in this experiment which is presented in Table 3. The value of monolayer moisture content of babool gum is determined and reported in Table 3. The isoster graph of babool gum samples at different moisture content obtained in this study is shown in Fig. 3a. The changes in net isosteric heat of sorption of babool gum with moisture content are presented in Fig. 3b.

Discussion

Moisture sorption behaviour of babool gum

It can be depicted in Fig. 1 that the most of the babool gum samples endure in fair condition as fresh sample during the analysis up to 70% RH, but the sample which was subjected to higher humidity (>75%) noticed the loss of their original physical state and colour turned into yellowish. Additionally, no incidence of mould growth was observed on gum samples throughout the process of experiment.

At lower temperature and higher relative humidity values, the variation in EMC was comparatively higher than at higher temperatures for the equal relative humidity (Fig. 2a). Further, the sorption isotherms of the gum reveal that on increasing the water activity at a particular temperature the EMC increases. The reason might have

been due to hydrophilic nature of carbohydrates present in the babool gum. Also, with the enhancement in temperature at a given water activity, the EMC decreased which is obvious and similar to other numerous foods. The reason for this could be that when there is rise in temperature, water molecules get stimulated because of their energy level and become unsteady and rupture away from water-binding positions of hygroscopic materials compels reduction in the number of active spots for water binding at higher temperature (Phillips and Williams 2000). The figure indicated that the isotherms of babool gum exhibit sigmoid shape. This type of isotherms is common for almost all food materials. Similar results have also been obtained by Ahmad and Islam (2018) for commercial flours (wheat, rice and corn) and Vishwakarma et al. (2011) for guar grain and guar gum splits. This study revealed that the moisture sorption behaviour is important for safe and long storage of the products as well as selection of packaging materials for storing the products under different environmental conditions.

Modelling of sorption isotherm

The sorption isotherm model parameters and goodness of fit factors like %RMSE, adjusted coefficient of determination (R^2), standard error (SE) and mean absolute percentage error (P) have been used to evaluate the adequacy

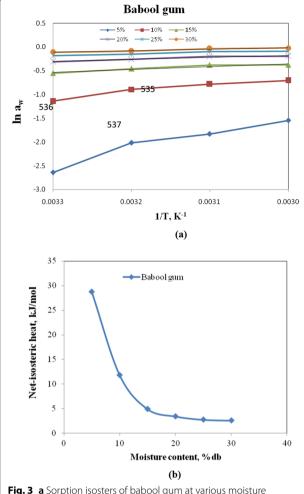


Fig. 3 a Sorption isosters of babool gum at various moisture contents. **b** Variation in net isosteric heat with moisture content in babool gum

of model in describing the relationship between water activity (a_w) and equilibrium moisture content (Sahu and Patel 2020). It can be observed from Table 3 that the GAB model has been found to have best representation of the sorption data because the values of the average grade statistical errors were found to be better in relation to other four models. The range of %RMSE and %P values of babool gum were found from 3.25 to 8.54% and 0.02 to 0.24%, respectively, for GAB model. High value of adjusted R^2 (0.96–0.99) also reveals its high adequacy of fitting the model. Hence, the GAB model gave the best fit for the adsorption isotherm of the test gum. The worst equation obtained for the test gums in BET model which was expected as it is thermodynamically suitable up to $0.50 a_{\rm w}$. Similar types of finding have also been obtained by Sahu and Patel (2020) for maize-millet-based soy-fortified extruded product and Ahmad and Islam (2018) for commercial flours (wheat, rice and corn).

Monolayer moisture content (M_o)

The monolayer moisture content $(M_{\rm o})$ is an indicator for the quantity of water firmly absorbed to definite sites at the exterior of foods, and this is a value that must be attained in order to satisfy food constancy. A graph of $M_{\rm o}$ versus absolute temperature is shown in Fig. 2b. It can be seen from Fig. 2b that the monolayer moisture content decreased with the increase in temperature which is obvious to many other food products. Similar result had demonstrated by Sahu et al. (2021) for extruded product. This may due to the fact that the increased temperature causes the drop in the extent of hydrogen bonding in the polymers, thus decreasing the accessibility of energetic portions for water binding and thereby, reduced the monolayer moisture content as narrated by Quirijns et al. (2005).

