Modelling of sorption characteristics of Nigerian green tea

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Abstract: Green tea (*Camellia sinensis*) processing variables have been shown to influence variation in chemical constituents and acceptability of resultant tea products. This therefore suggests that basic green tea processing variables may influence quality of stored green tea. Thus, the present study investigated the effect of steaming time, drying temperature and time on the sorption characteristics of optimized Nigerian green tea. Green tea samples were conventionally produced and optimized using experimental runs. Static gravimetric method employed involve eight levels of water activities (0.1-0.8) using concentrated H_2SO_4 at temperatures of 27°C, 35°C and 40°C to simulate mini atmospheric condition. Plots of equilibrium moisture content (EMC) against corresponding water activity (a_w) were generated for isotherm curves. The adsorption data were fitted using common isotherm models (Caurie, Brunauer, Emmett and Teller (BET), Guggenheim Anderson and de Boer (GAB), Henderson, Oswin, Peleg and Smith). Adsorptive behaviour of optimized green tea samples showed equilibrium moisture content increases over elevated level of water activity at constant temperature. The sorption curved displayed sigmoid shapes characterized as Type II isotherm. Peleg model gave a better fit of the adsorption data with highest R^2 (0.9992) and least values of standard error of estimate (0.3529) and residual sum of square (0.4981). The GAB monolayer moisture content at 27°C, 35°C and 40°C ranged between 1.94% and 6.83%, 2.94% and 6.47%, and 3.31% and 6.65%, respectively. Thus, GAB equation suggests optimized green tea prepared by steaming for 120 s, dried at 70°C for 150 min will retain safe storage moisture of 6.83% d.b under practical condition at 27°C.

Keywords: Nigerian green tea, green tea, steaming, drying, sorption characteristics

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1 Introduction

Tea plant (*Camellia sinensis* L. Kuntze) native to the Theaceae family is source of tea types. Tea shoots are harvested and processed into tea beverages by changing simple tea unit operations, which pronounce the differences in tea products. Among the common ones, green tea is prepared by steaming fresh tea leaves to inhibit enzymatic oxidation, rolling to minute particle size and drying to obtain ready to be infused dried leaves (Kosińska and Andlauer, 2014). The resultant product retains essential bioactive compounds with potent therapeutic, physiological and health-protecting effects (Saeed et al., 2017). Such derived benefits mean adequate processing and storage conditions are vital to ensure the quality of green tea for its final consumers. A previous study assessing the quality of available Nigerian tea clones has suggested that the tea clones meet recommended standards for green tea

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preparation (Aroyeun et al., 2017).

Sorption characteristics of agricultural commodities could either be adsorption (taking up moisture) or desorption (releasing moisture). Both adsorption and desorption describes the interrelationship between moisture content of foods and simulated surrounding atmospheric conditions (temperature, pressure and relative humidity or water activity) to establish equilibrium moisture content data which best illustrates storage stability and drying behavior of the experimental material. Subsequently, the obtained data is plotted against water activities which is used to depict isotherm curves and as well fitted in suitable models to provide true description of sorption isotherm of the assayed product. Typically, sorption isotherm estimation based on standard static gravimetric procedure have been extensively reported in the literature for food products, either utilizing saturated salt solution (Ghodake et al., 2007; Sinija and Mishra, 2008; Varghese et al., 2014; Mutlu et al., 2020) or acid solution (Oguntoyinbo et al., 2015; Sobowale et al., 2017) to create a mini atmospheric condition expressed in water activity or percentage relative humidity. Consequently, at a fixed temperature, the understudied food material demonstrates water vapor diffusion till equilibrium moisture content is attained.

