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Evaluation of low-temperature drying characteristics of fresh tea leaves (*Camellia assamica*) in an environmental chamber using mathematical models

Anindita Sharma, Partha Pratim Dutta*

Department of Mechanical Engineering, School of Engineering, Tezpur University, Tezpur, India

**Corresponding author: anindita0302@gmail.com*

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Abstract: Low-temperature drying (withering) is the first stage in black tea processing. Determination of appropriate end moisture content of green tea leaf as well as temperature and relative humidity (RH) maintained during withering eventually aid the final quality of the processed tea. Therefore, the tea leaf withering (partial drying) properties were evaluated in an environmental chamber using mathematical models. The temperatures and RH considered were 25, 30, 35 °C and 80, 85, 90%, respectively. A total of nine combinations of temperature and RH were considered by keeping one parameter constant. The conditions were taken adhering to that of the climatic conditions of Assam, India. The withering data from experiments were fitted into five drying models using the curve fitting method. The Page model gave better predictions with an R^2 value of 0.9989 at 30 °C temperature and 90% RH. The total phenolic and flavonoid contents of the tea leaf samples were evaluated. The best results were 50.60 ± 0.02 mg GAE·g^{-1} (GAE – gallic acid equivalent) and 22.47 ± 0.01 mg QCE·g^{-1} (QCE – quercetin equivalent) at 30 °C withering temperature.

Keywords: fit; green tea-leaf; humidity; moisture ratio; wither

Across the globe, tea can be included among the widely consumed brews. In the tea production process, tea leaves are first dried at a low temperature after harvesting, i.e. they undergo withering. Withering makes the leaves flabby and decreases the moisture content. The leaves get physically conditioned, which aids rolling and consequently the drying process. Physical as well as chemical transformations occur in withering (Deb and Jolvis Pou 2016). A certain degree of withering has to be attained in order to roll the leaves well. The moisture content generally reduces to a range of 66–50% during withering, depending on air temperatures (Sharma and Dutta 2018).

Studies have been made on the drying behaviour of different products like tomato (Akhijani et al. 2016), eggplant (Chayjan and Kaveh 2016), bitter gourd (Vijayan et al. 2016), peppermint leaves (Salarikia et al. 2017), curry leaves (Vijayan et al. 2017), stevia leaves (Lakshmi et al. 2019), *Garcinia pedunculuta* (Dutta et al. 2021), etc. Research has been conducted on the drying of tea, particularly for black tea production, to a considerable extent. The drying kinetics of black tea were determined (Temple and van Boxtel 1999). The drying properties of black tea were computed by using a producer gas-fired tea dryer (Dutta and Baruah 2014; Dutta 2014). Again, such characteristics of Assam

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CTC tea (CTC – crush, tear, curl method of processing black tea) were evaluated in a domestic microwave oven (Hatibaruah et al. 2013). Both energy consumption and energy efficiency were stated to be strongly reliant on vacuum pressure and microwave power in tea leaves drying (Jindarat et al. 2013). However, there has been less work on the withering (partial drying) features of green tea leaves. A review was provided on the various technological and scientific aspects of the drying and withering of tea (Sharma and Dutta 2018). The energy utilisation pattern in Assam's tea leaf processing unit was reported (Sharma et al. 2018). Further, the withering characteristics of the local tea variety of Assam were mathematically modelled in another work (Sharma and Dutta 2021a). The exergy analyses were performed for the withering of tea leaves in the solar-based withering trough (Sharma and Dutta 2021b; Sharma and Dutta 2022). Moisture content was simulated with 1D heat and mass transfer models during tea leaf withering (Botheju et al. 2011a). Moreover, the thin-layer drying kinetics were computed (Botheju et al. 2011b). For the temperature range of 20–45 °C, tea leaf withering qualities were studied at 1.1 $m \cdot s^{-1}$ velocity of air along with evaluation of sorption isotherms (Ghodake et al. 2006; Ghodake et al. 2007). Non-linear fuzzy methods were employed to foretell the withering standard (Gupta et al. 2012). The temperature, as well as velocity distributions, varied extensively in a 3D withering trough model (Gupta et al. 2014).

