Chapter-4

Modeling and experimental withering of tea leaves

As mentioned in the introduction chapter, tea leaves are dried in low temperature after harvest. This process is termed as withering. The withered leaves become flabby after the decrease of the surface moisture in them. The reduction in moisture content mainly depends on temperature and relative humidity. Both physical as well as chemical changes occur during withering. A certain point of withering has to be reached to aid the rolling process [43,153].

The drying behaviour of products like tomato [9], eggplant [36], bitter gourd [189], cocoa beans [122,123], chicken breast [112], olive leaves [62], peppermint leaves [148], curry leaves [190], *vernonia amygdalina* leaves [12], stevia leaves [106] and many more have been studied upon. The literature survey summarizes that there have been considerable research on the drying operation of black tea processing. From the available literature, it is quite clear that very little emphasis has been given on the tea withering operation, especially for the local tea variety of Assam, India. This chapter discusses an attempt made to determine the low temperature drying or withering characteristics of local tea leaves in an environmental chamber by controlling the temperature and relative humidity as per requirement. As the tea leaf withering process depends mainly on these two factors, a properly controlled ambience would provide an insight into the suitable conditions for this stage in tea processing.

4.1 Materials and methods

Determination of suitable combinations of tea withering temperature and humidity gives superior product quality as well as an ideal energy cost for black tea processing. The proper withering or partial drying attributes of freshly plucked tea leaves were obtained by giving importance to physical parameters of temperature and relative humidity. An environmental chamber (EC) with a humidity and temperature controlled environment was used to carry out the experimental studies for fresh tea leaf samples of Assam, India. In general practice, the tea withering process in this region is operated for a temperature range of (25-35) °C. The RH remains mostly between (80-90) % during the peak black tea production season. This experimental and modelling attempt considered these indigenous climatic variables by taking the extreme values with an increase of five units in both the parameters. Additionally, the TPC and TFC have been determined for the withered tea leaf samples.

4.1.1 Experimental technique

Local variety of fresh tea leaves were plucked from a tea garden near Tezpur University, Tezpur, Assam, India (26.65 ºN latitude and 92.79 ºE longitude). The collected tea leaves were kept in an airtight container and placed in a refrigerator for prevention of moisture loss. The tea withering or partial drying process was carried out in an EC (Model-REC-22038A2T, Make: REICO, India) by emphasizing on the air temperature and RH. The schematic illustration of the EC is shown in Fig. 4.1. The EC and the lea teaves wilted in it are shown in Fig. 4.2(a) and 4.2(b) respectively.

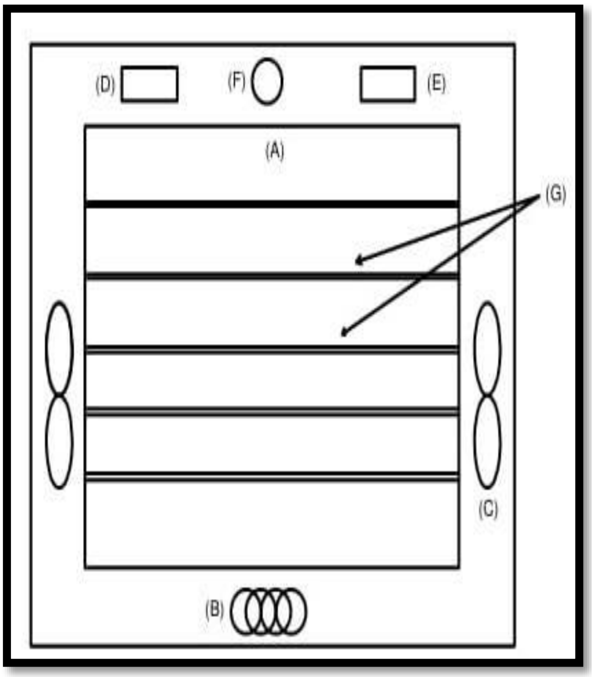

Fig. 4.1. Schematic illustration of the EC

(A) Chamber area
(B) Heating element
(C) Fan
(D) Temperature display and control
(E) Humidity display and control
(F) Timer
(G) Chamber trays

Fig. 4.2(a). The environmental chamber

Fig. 4.2(b). Tea leaves being withered inside the EC

The initial moisture of the tea leaf samples was estimated by the oven-dry method. Tea samples weighing 50 g was kept at temperature 103 °C for 6 h in a hot-air oven (Make: REICO, India). The initial moisture content was calculated from weight loss of the leaves by repeating three times [74]. The experiments in the EC were conducted by considering air temperatures (25-35) °C and RH of (80-90) % in increments of five units each. The parameters were selected and set in the EC as required in agreement with the climatic condition of Assam during peak tea production season. Nine different combinations of temperature and RH were set by keeping one variable constant at a time. The experiments were conducted at an average air velocity 1 m/s [33, 66]. The chamber was allowed to run devoid of loading initially till it became steady with the set values of temperature and humidity. The green tea samples were divided in equal weights on six trays and placed over the trays of the EC. The experiment was allowed to run for 9 h after loading. The test samples were weighed hourly using an electronic balance (HT-120, A&D Company Limited). The weighing of the samples took around 2 minutes. No disturbance was assumed to take place due to the lesser time of weighing as compared to the total withering period. The experiments were repeated three times for each combination and the mean values of the readings were considered.