The BET and GAB models admit the prediction of monolayer moisture content ($M_{\rm o}$) of grain and food commodities. The remaining equations do not entertain the monolayer moisture content ($M_{\rm o}$) and unable to evaluate. The monolayer moisture content of the babool gum samples was obtained to be 8.46 g/100 g and 7.32 g/100 g, respectively, from GAB and BET models at 30 °C, and their equivalent values for 60 °C are 7.49 g/100 g and 4.18 g/100 g, respectively (Table 3). The monolayer moisture content calculated through the BET model is to some extent lower than the value obtained from the GAB experiment. Similar types of finding have also been resulted in the gellan gum (Abramovic and Klofutar 2005) and *gulabjamun* mix (Pushpadas et al. 2014).

The relationship between monolayer moisture content $(M_{\rm o})$ and absolute temperature (K) has also been obtained by using regression analysis. The linear equation (Eq. 6) for babool gum has been established to relate the relationship.

$$Y = -0.03x + 18.08 \quad \left(R^2 = 0.96\right) \tag{6}$$

where Y is monolayer moisture, and % db and x are absolute temperature (K).

The high value of coefficient of determination $(R^2 = 0.96)$ indicated that the developed linear relationship between monolayer moisture content and absolute temperature is adequately fitted the experimental data.

Isosteric heat of sorption

In order to establish the isosteric heat of sorption, a plot of natural logarithm of water activity $(a_{\rm w})$ as a function of the inverse of absolute temperature (1/T) for selected moisture contents was developed (Fig. 3a). From the graph it can be depicted that as the moisture content

increases, the slopes of isosters decreased. The slope of the isosters was employed to determine the net isosteric heat of sorption.

Figure 3b indicates that the changes in net isosteric heat of sorption, the moisture content also varied. It can be clearly revealed that net isosteric heat of sorption (q_{st}) has a negative dependency on moisture content. This reduction can mostly be described by the reality that when moisture content is less, the sorption takes place at the most energetic locations and offers the greatest interfacial energy. However, with the increase in moisture content the decreased number of dynamic portions accessible for water vapour sorption is quite possible. As the isosteric heat of sorption inclined and approaching to zero, the impact of the adsorbent on the absorbed molecules turned insignificant. Maximum and minimum values of net isosteric heat were 28.77 kJ mol⁻¹ and 2.58 kJ mol⁻¹, respectively, for corresponding moisture content 5% (db) and 30% (db) for babool gum. It can be additionally added that the net isosteric heat of sorption initially decreased very fast up to 15% moisture content (db) for babool gum, beyond which that the decline in isosteric heat was not appreciable. Similar type of finding has also found in gulabjamun mix (Pushpadass et al. 2014). Ahmad and Islam (2018) also reported the same results of net isosteric heat of sorption for commercial flours (wheat, rice and corn) of Bangladesh.

Conclusions

The sorption behaviours of babool gum at four experimental temperatures (30, 40, 50 and 60 °C) were found to be the curves of sigmoid shaped. At lower temperature and higher relative humidity levels, the variation in EMC was comparatively higher than at higher temperatures for the equal relative humidity. This means that moisture sorption isotherm of babool gum was temperature dependent. Based on the values of numerous error functions, GAB equation was found to be the most suitable model to describe the observed data as compared to other models applied in the experiments. The monolayer moisture content Mo, concluded from GAB model, was found to be 8.47 g/100 g and 7.49 g/100 g at 30 °C and 60 °C, respectively. Maximum and minimum values of net isosteric heat were 28.77 kJ mol⁻¹ and 2.58 kJ mol⁻¹, respectively, for corresponding moisture content 5% (db) and 30% (db) for babool gum.

Abbreviations

EMC: Equilibrium moisture content; GAB: Guggenheim–Anderson–de Boer; BET: Brunauer–Emmett–Teller; BSS: British Standard Sieve; BOD: Biological oxygen demand; RH: Relative humidity.

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Author contributions

SSS performed experiment work, material preparation, data collection and analysis of data. The first draft of the manuscript was written by SSS and CS and also interpreted the data. SP did the conception and design of the work. All authors commented on previous versions of the manuscript. PSP supported for the analysis of data and writing of paper. PS supported in experiment, data analysis and writing the manuscript. SP, CS and DK helped in the correction of different sections of the paper. All authors read and approved the final manuscript.

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Availability of data and materials

The authors declare that the data supporting the findings of this research are available within the article.

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Competing interests

There is no financial/personal interest or belief that could affect the objective, or if there is stating the source and nature of that potential conflict.

Author details

¹Department of Agricultural Processing and Food Engineering, Faculty of Agricultural Engineering, IGKV, 492 012, Raipur, India. ²Department of Dairy Engineering, College of Dairy Science and Food Technology, CGKV, 492 012, Raipur, India.

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