The above literature summarises the fact that there have been considerable works on the drying operation of black tea processing. It becomes quite clear that very little focus has been given to the withering tea aspect, especially for the local tea samples of Assam, India. No such work has been reported for determining the withering characteristics of the Assam tea variety in an environmental chamber by controlling the temperature and relative humidity. Hence, an effort has been made to study the withering (low-temperature drying) properties of tea leaves adhering to this region specifically. As tea leaf withering depends mainly on temperature and relative humidity, a properly controlled environment would provide insight into the appropriate conditions for this stage in tea processing. The objectives/aims of the present study are:

(*i*) To determine the appropriate withering (partial drying) properties of green tea leaves in an environmental chamber, emphasising temperatures and RHs of drying air.

(*ii*) To determine the suitable combinations of green tea leaf physical withering temperature and RH by fitting the withering properties into five standard drying models.

(*iii*) To determine the total phenolic (TPC) and flavonoid contents (TFC) of the withered tea leaf samples.

MATERIAL AND METHODS

The current work attempted to determine the appropriate withering (partial drying) features of green tea leaves, emphasising physical variables of temperature and RH. Determination of the best combinations of green tea leaf physical withering temperature and humidity gives better product quality as well as ideal black tea manufacturing energy cost. Experimental studies were carried out in an environmental chamber (EC) with a temperature and humidity-controlled environment that had not been reported in the available literature for local tea samples of Assam, India. Usually, the tea withering process is carried out for a temperature range of 25–35 °C in this region. The RH is mostly high around 80–90% during the peak production season of black tea production. The present experimental and modelling work considered the local climatic variables as mentioned above by taking the extreme values with an increment of 5 units in both temperature and humidity. Moreover, the total phenolic and flavonoid contents have been determined for the partially dried tea leaf samples.

Experimental technique

Fresh green tea leaves were collected from a tea garden near Tezpur University, Tezpur, Assam, India (26°39'00.0"N latitude and 92°47'24.0"E longitude). The plucking was done as per the standard of plucking in the tea estates. The tea leaves were sealed in an airtight container and stored in a refrigerator to preserve moisture. The withering process was carried out in an EC (Model-REC-22038A2T; REICO, India), giving importance to the air temperature and RH. The EC and its schematic illustrations are shown in Figures 1A and 1B, respectively.

The oven-dry method was used to calculate the initial moisture of the leaf samples. A tea sample weighing 50 g was kept for 6 h at a temperature of 103 °C in a hot-air oven (114/14; REICO, India).

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(A) (B)

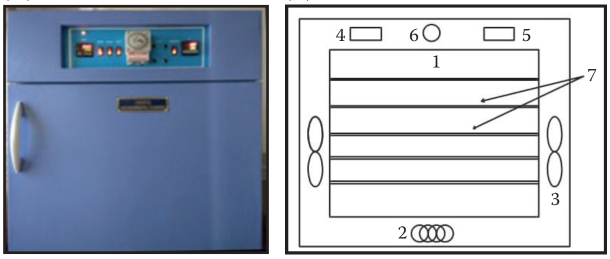

Figure 1. (A) The environmental chamber and (B) schematic diagram of the EC

1 – chamber area; 2 – heating element; 3 – fan; 4 – temperature display and control; 5 – humidity display and control; 6 – timer; 7 – chamber trays

The weight loss of the leaves gave the initial moisture content (Hatibaruah et al. 2013). The experiments were conducted by considering air temperatures of 25–35 °C and RH of 80–90% in increments of 5 units each. The temperatures and RHs were selected in accordance with the local climate of Assam during the peak tea production period and set in the EC as per requirement. A total of nine combinations of temperatures and RHs were considered by keeping one parameter constant. The experiments were conducted at an average velocity of 1 $m.s^{-1}$ of air. The EC was allowed to run without loading till it became stable with the set values of temperature and RH. The tea samples were loaded on six trays after dividing them into equal weights on top of the chamber trays. The trays were placed in the EC, and the experiment was allowed to run for 9 h. Weights of the samples were taken regularly after each hour in an electronic balance (HT-120; A&D Company Limited, Japan). The weighing process took an average of 2 min, and it was assumed that no disturbance took place due to the less time of weighing compared to the total withering period. Three repetitions for each combination of temperatures and RHs were done during the experimentation.