Some assumptions made for the study are as follows-

- Heat transfer through conduction between the leaves is very small.
- Absence of hysteresis effect between adsorption-desorption isotherms.
- Uniform temperature and RH throughout the experiment.
- Insignificant time to weigh the samples.
- Initial boundary condition: $M_{(t)} = M_{(0)}$ at $t = 0$.

4.1.2 Analysis of withering data

The tea-leaf drying rate (D_r) is given by Eq. (4.1) [32]:

$$D_r = \frac{dM_{(t)}}{dt} = -k\left(M_{(0)} - M_{(e)}\right) \tag{4.1}$$

where, k = drying coefficient (s^{-1}). As the initial moisture content for all the leaf samples were different from one another, the moisture ratio (MR) was calculated to normalize the data. The moisture ratio was calculated by Eq. (4.2) [66]:

$$MR = \frac{M_{(t)} - M_{(e)}}{M_{(0)} - M_{(e)}} \tag{4.2}$$

where, $M_{(0)}$ = initial moisture content, $M_{(t)}$ = moisture content at a given time (t) and $M_{(e)}$ = equilibrium moisture content.

The $M_{(e)}$ of the tea leaves were determined by the GAB equation as shown in Eq. (4.3) [136]:

$$M_{(e)} = \frac{(HM_m m_1 m_2)}{[(1-m_2H)(1+m_1m_2H-m_2H)} \tag{4.3}$$

where H is the RH. The values of the constants M_m, m_1 and m_2 were 0.04354, 8.40585 and 0.94255 respectively for 25 °C and 30 °C. For 35 °C, they were 0.04237, 8.38243 and 0.93886.

4.1.3 Drying models and determination of drying coefficients

Five standard drying models were selected from the literature to fit the withering or partial drying data obtained from the experiments [33, 66, 74]. Table 4.1 gives the 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 estimated from the curve fitting.

Table-4.1. Drying models [33, 66, 74]

Sl. 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	$MR = Ae^{(-k_1 t)} + Be^{(-k_2 t)}$

The non-linear regression method was used to find the best fit among the drying models for the withering data obtained. The Curve Fitting Tool in MATLAB was used for the purpose. The experimental results were statistically significant. The goodness of fit for the data was evaluated by the coefficient of determination (R^2), adjusted R^2 and root mean square error (*RMSE*). The more the value of R^2 approaches 1, the finer is the fit. On the other hand, a smaller value of *RMSE* gives a better fit. Adjusted R^2 is always less than or equal to the value of R^2. On adding new variables to a regression model, the value of R^2 never decreases. In fact it remains the same or increases while doing so. Whereas, the adjusted R^2 considers the addition of such variables in a model. The R^2, *RMSE* and R^2_{ad} are calculated by Eq.(4.4), Eq.(4.5) and Eq.(4.6) respectively [79].

$$R^2 = 1 - \frac{S_r}{S_t} \qquad (4.4)$$

where, S_r and S_t are residual sum square and total sum square respectively.

$$RMSE = \left[\frac{1}{N} \sum_{i=1}^{N} (MR_{ei} - MR_{pi})^2 \right]^{\frac{1}{2}} \qquad (4.5)$$

where, MR_{ei} = experimental MR and MR_{pi} = predicted MR.

$$R^2_{ad} = 1 - \frac{MS_r}{MS_t} \qquad (4.6)$$

where, MS_r and MS_t are residual mean square error and total sum square error respectively.

4.1.4 Specific energy consumption

The specific energy consumption (SE) during the low temperature drying process of fresh tea leaves can be determined by [148]:

$$SE = \frac{E_t}{M_w} \qquad (4.7)$$

where, E_t = total energy supplied in the EC (MJ) and M_w = mass of water removed by drying (kg).

4.1.5 Determination of Total Phenolic Content (TPC) and Total Flavonoid Content (TFC)

The phenols and flavonoids are certain chemical substances effecting the total health and flavour quality of made tea. Tea contains higher amount of flavonoids than that in similar categories of beverages. Hence, it becomes essential to estimate the TPC and TFC in the withered or partially dried tea leaves.

The tea leaves were withered to a level such that they can be ground easily. The TPC and TFC of the withered tea-leaf samples were estimated using standard procedures. The contents were evaluated for the tea-leaf samples withered at the temperatures of 25 °C, 30 °C and 35 °C.

4.1.5a Method of extraction

The partially dried tea leaves were ground finely. A withered 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, bestowed into the extraction tubes and a vortex mixer was used to mix it. The extraction tubes were incubated for 10 minutes in the water bath. They were vortexed after 5 and 10 minutes respectively. The tubes were centrifuged for 10 minutes at 3500 rpm after cooling them. Similarly, a second extraction was carried out. Both the extracts were merged and made up to 10 ml with cold methanol/water extraction mixture and then mixed in a vortex mixer [95].