Fewer studies have examined the influence of temperature differences on sorption isotherm characteristics of tea, with results indicating that EMC increased with decreasing temperature, at constant relative humidity (Ghodake et al., 2007; Sinija and Mishra, 2008; Chen and Weng, 2010; Dalgic et al., 2012). Available literatures on effect of processing variables on green tea revealed variation in chemical component and sensory characteristics (Odunmbaku et al., 2015; Donlao and Ogawa, 2019). Odunmbaku et al. (2015) reported epigallocatechin gallate, epigallocatechin, epicatechin gallate and epicatechin levels of 46.90-178 mg g^{-1}, 0.30-4.24 mg g^{-1}, 1.03-8.83 mg g^{-1} and 8.05-33.96 mg g^{-1}, respectively. Likewise, 0.6-1.25 g 100 g^{-1}, 5.16-8.36 g 100 g^{-1}, 0.3-0.44 g 100 g^{-1}, 0.44-0.71 g 100 g^{-1}, 3.82-8.35 g 100 g^{-1}, 0.12-0.48 g 100 g^{-1}, 0.5-0.79 g 100 g^{-1}, 0.08-0.13 g 100 g^{-1}, 0.12-0.2 g 100 g^{-1} and 2.12-4.44 g 100 g^{-1}, gallocatechin, epigallocatechin, catechin, epicatechin, epigallocatechin gallate, gallocatechin gallate, epicatechin gallate, catechin gallate, caffeine and total chloropyll, respectively were reported by Donlao and Ogawa (2019). This therefore suggests notable quality changes would be initiated during storage life of green tea (approximately 18 months) with varying processing variables. With this in view, the present study aimed to establish the effect of Nigerian green tea processing variables (steaming time, drying temperature and time) on its sorption characteristics. Sorption isotherm data were interpreted graphically and as well analyzed using common sorption isotherm models (Al-Muhtaseb et al., 2002; Basu et al., 2007; Oguntoyinbo et al., 2015; Sobowale et al., 2017) to determine best suited model to characterize the isotherms of Nigerian green tea.

2 Materials and methods

2.1 Preparation of green tea

Tea shoots made up of apical bud and two leaves were harvested in 2015 from the Cocoa Research Institute of Nigeria (CRIN) experimental tea plots, Taraba State, Nigeria (7°20'N 11°43'E). Conventional processing and optimized processing variables using experimental runs (Table 1) generated in our earlier study were adopted for the production of green tea samples (Odunmbaku et al., 2015). Green tea was produced from cleaned tender shoot (100 g) following the basic unit operations of steaming at 115°C, shaping by rolling manually and oven drying (Hilal and Engelhardt, 2007). Average initial moisture content of the tea was 4%.

2.2 Determination of adsorption isotherm

The adsorption isotherm of green tea samples were determined using static gravimetric method described by Sobowale et al. (2017). This was carried out by creating eight levels of concentrated H_2SO_4 at temperatures of 27°C, 35°C and 40°C (Table 2). Three replicates of green tea (2.0 g each) was weighed into moisture pans and positioned on wire gauze in the desiccators. Initially, the desiccators containing the sample were conditioned at different water activities and kept at 27ºC, and intermittently weighed until constant weight was attained. The process was also repeated at 35ºC and 40ºC and average values were reported. The equilibrium moisture content (EMC) was obtained by estimating the initial moisture content and the known change in weight on dry basis (Oguntoyinbo et al., 2015). Plots of EMC against corresponding water activity (a_w) were generated for isotherm curves of green tea samples.

2.3 Modeling and validation of sorption isotherm

Seven commonly investigated mathematical models including Caurie, Brunauer, Emmett and Teller (BET), Guggenheim Anderson and de Boer (GAB), Henderson, Oswin, Peleg and Smith equations (Table 3) were used to fit experimental adsorption data. The sorption equation coefficients were determined by standard regression technique using DataFit version 9.0.59 statistical software. Standard error of estimate (SEE), residual sum of square (RSS) and coefficient of regression (R^2) were used as indicators to determine the goodness of fit of the isotherm models. SEE and RSS are expressed in Equation 1-2;

$$SSE = \sqrt{\sum_{i=1}^{n} (M_{cal}-M_{pred})^2 / N-1} \quad (1)$$

$$RSS = \sum_{i=1}^{n} (M_{cal} - M_{pred})^2 \quad (2)$$

Table 1 Experimental runs for green tea sample production

Sample	Steaming time (s)	Drying temperature (ºC)	Drying time (min)
292	90	65	150
194	60	70	150
321	120	60	150
573	90	70	120
250	120	65	120
926	60	60	150
531	60	65	120
756	90	60	120
430	90	65	120
271	120	70	90
980	90	65	90
658	120	60	90
621	60	60	90
738	60	70	90
564	120	70	150

Table 2 Quantity of concentrated H_2SO_4 per 250 mL water at different temperature

Water activity	27ºC	35ºC	40ºC
0.1	164.43	164.84	165.50
0.2	146.17	146.52	147.07
0.3	133.27	133.55	133.99
0.4	120.89	121.04	121.28
0.5	109.46	109.43	109.39
0.6	97.74	97.47	97.03
0.7	84.59	83.96	82.96
0.8	69.89	68.47	66.19