Following were some assumptions made during the study:

(*i*) Conduction heat transfer between leaves was negligible.

(*ii*) In between adsorption-desorption isotherms, there was no hysteresis effect.

(*iii*) Uniform and constant temperature and RH distribution amid the experiment.

(*iv*) Negligible time duration for weighing the samples hourly.

(*v*) Initial boundary condition: $M_{(t)} = M_{(0)}$ at $t = 0$.

Analysis of withering data

The tea leaf drying rate (D_r) is given by Equation (1) (Botheju et al. 2011a):

$$D_r = \frac{dM_{(t)}}{dt} = -K\left(M_{(0)} - M_{(e)}\right) \quad (1)$$

where: K – drying coefficient (h^{-1}); $M_{(t)}$ – moisture content at a given time t (% w.b.); $M_{(0)}$ – initial moisture content (% w.b.); $M_{(e)}$ – equilibrium moisture content (% w.b.).

However, as the initial moisture content for all the leaf samples differed from one another, the moisture ratio (MR) was determined to normalise the data. The moisture ratio was calculated by Equation (2) (Ghodake et al. 2006):

$$MR = \frac{M_{(t)} - M_{(e)}}{M_{(0)} - M_{(e)}} \quad (2)$$

The equilibrium moisture contents of the tea leaves were determined by the Guggenheim-Anderson-de Boer (GAB) equation as shown in Equation (3) (Panchariya et al. 2001):

$$M_{(e)} = \frac{(hM_m m_1 m_2)}{[(1 - m_2 h)(1 + m_1 m_2 h - m_2 h)} \quad (3)$$

where: h – relative humidity (%); M_m, m_1, m_2 – constants (values were 0.04354, 8.40585 and 0.94255, respectively for 25 and 30 °C; for 35 °C, they were 0.04237, 8.38243 and 0.93886).

Drying models and determination of drying coefficients

Five drying models were selected from the literature to fit the withering data (Hatibaruah et al. 2013). Table 1 gives the various models used for the analysis. The drying coefficients (k, k_1, k_2) for all the models were determined accordingly. The coefficients A, B and N were found from the curve fitting.

The non-linear regression method was employed to find the best fit among the models to the withering data obtained. The Curve Fitting Tool in MATLAB (version 15) was used for the purpose. The experimental results were found to be statistically significant. The goodness of fit was evaluated by the coefficient of determination (R^2) and root mean square error (RMSE). The more the value of R^2 approaches 1, the better is the fit. On the other

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Table 1. Drying models

Serial No.	Models	Equations
I.	Lewis	$MR = e^{(-kt)}$
II.	Henderson & Pabis	$MR = Ae^{(-kt)}$
III.	Logarithmic	$MR = Ae^{(-kt)} + B$
IV.	Page	$MR = e^{\left(-kt^{N}\right)}$
V.	Two-term	$MR = Ae^{(-k_1 t)} + Be^{(-k_2 t)}$

MR – moisture ratio; e – exponential; k, k_1, k_2 – drying coefficients; A, B, N – coefficients of the drying model

hand, a lesser value of RMSE indicates a better fit. The R^2 and RMSE are calculated by Equation (4) and (5) (Holman 2007).

$$R = -\frac{S_r}{S_t} \tag{4}$$

where: S_r – residual sum square; S_t – total sum square.

$$\text{RMSE} = \left[\frac{1}{N}\sum_{i=1}^{N}(MR_{ei} - MR_{pi})^2\right]^{\frac{1}{2}} \tag{5}$$

where: MR_{ei} – experimental MR; MR_{pi} – predicted MR.

Specific energy consumption

The specific energy consumption (SEC) during the low-temperature drying process of fresh tea leaves can be determined by (Salarikia et al. 2017):

$$SEC = \frac{E_t}{M_w} \tag{6}$$

where: E_t – total energy supplied in the chamber (MJ); M_w – mass of water removed by drying (kg).

Determination of total phenolic content (TPC) and total flavonoid content (TFC)

Phenols and flavonoids are some chemical compounds which affect the overall health benefits and flavour quality of made tea. Tea contains a higher amount of flavonoids among similar varieties of food and beverages. It is therefore essential to assess the total phenolic and flavonoid contents in the partially dried tea leaves.