TPC was estimated by using Folin-Ciocalteu’s reagent according to ISO 14502-1-2005E standard. For the sample extract, 1 ml was put in a volumetric flask of 100 ml and made upto mark with distilled water. 1 ml of the diluted sample was amalgamated with 5 ml reagent and 4 ml of 7.5% Na_2CO_3 solution for 1 h before doing the spectrometric analysis. TPC was determined by using gallic acid as a standard. The results were expressed as milligrams of gallic acid equivalent (GAE) per gram extract (mg GAE/g) [95]. Quercetin was used as standard to determine TFC with colorimetric assay. The results of flavonoid were expressed as milligrams of quercetin equivalent (QCE) per gram extract (mg QCE/g) [100].

4.2 Results and discussion

4.2.1 Withering characteristics

The withering or low temperature drying properties of fresh tea leaves collected from a local tea garden were determined in an EC. Nine different combinations of temperatures and RHs in agreement with the regional climatic conditions were considered. The oven-dry method gave the moisture content of the tea leaves as 76% initially. Figures 4.3 and 4.4 show the tea-leaf samples before and after withering respectively. The oven-dried tea sample is shown in Figure 4.5.

Fig. 4.3. Fresh tea leaves

Fig. 4.4. Withered tea leaves

Fig. 4.5. Oven-dried tea leaves

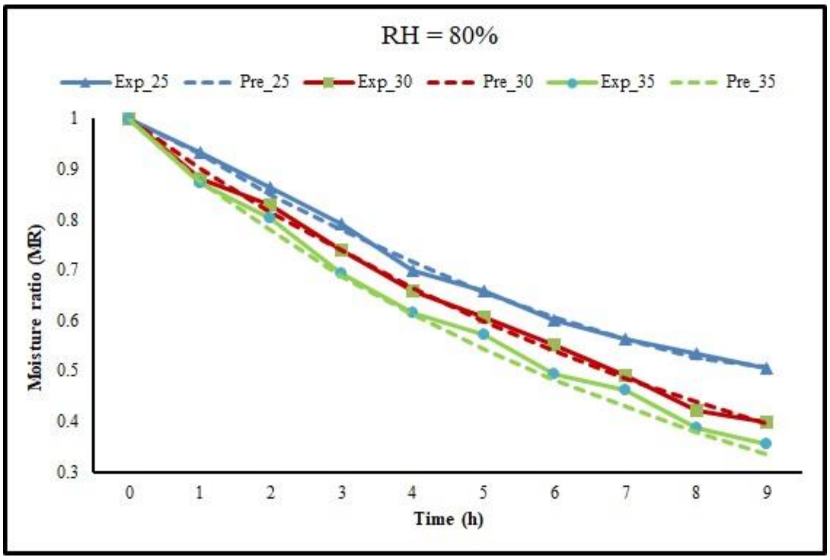

Fig. 4.6. *MR* vs Time (h) at RH = 80%

The experimental and predicted withering or low temperature drying attributes of fresh tea leaves over a period of 9 h are represented in Figures 4.6-4.8. Fig. 4.6 depicts the withering properties at a constant RH of 80% and temperature range of (25-35) ºC with an increment of 5 ºC. As per general trend, there is an increase in the withering rate with a rise in temperature. The moisture ratio (*MR*) decreases to 0.51, 0.4 and 0.35 after 9 h for the considered temperatures range, thus showing a high rate of withering as the temperature surges. A constant rate withering is observed after 6 h at 25 ºC. The average withering rates at the three temperatures are respectively 0.28, 0.30 and 0.32 g water/g dry solid-h. The predicted results showed good agreement with the experimental ones. Such falling rates were reported when similar conditions of temperatures and RHs were adopted to wither fresh tea leaves [33].