Table 3 Sorption isotherm models (Oguntoyinbo et al., 2015; Chen, 2019; and Mutlu et al., 2020)

Models	Equations
Caurie	$M= f_0 Exp(f_1 * a_w)$
Brunauer, Emmett and Teller (BET)	$M= \frac{a_w m_0 C}{(1 - a_w)(a_w(C -1) +1)}$
Guggenheim Anderson and de Boer (GAB)	$M = \frac{A*B*C*a_w}{(1-b*a_w)(1-B*a_w +B*C*a_w)}$
Henderson	$M = a_0(-\ln(1 - a_w)^{a_1}$
Oswin	$M= d_0 \left(\frac{a_w}{1-a_w}\right)^{d_1}$
Peleg	$M=Aa_w^B +Ca_w^D$
Smith	$M=A+B\ln(1 - a_w)$

Note: M, equilibrium moisture content (% dry basis); a_w, water activity; a_0, d_0, f_0, m_0, a_1, d_1, f_1, A, B, C and D are isotherm model constant

3 Results and discussion

The average equilibrium moisture contents obtained at different water activity and temperature (27°C, 35°C and 40°C) were used to graphically depict the experimental moisture adsorption isotherm curves of conventionally and optimized Nigerian green tea samples (Figures 1 - 4). For the control green tea sample, the sorption curve demonstrated initial upkeep increase in EMC (by 0.35 mg g^{-1}), which further increased from 27°C to 35°C and 40°C at constant water activity value of 0.8. This maybe generally attributed to maximum excitation state of moisture molecules present in the green tea samples at elevated temperature, therefore displaying reduction in their attractive forces and consequently lower level of moisture sorption at a specific water activity with increasing temperature from 27°C to 35°C and 40 °C. Similar observation has been reported for other tea samples (Ghodake et al., 2007; Sinija and Mishra, 2008; Donlao and Siriwattanayotin, 2012). On the other hand, with influence of processing variables on green tea samples, its adsorptive behaviour indicated EMC increases over a range of water activity at constant temperature of 27°C, 35°C and 40 °C. The adsorption isotherms of green tea samples were sigmoid shapes with respect to Brunauer, Emmett and Teller (BET) suggests a Type II isotherm (Mathlouthi and Rogé, 2003; Donlao and Siriwattanayotin, 2012). Similar isotherm type was also reported for green tea powder and granules (Sinija and Mishra, 2008; Tantala et al., 2019).

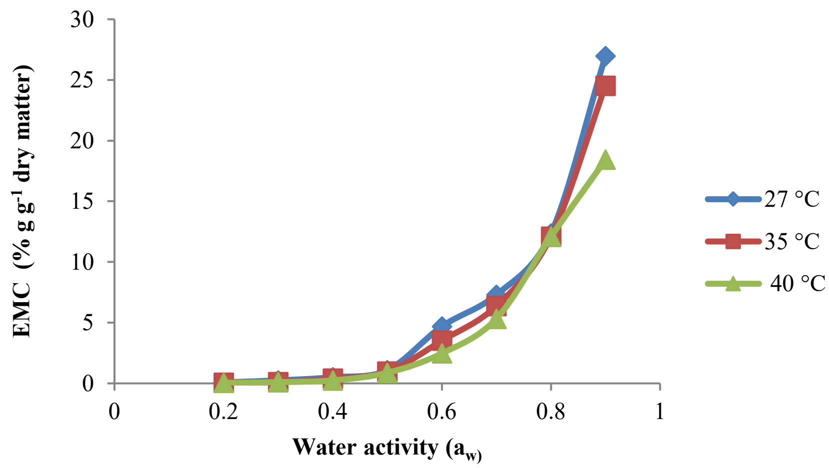

Figure 1 Experimental moisture adsorption isotherm of Nigerian green tea at 27°C, 35°C and 40°C