The withered tea leaves were dried to an extent such that they can be coarsely ground. The TPC and TFC of the samples were determined using standard procedures. The contents were found out for the tea leaf samples withered at the temperatures of 25, 30, and 35 °C.

Method of extraction. The withered tea leaves were finely ground. A partially dried tea leaf sample of (0.2 ± 0.001) g was balanced into 10 mL graduated extraction tubes and 5 mL of 70% hot water/methanol extraction mixture at 70 °C, dispensed into the extraction tubes, and a vortex mixer was used to mix it. 10 min of incubation was done for the extraction tubes in the water bath and vortexed after 5 and 10 min, respectively. The tubes were allowed to cool and centrifuged for 10 min at 3 500 rpm after removing from the water bath. Similarly, a second extraction was done. The extracts were combined and made up to 10 mL with a cold methanol/water extraction mixture and mixed in a vortex mixer (Kerio et al. 2013).

TPC was determined by using Folin-Ciocalteu's reagent as per ISO 14502-1-2005E. For the sample extract, one millilitre was put in a volumetric flask of 100 mL and made up to mark with distilled water. 1 mL of diluted sample was mixed with 5 mL reagent and 4 mL of 7.5% Na_2CO_3 solution for one hour before performing the spectrometric analysis. Gallic acid was used as a standard for determining TPC. The results were expressed as milligrams of gallic acid equivalent (GAE) per gram extract (mg GAE·g^{-1}) (Kerio et al. 2013). The TFC were determined by taking quercetin as standard with colourimetric assay. The results of TFC were expressed as milligrams of quercetin equivalent (QCE) per gram extract (mg QCE·g^{-1}) (Kritsadaruangchai et al. 2019).

RESULTS AND DISCUSSION

Withering characteristics. Fresh tea leaves from a local tea garden were collected to determine the low-temperature drying or withering properties in an environmental chamber. Nine different combinations of temperatures and RHs in accordance with the climatic conditions of Assam, India, were studied. The moisture content of the tea leaves was obtained a 76% initially. Figures 2A, B show the tea leaf samples before and after withering, respectively. The oven-dried tea sample is shown in Figure 2C.

Figures 3A–C represent the experimental and simulated withering (drying) properties of green tea leaves over a period of 9 h. Figure 3A shows the

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Figure 2. (A) Fresh tea leaves, (B) withered tea leaves, and (C) oven-dried tea leaves

withering curves at a constant humidity of 80% and temperature range of 25–35 °C. As expected, with a temperature rise, there is an increase in the withering rate. The moisture ratio (*MR*) decreases by 49, 60, and 64% after 9 h for the considered temperature range, thus showing a surge in the rate of withering at the highest temperature. The predicted results show good agreement with the experimental ones. Botheju et al. (2011b) reported such falling rates when similar conditions of temperatures and RHs were adopted to wither fresh tea leaves.

Figure 3B shows the withering curves at a constant humidity of 85% and temperature range of 25–35 °C. The moisture ratios change by 45, 49, and 56% after 9 h for the three considered temperatures, respectively. The decrease in moisture content is comparatively lower than in the previous case because, with the increase in humidity, there is a decrease in the capacity of air to remove moisture from the leaves. A similar pattern of deviance of the experimental data from the simulated results is observed here too.

The withering curves at 90% humidity and temperature range of 25–35 °C are shown in Figure 3C. The moisture ratio decreases from 38 to 52% after 9 h of partial drying for the same rise in the temperature range. Due to an increase in the RH, withering is the slowest in this case. Botheju