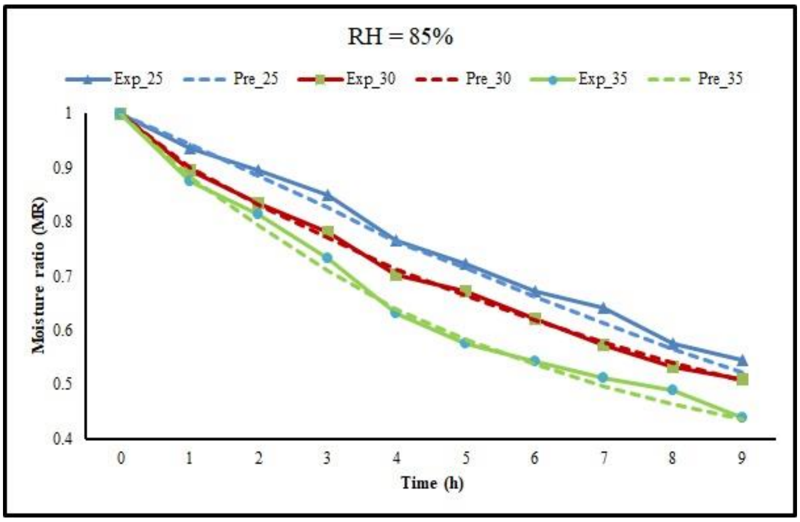

Fig. 4.7. *MR* vs Time (h) at RH = 85%

The withering properties at air temperature range of (25-35) ºC and a constant humidity of 85% are shown in Fig. 4.7. The moisture ratios change to 0.54, 0.51 and 0.44 after 9 h for the three considered temperatures respectively. The decrease in moisture content was comparatively lower than the previous case because with the increase in humidity, there is a decrease in the capacity of air to remove moisture from the leaves. The partial drying rates on an average are 0.27 g water/g dry solid-h at 25 ºC, 0.28 g water/g dry solid-h at 30 ºC and 0.30 g water/g dry solid-h at 35 ºC respectively. The similar pattern of deviance of the experimental data from the simulated results is observed here too.

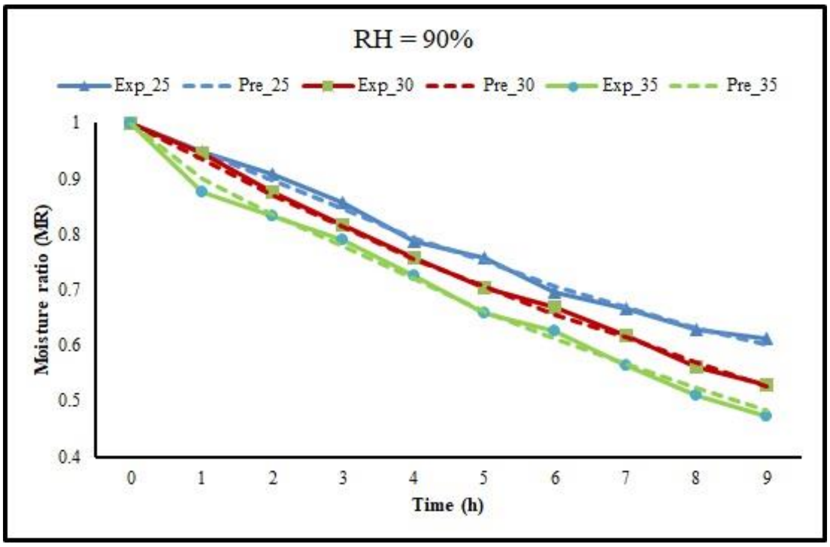

Fig. 4.8. *MR* vs Time (h) at RH = 90%

Fig. 4.8 shows the withering properties at temperature range of (25-35) ºC and 90% humidity. The moisture ratio decreases to 0.61, 0.53 and 0.47 after 9 h of partial drying for the same rise in temperature range. Due to an increase in the RH, the withering was the slowest in this case. At 25 ºC, the average rate of partial drying was 0.26 g water/g dry solid-h whereas it was 0.28 and 0.29 g water/g dry solid-h respectively at 30 ºC and 35 ºC. At 25 ºC, a constant rate of withering is seen after 8 h in this case. Botheju et al. and Ghodake et al. obtained such increase in the withering rate with an increase in air temperature [33,66]. The predicted values are in better agreement with the experimental values than the rest in this combination.

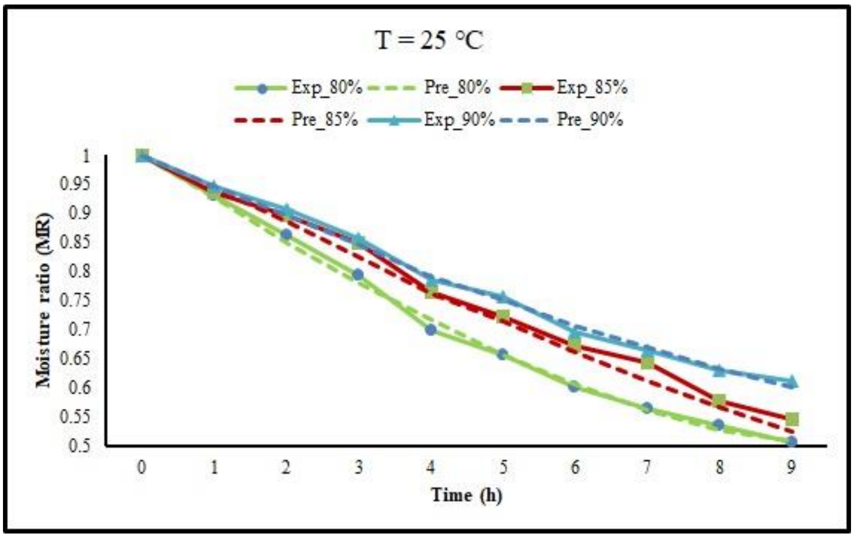

Fig. 4.9. *MR* vs Time (h) at T = 25 ºC

Figures 4.9-4.11 illustrate the withering characteristics for experimental and predicted results at constant temperature and variable humidity. The withering properties of the tea leaves at RHs 80%, 85% and 90% and at constant temperature of 25 ºC are represented in Fig.4.9. The MRs decrease to 0.51, 0.54 and 0.61 at the end of 9 h when withering took place at 80%, 85% and 90% RH respectively. The rate of withering 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. The average withering rates at these three levels of RH were accordingly 0.28, 0.27 and 0.26 g water/g dry solid-h.