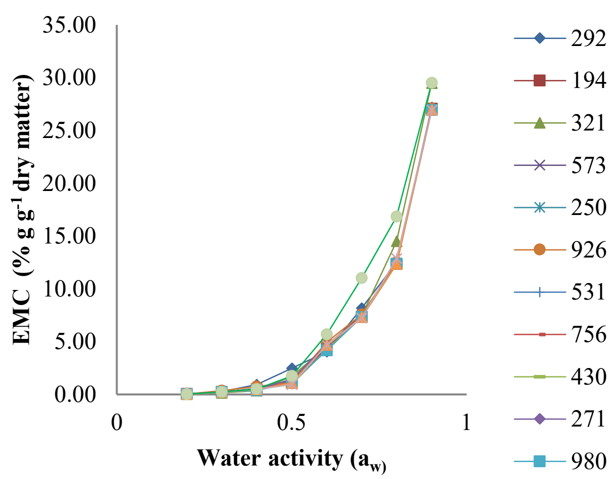

Figure 2 Moisture sorption isotherm of optimized green tea samples at 27°C

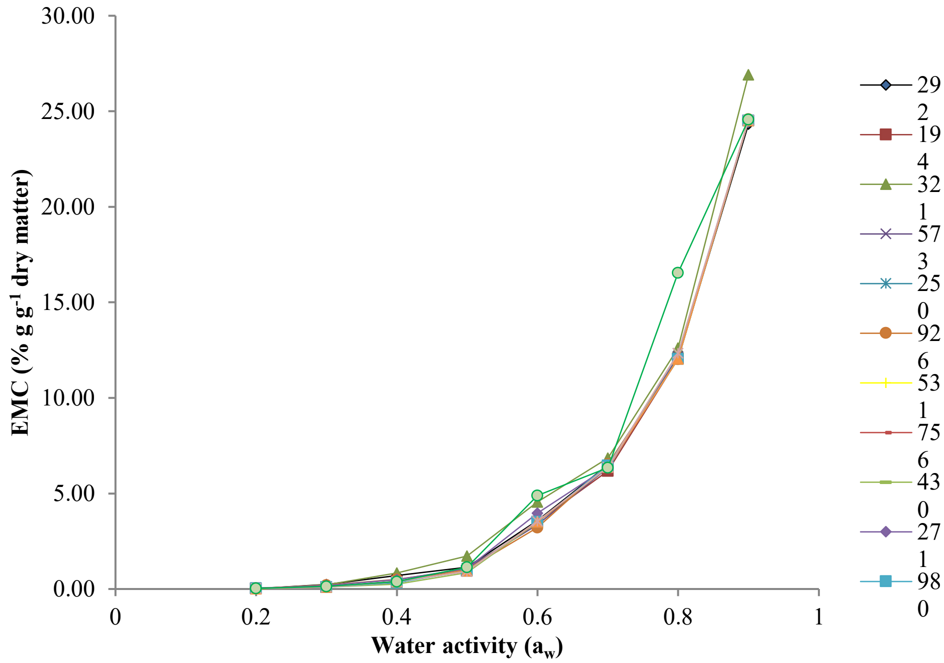

Figure 3 Moisture sorption isotherm of optimized green tea samples at 35°C

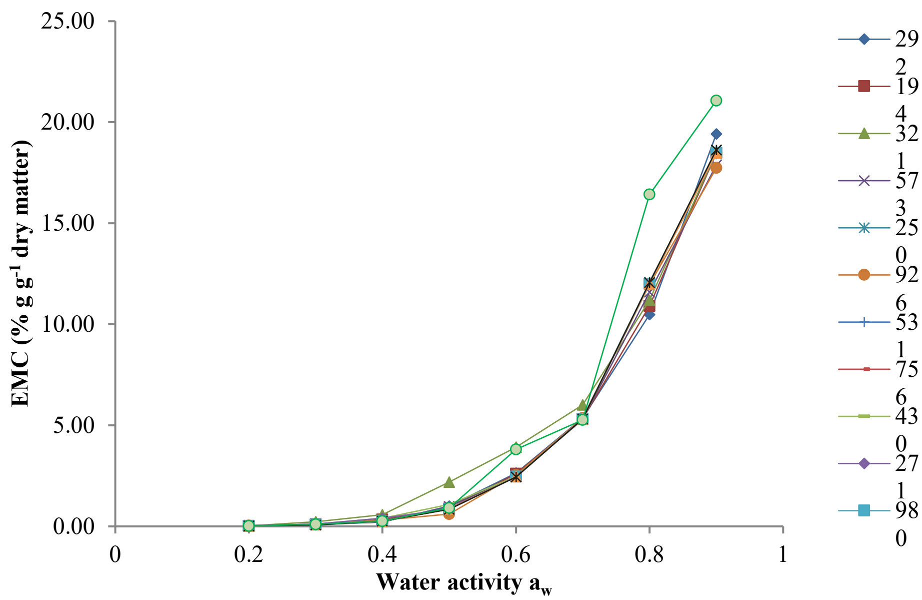

Figure 4 Moisture sorption isotherm of optimized green tea samples at 40 °C.