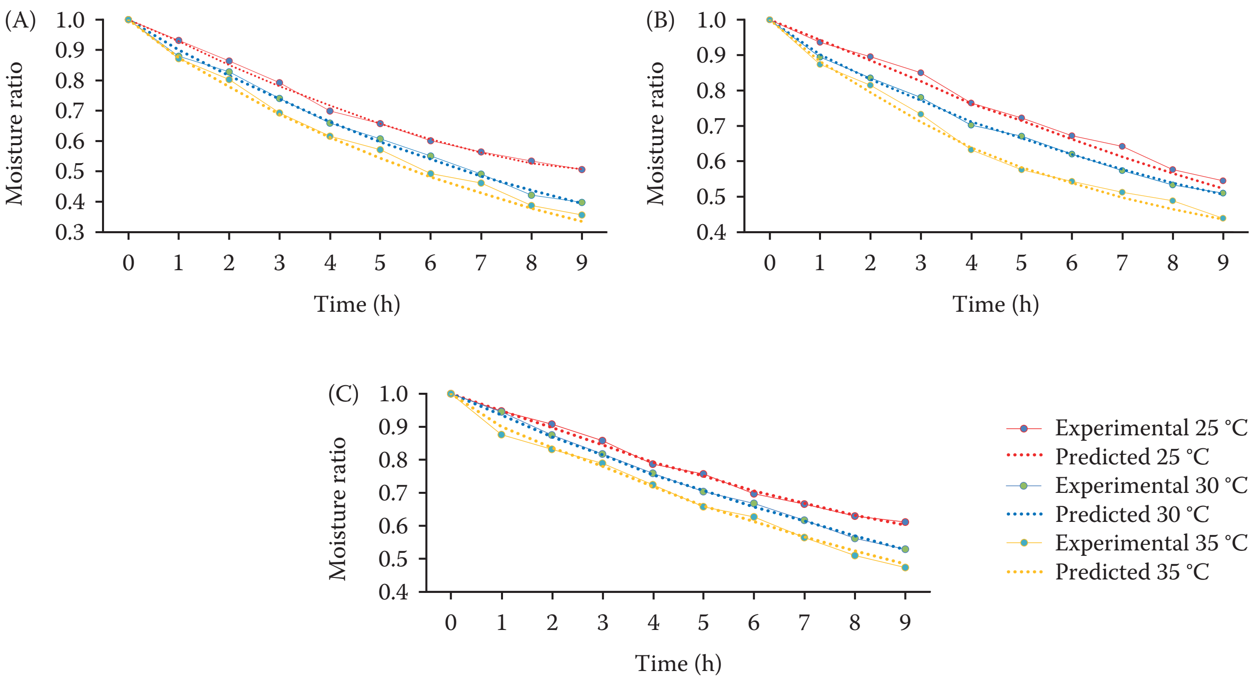

Figure 3. Moisture ratio *vs*. time at (A) RH = 80%, (B) RH = 85%, and (C) RH = 90%

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et al. (2011b) and Ghodake et al. (2006) obtained such an increase in the withering rate with an increase in air temperature.

The withering curves at constant temperature and variable RHs are shown in Figures 4A–C for both experimental and simulated results. Figure 4A represents the withering curves at a constant temperature of 25 °C with humidity of 80, 85, and 90%. The *MR*s decrease by 49, 45, and 39% at the end of 9 h for the three RHs, respectively. The withering rate is observed to be the lowest and highest at 90 and 80% humidity, respectively, thus following the general trend of reduction of drying rate at higher humidity.

Figures 4B and 4C show the withering characteristics at 30 and 35 °C, respectively, with the same range of RHs. A similar trend of withering rate is followed in both cases as in the previous instance. At constant temperatures of 30 and 35 °C, the moisture ratios come down by 60, 49, 47%, and 64, 56, 52% respectively, after 9 h of withering at the three levels of RH. This clearly shows that the moisture contents reduce with the rise in RH at a constant temperature. However, the rate of withering is faster when the temperature increases. Botheju et al. (2011b) and Ghodake et al. (2006) reported similar withering properties again.

From all the considered cases, it is evident that the withering rate is the highest at an air temperature of 35 °C with 80% humidity. The arrangement of 25 °C and 90% has the lowest withering rate. This is again in compliance with the findings of Botheju et al. (2011b) and Ghodake et al. (2006). A moderate withering rate is seen at an air temperature of 30 °C and 90% humidity as compared to the rest.