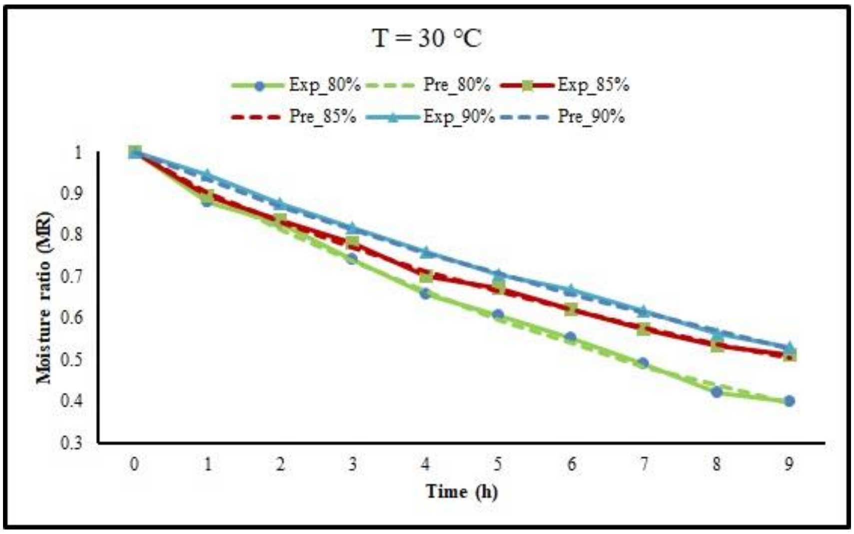

Fig. 4.10. *MR* vs Time (h) at T = 30 ºC

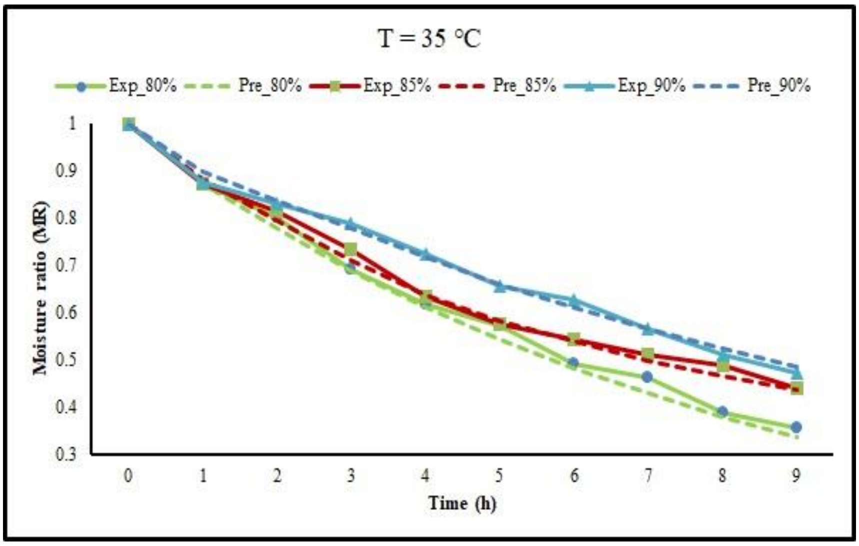

Fig. 4.11. *MR* vs Time (h) at T = 35 ºC

The withering characteristics at 30 ºC and 35 ºC with the same range of RHs are shown in Figures 4.10 and 4.11. Similar trends are observed in the withering rate for both the cases as in the previous condition. At constant temperatures of 30 ºC and 35 ºC, the moisture ratios came down to 0.4, 0.51, 0.53 and 0.36, 0.44, 0.47 respectively after 9 h of withering at the three levels of RH, i.e., 80%, 85% and 90%. This clearly shows that the moisture contents reduce with the rise in RH at constant temperature. However, rate of withering is faster when the temperature increases. The mean withering rates were 0.30, 0.29 and 0.28 g water/g dry solid-h at 30 ºC while they were 0.32, 0.30 and 0.29 g water/g dry solid-h at 35 ºC for the three considered RH levels. Botheju et al. and Ghodake et al. again reported similar withering properties [33,66].

From all the considered cases, it is evident that the withering rate is the highest at air temperature of 35 ºC with 80% humidity. The arrangement of 25 ºC-90% has the lowest withering rate. This is again in agreement with findings of [33,66]. A moderate rate of withering is seen at air temperature of 30 ºC and 90% humidity as compared to the rest.