Table 4 Evaluation models indicators (adsorption)

Rank	Model	SSE	RSS	R^2	Ra^2
1	Peleg	0.3529	0.4981	0.9992	0.9985
2	Henderson	0.3797	0.8653	0.9985	0.9983
3	GAB	0.5872	1.7238	0.9971	0.9959
4	BET	0.5825	2.0360	0.9966	0.9960
5	Caurie	0.5970	2.1387	0.9964	0.9958
6	Oswin	0.8500	4.3352	0.9927	0.9915
7	Smith	2.4964	37.3927	0.9371	0.9266

Note: *SSE* – standard error of estimate; *RSS* – residual sum of squares; R^2 – coefficient of regression; Ra^2 – adjusted coefficient of regression

Table 5 GAB and BET mono layer moisture content of optimized green tea at different temperature

		GAB Monolayer Moisture content			BET Monolayer Moisture Content		
S/N	Process Variables	27°C	35°C	40°C	27°C	35°C	40°C
1	90sST:65ºCDT:150minDt	1.94	6.02	3.31	3.81	3.47	2.41
2	60sST:70ºCDT:150minDt	3.23	3.21	3.78	3.76	3.48	2.28
3	120sST:60ºCDT:150minDt	3.61	2.94	4.13	3.82	2.39	2.39
4	90sST:70ºCDT:120minDt	3.08	3.35	4.28	3.85	2.41	2.41
5	120sST:65ºCDT:120minDt	3.13	3.25	4.51	3.81	3.47	2.41
6	60sST:60ºCDT:150minDt	2.89	3.24	4.73	3.67	3.15	2.70
7	60sST:65ºCDT:120minDt	3.16	3.25	4.85	3.85	3.49	2.43
8	90sST:60ºCDT:120minDt	3.33	3.26	4.85	3.79	3.42	2.40
9	90sST:65ºCDT:120minDt	3.41	3.29	4.85	4.15	3.80	2.28
10	120sST:70ºCDT:90minDt	2.89	4.13	4.90	3.85	3.47	2.40
11	90sST:65ºCDT:90minDt	3.44	3.27	4.92	3.85	3.48	2.40
12	120sST:60ºCDT:90minDt	3.17	3.26	4.92	3.73	3.38	2.64
13	60sST:60ºCDT:90minDt	3.96	3.43	4.93	3.76	3.44	2.43
14	60sST:70ºCDT:90minDt	3.97	3.43	4.93	3.76	3.44	2.43
15	120sST:70ºCDT:150minDt	6.83	6.47	6.65	3.77	3.46	2.31

The evaluation model indicators for adsorption isotherm of the Nigerian green tea samples are presented in Table 4. A good fit model is characterized by lower values of standard error of estimate (*SSE*) and residual sum of squares (*RSS*), and higher coefficient of regression (R^2) value. The various models fitted with adsorption data gave good fit with closely related R^2 values (0.9371-0.9992) and lower SSE (0.3529-0.8500) and RSS (0.4981-4.3352) except for the Smith model. However, the Peleg model demonstrated better fit with the highest R^2 (0.9992) and least *SSE* (0.3528) and *RSS* (0.4981). Therefore, Peleg model is best suited for predicting the adsorption data of green tea under simulated storage conditions. The shelf stability of green tea samples during storage could be predicted using monolayer moisture content (M_o) (Iglesias et al., 1975). The GAB and BET monolayer moisture content of the optimized green tea samples at different temperatures (Table 5) revealed general increases in GAB monolayer moisture content values with increase in temperature whereas BET monolayer moisture content showed opposite trend with increases in temperature. The observed differences could be due to higher temperature effect on structural active sites of the understudy food material. In addition, GAB model gave higher monolayer moisture content values compared to BET equation. Similar observation has been reported for some food samples (Labuza et al., 1985).

4 Conclusion

This study investigated the effect of Nigerian green tea processing variables (steaming time, drying temperature and time) on its sorption characteristics. Adsorptive behaviour of optimized green tea samples showed equilibrium moisture content increases over elevated level of water activity at constant temperature. The adsorption isotherm curves were sigmoid shapes with respect to Brunauer, Emmett and Teller (BET) description of Type II isotherm. Peleg model gave a better fit of the adsorption isotherm data for optimized Nigerian green tea samples. The GAB equation seemed to have effectively indicated a safe storage moisture for green tea at 6.83% d.b. At this condition, the best steaming time was for 120 s and drying at 70ºC for 150 min.

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