Five drying models were used to fit the experimental withering data. Table 2 illustrates the drying curve parameters obtained from the regression analysis. Out of all the five models, the Page model gave the best fit at 30 °C and 90% with the coefficient of determination (R^2) value of 0.9989 and RMSE value of 0.0051. In previous work, Botheju et al. (2011b) found the Page and Two-term models as the best-suited drying models for drying tea leaves in Sri Lanka. On the other hand, Ghodake et al. (2006) reported that the Henderson and Pabis model described the lower temperature properties suitably and the Page model suited the higher temperatures.

The mathematical model for the best fit is as follows:

$$MR = e^{\left(-0.0636t^{1.0020}\right)} \tag{7}$$

where: e – exponential.

Specific energy consumption. The specific energy consumed at a constant RH of 90% and tem-

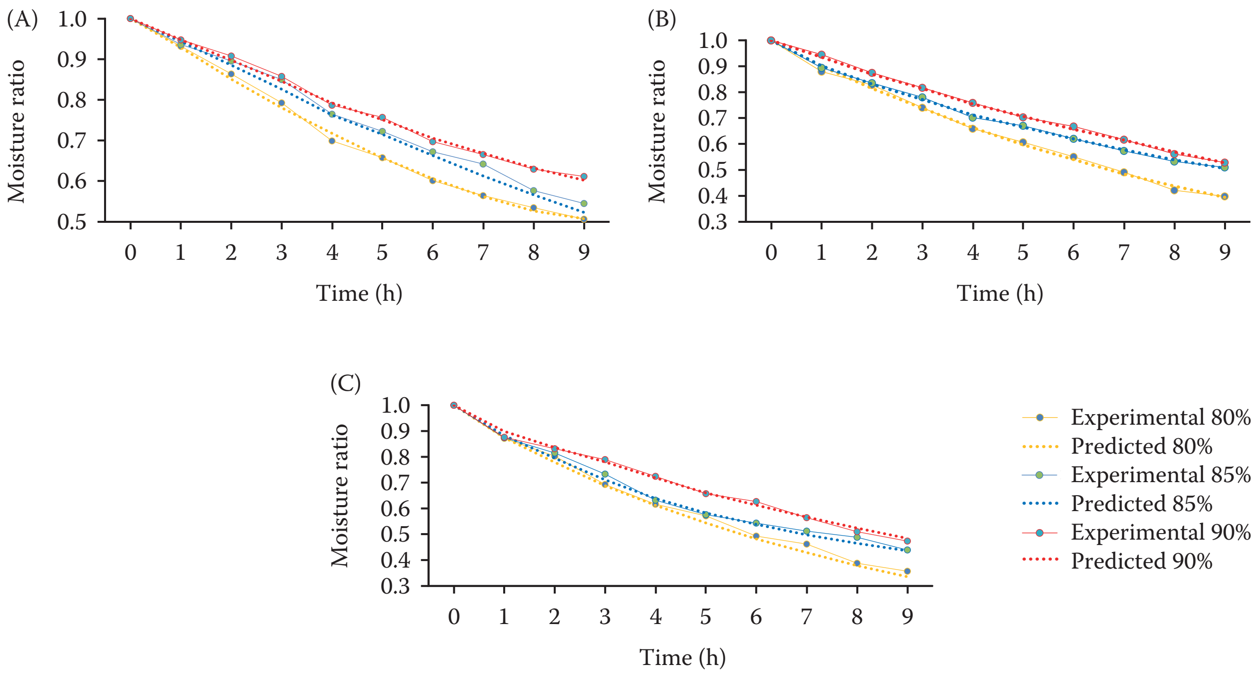

Figure 4. Moisture ratio *vs.* time at (A) T = 25 °C, (B) T = 30 °C, and (C) T = 35 °C

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Table 2. Drying curve parameters for the withering data