Five standard drying models were used for fitting the experimental data obtained from the EC. Table 4.2 elaborately shows the withering curve parameters obtained from the regression analysis. The best fit was given by the Page model at 30 °C and 90% with R^2 value of 0.9989 and *RMSE* value of 0.0051. The adjusted R^2 value was found as 0.9988. The Lewis model gave the second best fit for the withering properties followed by the Henderson-Pabis model. The respective R^2 values for these two models were computed as 0.9988 and 0.9988 with corresponding adjusted R^2 values of 0.9988 and 0.9987. The *RMSE* values were respectively 0.0054 and 0.0057 for these two models. The second and third best models showed these fittings for the same conditions of 30 °C and 90%.

Previously, Page and Two-term models were obtained as the best suited drying models for drying tea leaves in Sri lanka [33]. Again, it was reported that the Henderson-Pabis model fitted the withering properties at lower temperature suitably whereas the Page model suited the higher temperature characteristics [66].

The mathematical models for the first, second and third best fits are as follows:

$$MR = e^{\left(-0.0636t^{1.0020}\right)} \tag{4.8}$$

$$MR = e^{(-0.0640t)} \tag{4.9}$$

$$MR = 1.001e^{(-0.0641t)} \tag{4.10}$$

Table-4.2. Drying curve parameters for the withering data

Model	**T (°C)**	**RH (%)**	**Constants**						***R²***	**Adj. *R²***	***RMSE***
			k	**k₁**	**k₂**	**A**	**B**	**N**			
Lewis	25	80	0.0806						0.9930	0.9930	0.0145
	30	80	0.1025						0.9965	0.9965	0.0122
	35	80	0.1160						0.9967	0.9967	0.0120
	25	85	0.0655						0.9926	0.9926	0.0134
	30	85	0.0800						0.9903	0.9903	0.0160
	35	85	0.0994						0.9801	0.9801	0.0263
	25	90	0.0569						0.9943	0.9943	0.0133
	30	**90**	**0.0640**						**0.9988**	**0.9988**	**0.0053**
	35	90	0.0827						0.9894	0.9894	0.0175
Henderson & Pabis	25	80	0.0809			1.0010			0.9930	0.9922	0.0154
	30	80	0.1019			0.9971			0.9965	0.9961	0.0126
	35	80	0.1145			0.9920			0.9970	0.9967	0.0123
	25	85	0.0675			1.0110			0.9939	0.9931	0.0139
	30	85	0.0761			0.9788			0.9945	0.9938	0.0138
	35	85	0.0942			0.9736			0.9847	0.9828	0.0244
	25	90	0.0578			1.0060			0.9948	0.9941	0.0135
	30	**90**	**0.0641**			**1.0010**			**0.9988**	**0.9987**	**0.0054**
	35	90	0.0795			0.9830			0.9919	0.9909	0.0162
Logarithmic	25	80	0.1182			0.7887	0.2244		0.9953	0.9940	0.0135
	30	80	0.0848			1.1240	-0.1324		0.9970	0.9961	0.0116

	35	80	0.1209			0.9612	0.0332		0.9971	0.9963	0.0111
	25	85	0.0350			1.7150	-0.7130		0.9956	0.9943	0.0127
	30	85	0.1188			0.7372	0.2543		0.9972	0.9964	0.0097
	35	85	0.1733			0.7020	0.2980		0.9941	0.9925	0.0162
	25	90	0.0646			0.9252	0.0823		0.9949	0.9934	0.0159
	30	**90**	**0.0646**			**0.9944**	**0.0067**		**0.9988**	**0.9985**	**0.0057**
	35	90	0.0632			1.1610	-0.1832		0.9923	0.9901	0.0169
Page	25	80	0.0844					0.9749	0.9933	0.9924	0.0151
	30	80	0.1010					1.0080	0.9965	0.9961	0.0126
	35	80	0.1232					0.9658	0.9972	0.9968	0.0121
	25	85	0.0544					1.1010	0.9959	0.9954	0.0106
	30	85	0.1009					0.8708	0.9978	0.9976	0.0080
	35	85	0.1313					0.8420	0.9920	0.9909	0.0178
	25	90	0.0536					1.0330	0.9947	0.9941	0.0136
	30	**90**	**0.0636**					**1.0020**	**0.9989**	**0.9988**	**0.0051**
	35	90	0.0927					0.9366	0.9910	0.9898	0.0171
Two-term	25	80		0.0866	-0.6736	1.0110	0.0001		0.9969	0.9953	0.0149
	30	80		0.1023	0.1004	0.9136	0.0845		0.9965	0.9948	0.0156
	35	80		0.1251	0.1150	-0.0392	1.0320		0.9970	0.9956	0.0143
	25	85		0.5347	0.0737	-0.0578	1.0550		0.9960	0.9941	0.0160
	30	85		3.2140	0.0728	0.0400	0.9610		0.9979	0.9968	0.0092
	35	85		-0.0850	0.1352	0.0824	0.9161		0.9943	0.9915	0.0172
	25	90		0.0460	0.0538	-0.5042	1.5100		0.9948	0.9922	0.0162
	30	**90**		**0.4696**	**0.0640**	**0.0011**	**1.0010**		**0.9988**	**0.9982**	**0.0062**
	35	90		0.0887	0.0786	0.1045	0.8792		0.9919	0.9878	0.0187

4.2.2 Comparison with open sun withering of tea leaves

Fig. 4.12 shows the withering curve of tea leaves when withered under the open sun. The ambient temperature and RH were 32 °C and 77% respectively. The moisture ratio comes down to 0.37 after 4 h of withering which is almost half of the time taken to wither under controlled conditions in the environmental chamber. The mean withering rate is 0.71 g water/g dry solid-h under direct sunlight. This increase in the withering rate may be described as the presence of dry air in the open environment.

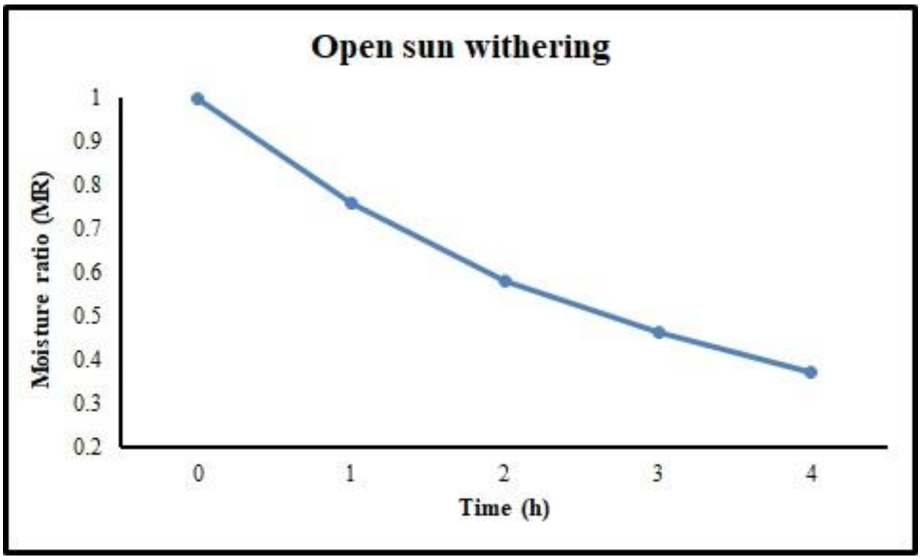

Fig. 4.12. Open sun withering of tea leaves

4.2.3 Specific energy consumption

The specific energy consumed at temperatures 25 °C, 30 °C and 35 °C with a constant RH of 90% is shown in Fig. 4.13.

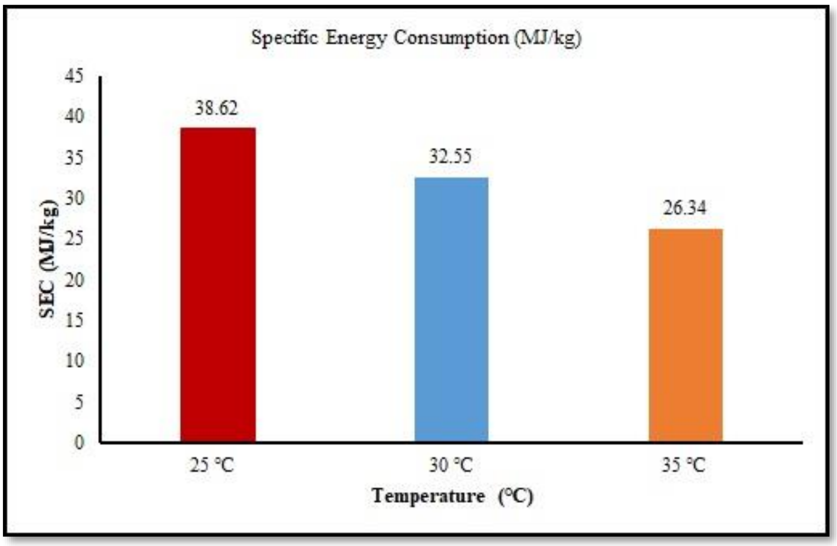

Fig. 4.13. Specific energy consumption at different temperatures

It is observed that the specific energy consumed during the process reduces with the rise in drying air temperature. The lowest value of SE was obtained as 26.34 MJ/kg at the temperature of 35 °C. This decline may be explained as the result of quicker withering or drying at higher temperature. Similar trends for SE were obtained while drying peppermint leaves [148].

4.2.4 Total Phenolic and Flavonoid contents

Standard procedures were used to determine the overall phenolic and flavonoid contents of the tea-leaf samples withered at temperatures of 25 °C, 30 °C and 35 °C. Figures 4.14 and 4.15 represent respectively the TPCs and TFCs at all the withering temperatures.