Model	T (°C)	RH (%)	Constants						R^2	RMSE
			k	k_1	k_2	A	B	N		
Lewis	25	80	0.0806	–	–	–	–	–	0.9930	0.0145
	30	80	0.1025	–	–	–	–	–	0.9965	0.0122
	35	80	0.1160	–	–	–	–	–	0.9967	0.0120
	25	85	0.0655	–	–	–	–	–	0.9926	0.0134
	30	85	0.0800	–	–	–	–	–	0.9903	0.0160
	35	85	0.0994	–	–	–	–	–	0.9801	0.0263
	25	90	0.0569	–	–	–	–	–	0.9943	0.0133
	30	90	0.0640	–	–	–	–	–	0.9988	0.0053
	35	90	0.0827	–	–	–	–	–	0.9894	0.0175
Henderson & Pabis	25	80	0.0809	–	–	1.0010	–	–	0.9930	0.0154
	30	80	0.1019	–	–	0.9971	–	–	0.9965	0.0126
	35	80	0.1145	–	–	0.9920	–	–	0.9970	0.0123
	25	85	0.0675	–	–	1.0110	–	–	0.9939	0.0139
	30	85	0.0761	–	–	0.9788	–	–	0.9945	0.0138
	35	85	0.0942	–	–	0.9736	–	–	0.9847	0.0244
	25	90	0.0578	–	–	1.0060	–	–	0.9948	0.0135
	30	90	0.0641	–	–	1.0010	–	–	0.9988	0.0054
	35	90	0.0795	–	–	0.9830	–	–	0.9919	0.0162
Logarithmic	25	80	0.1182	–	–	0.7887	0.2244	–	0.9953	0.0135
	30	80	0.0848	–	–	1.1240	–0.1324	–	0.9970	0.0116
	35	80	0.1209	–	–	0.9612	0.0332	–	0.9971	0.0111
	25	85	0.0350	–	–	1.7150	–0.7130	–	0.9956	0.0127
	30	85	0.1188	–	–	0.7372	0.2543	–	0.9972	0.0097
	35	85	0.1733	–	–	0.7020	0.2980	–	0.9941	0.0162
	25	90	0.0646	–	–	0.9252	0.0823	–	0.9949	0.0159
	30	90	0.0646	–	–	0.9944	0.0067	–	0.9988	0.0057
	35	90	0.0632	–	–	1.1610	–0.1832	–	0.9923	0.0169
Page	25	80	0.0844	–	–	–	–	0.9749	0.9933	0.0151
	30	80	0.1010	–	–	–	–	1.0080	0.9965	0.0126
	35	80	0.1232	–	–	–	–	0.9658	0.9972	0.0121
	25	85	0.0544	–	–	–	–	1.1010	0.9959	0.0106
	30	85	0.1009	–	–	–	–	0.8708	0.9978	0.0080
	35	85	0.1313	–	–	–	–	0.8420	0.9920	0.0178
	25	90	0.0536	–	–	–	–	1.0330	0.9947	0.0136
	30	**90**	**0.0636**	–	–	–	–	**1.0020**	**0.9989**	**0.0051**
	35	90	0.0927	–	–	–	–	0.9366	0.9910	0.0171
Two-term	25	80	–	0.0866	–0.6736	1.0110	0.0001	–	0.9969	0.0149
	30	80	–	0.1023	0.1004	0.9136	0.0845	–	0.9965	0.0156
	35	80	–	0.1251	0.1150	–0.0392	1.0320	–	0.9970	0.0143
	25	85	–	0.5347	0.0737	–0.0578	1.0550	–	0.9960	0.0160
	30	85	–	3.2140	0.0728	0.0400	0.9610	–	0.9979	0.0092
	35	85	–	–0.0850	0.1352	0.0824	0.9161	–	0.9943	0.0172
	25	90	–	0.0460	0.0538	–0.5042	1.5100	–	0.9948	0.0162
	30	90	–	0.4696	0.0640	0.0011	1.0010	–	0.9988	0.0062
	35	90	–	0.0887	0.0786	0.1045	0.8792	–	0.9919	0.0187

T – temperature; RH – relative humidity; k, k_1, k_2 – drying coefficients; A, B, N – coefficients of the drying model; bold – best fitting model

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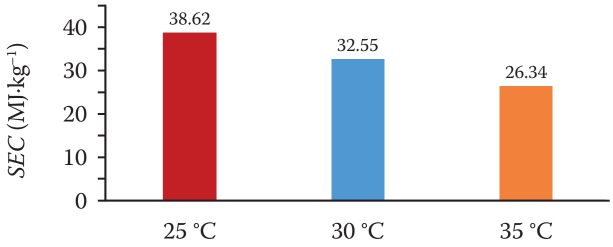

Figure 5. Specific energy consumption (*SEC*) at different temperatures

peratures of 25, 30, and 35 °C is shown in Figure 5. It is observed that the *SEC* decreases with the increase in drying air temperature, the lowest being 26.34 MJ·kg^{-1} at 35 °C. This may be explained as the result of faster drying or withering at a higher temperature. Similar trends in *SEC* were observed in peppermint leaves drying (Salarikia et al. 2017).