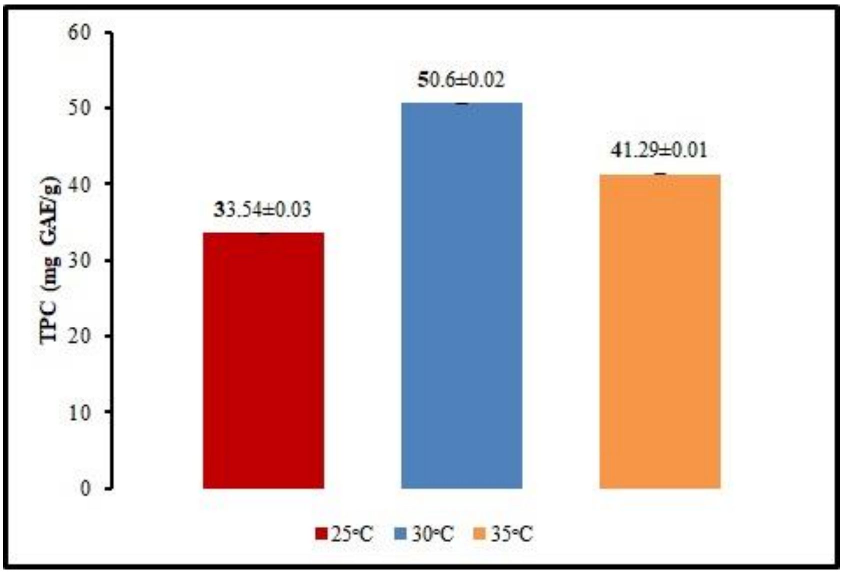

Fig. 4.14. Total Phenolic Content

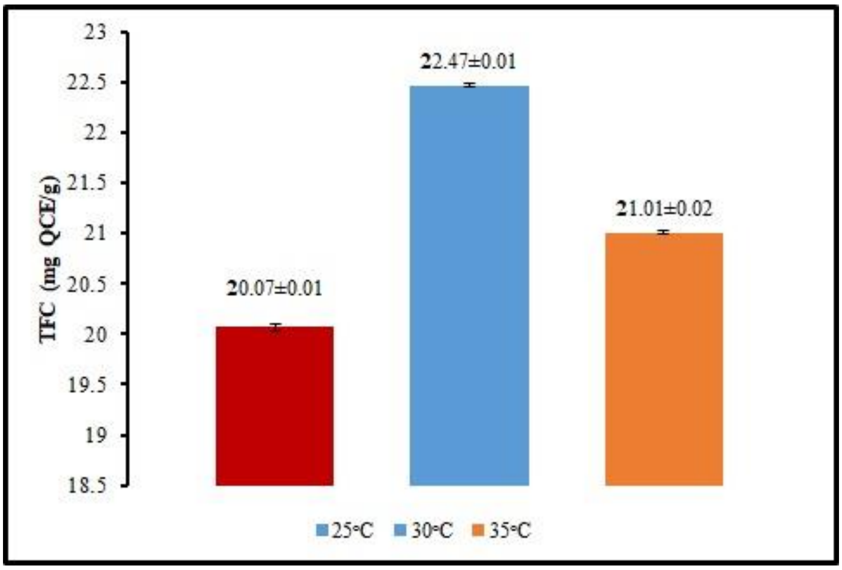

Fig. 4.15. Total Flavonoid Content

It is clearly seen that the maximum TPC and TFC are found at 30 °C as (50.6 ± 0.02) mg GAE/g and (22.47 ± 0.01) mg QCE/g respectively. The sudden spike in tea's polyphenol and flavonoid content at 30 °C is a notable phenomenon caused by increased enzymatic activity, reduced microbial growth, and the interplay between temperature and moisture content promoting their accumulation within this specific temperature range. [212]. The TPC for unfermented tea samples of Assam was estimated as (89.20 ± 1.05) mg GAE/g and TFC as (77.43 ± 0.67) mg QCE/g [100]. The polyphenol content in green tea was reported to be higher than that in black tea for Indian tea samples [92].

4.3 Conclusions

The chapter presents the low temperature drying or withering properties of fresh tea leaves determined from the experiments conducted in an environmental chamber. The following are the conclusions which may be drawn from the study-

- The withering of fresh tea leaves occurred in a combination of both constant-rate as well as falling-rate drying period.
- The withering characteristics are affected by drying air temperature and humidity.
- The highest withering rate of 0.32 g water/g dry solid-h was observed at 35 °C-80% combination while the least rate was found as 0.26 g water/g dry solid-h at 25 °C-90%. A moderate wilting rate of 0.28 g water/g dry solid-h was observed in 30 °C-90%.
- The first, second and third best fits were given by the Page, Lewis and Henderson and Pabis models respectively at drying air temperature 30 °C and 90% RH.
- The specific energy consumed at drying air temperatures 25, 30 and 35 °C for a constant RH of 90% were computed as 38.62, 32.55 and 26.34 MJ/kg respectively.
- The maximum TPC and TFC were estimated to be (50.60 ± 0.02) mg GAE/g and (22.47 ± 0.01) mg QCE/g respectively at 30 °C.

As a further step, the withering or low temperature drying features of the fresh tea leaves may be determined under similar environmental conditions by developing an experimental set-up and application of solar thermal energy. The next chapter elaborates such an experimental study done on the intervention of solar thermal energy to wither tea leaves.