Total phenolic and flavonoid contents. The TPC and TFC of the partially dried tea leaf samples were determined using standard procedures for the samples withered at 25, 30, and 35 °C. Figures 6 and 7 show the TPCs and TFCs, respectively, at all mentioned temperatures. It is clearly observed that the highest contents in both cases are found at 30 °C as 50.6 ± 0.02 mg GAE·g^{-1} and 22.47 ± 0.01 mg QCE·g^{-1}, respectively. No relevant literature has been found as such for the phenolic and flavonoid contents of withered tea samples at specific temperatures. However, Kritsadaruangchai et al. (2019) reported the TPC and TFCs of unfermented Assam tea samples as 89.20 ± 1.05 mg GAE·g^{-1} and 77.43 ± 0.67 mg QCE·g^{-1}, respectively. Kaur et al. (2015) observed greater polyphenol content in green tea than in black tea for Indian samples.

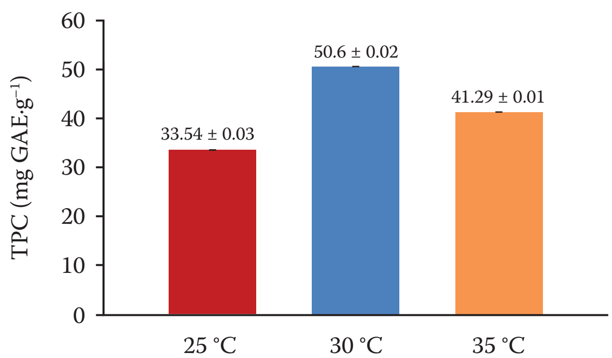

Figure 6. Total phenolic content (TPC)

GAE – gallic acid equivalent

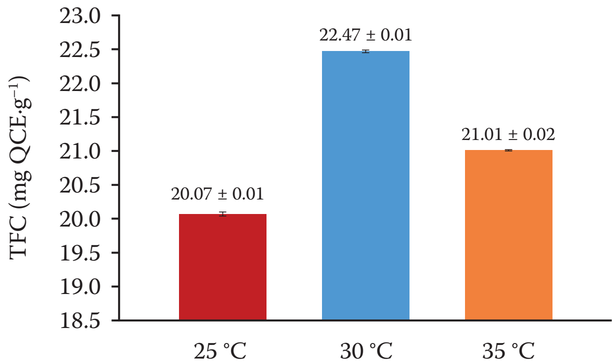

Figure 7. Total flavonoid content (TFC)

QCE – quercetin equivalent

CONCLUSION

In the present work, the withering characteristics of green tea leaves were determined from the experiments conducted in an environmental chamber. The data obtained from experiments were fitted in five models. The following are the conclusions which are drawn from the study:

(*i*) Withering of fresh tea leaves took place in a combination of both constant-rate as well as falling-rate drying period.

(*ii*) Both drying air temperatures and RHs affected the withering characteristics.

(*iii*) The withering rate was maximum at 35 °C and 80% RH with the moisture ratio coming down by around 64% at the end of 9 h. At 25 °C and 90% RH, there is a minimum fall in moisture ratio by around 38%, thus showing the least withering rate.

(*iv*) The Page model gave the best fit at a drying air temperature of 30 °C and 90% RH.

(*v*) The *SEC* were 38.62, 32.55, and 26.34 MJ·kg^{-1} at drying air temperatures 25, 30, and 35 °C for a constant RH of 90%, respectively.

(*vi*) The highest TPC and TFC were estimated at 30 °C as 50.60 ± 0.02 mg GAE·g^{-1} and 22.47 ± 0.01 mg QCE·g^{-1}, respectively.

As a future work, the withering (partial drying) features of the fresh tea leaves may be obtained under similar conditions by developing an experimental set-up and application of renewable energy.

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