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Drying kinetics and mathematical modelling of *Ailanthus excelsa* leaves

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Abstract
Ailanthus excelsa (*Simaroubaceae*) is a plant used in Indian system of medicine by providing a range of potential health beneficiary activities. The present investigation was carried out to study the drying kinetics of *Ailanthus excelsa* leaves using different drying methods *viz.*, shade drying, tray drying (40±1 °C) and dehumidified air drying (40±1 °C and 15±1% RH). Irrespective of drying methods for *Ailanthus excelsa* leaves, a reduction in moisture content was observed. The *Ailanthus excelsa* leaves took 28 h, 9 h and 6 h to dry under shade drying, tray drying and dehumidified drying respectively, from 238.64% (d.b.) till the desired moisture content (less than 5%) was attained. Six thin layer models were selected for describing the drying process of *Ailanthus excelsa* leaves. Among the different drying methods, the Page model was best fitted with the samples dried under dehumidified air dryer with the highest R^2 value of 0.9996.

Keywords: *Ailanthus excelsa*, drying, modeling, page model

1. Introduction

Over the past decade, herbal and ayurvedic drugs have become a subject of world importance, with both medicinal and economical implications. A regular and widespread use of herbs throughout the world has increased serious concerns over their quality, safety and efficacy [Kumar *et al*., 2010] [8]. In developing countries, the traditional medicines having wide historical background are utilized for primary health care and in those plants only few have been explored for their potential as drug. In India, the World Health Organization (WHO) has exhibited 2500 species with medicinal purpose and from that 150 species were used on larger scale [Umashankar and Shruti, 2011] [17].

Ailanthus excelsa (*Simaroubaceae*) is a plant used in Indian system of medicine for a variety of purposes. It is commonly known as "Mahanimba" due to its resemblance with neem tree (*Azadirachta indica*). The term *Ailanthus* is derived from *ailanto* which means "Tree of Heaven" and is the name for one of the species in the Moluccas, while in Latin *excelsa* means "tall" [Lavhale and Mishra, 2007] [9]. *Ailanthus* is distributed in Indo-Malaya, China, Japan and Australia. It is called as *Ailanthus excelsa* in India (leaflets coarsely toothed and filaments shorter than anthers) and *Ailanthus malbarica* in Indo-China (leaflets entire and filaments larger than anthers) [Kirtikar and Basu, 1995] [7].

Ailanthus excelsa is one of the species that has a major source of natural secondary metabolites and acts as an active component by providing a range of potential health beneficiary activities [Kalaskar *et al*, 2019] [6]. The various parts (leaves, bark, stem) of *Ailanthus excelsa* tree have antitumor, antiviral, antimalarial, antileukemic, antifeedent, hepatoprotective, antiasthmatic, antifertility and antibacterial activity [Vaibhav *et al*, 2014] [19]. *Ailanthus excelsa* is used in treatment of skin eruption and for the cure of wounds and the extract showed potent antibacterial and antifungal activities [Malviya and Dwivedi, 2019] [10].

Drying is the most common and fundamental method for post-harvest preservation as it allows for the quick conservation of the qualities of the plant material in an uncomplicated manner.

Due to high investment and energy costs, drying is also a large expense in medicinal plant production. Drug quality and consequently earnings are significantly influenced by the drying regime. Conventionally, low drying temperatures between 30 and 50 °C are recommended to protect sensitive active ingredients [Bogers *et al*., 2006] [3]. The method and temperature used for drying generally have a considerable impact on the quality and stability of bioactive compounds [Pham *et al*., 2015] [12]. Hence, drying of *Ailanthus excelsa* leaves is an important post-harvest treatment to reduce the moisture content and to increase the shelf-life. Since the bioactive compounds of *Ailanthus excelsa* are heat sensitive materials, drying under low temperature could be beneficial. The literature is almost silent on drying of *Ailanthus excelsa* leaves. By keeping this in view, the present work was carried out to study the drying kinetics and mathematical modelling of *Ailanthus excelsa* leaves.

2. Materials and Methods

2.1 Drying of *Ailanthus excelsa* leaves

The fresh *Ailanthus excelsa* stems of uniform colour and freshness were harvested from the tree. The fresh leaves of *Ailanthus excelsa* were separated from the stems and used for the drying. The *Ailanthus excelsa* tree and fresh leaves of it are depicted in Plate 1 and 2, respectively. The fresh leaves (1.0 kg for each treatment) were subjected to drying using three different drying methods *viz*., shade drying, tray drying (40±1 °C) and dehumidified air drying (40±1 °C and 15±1% RH) till the desired moisture content (less than 5%).

Plate 1: *Ailanthus excelsa* tree

Plate 2: Fresh *Ailanthus excelsa* leaves

The drying kinetics of *Ailanthus excelsa* leaves was studied by analysing the reduction in moisture content, drying rate and moisture ratio during drying period. The weight of dried sample was taken at an interval of 30 min. until completion of drying process of less than 5% (d.b.) moisture content [Ali *et al*., 2014] [1] in shade drying, tray drying and dehumidified drying. Reduction in moisture content v/s drying time curves were plotted for shade drying, tray drying and dehumidified air drying.

2.2 Moisture content

The moisture content of fresh leaves of *Ailanthus excelsa* was determined by following AOAC (2005, Method No. 945.43) using hot air oven (Kemi, KOS.6FD, Perumbavoor, India) [AOAC, 2005] [2]. Three grams of *Ailanthus excelsa* leaves were kept in a pre-dried moisture box. The mass of the sample was recorded as W_1. The box was placed in hot air oven maintained at 100±2 °C for 8 h. After drying, the box was kept in the desiccator and then weighed. The mass of the dried sample was recorded as W_2. The moisture content of the sample was calculated by using the following equation [Wankhade *et al*., 2012] [20],

$$\text{Moisture content (\% d.b.)} = \frac{W_1 - W_2}{W_2} \times 100$$

Where,
W_1 = Initial weight of sample, g
W_2 = Final weight of sample, g

2.3 Drying rate

The drying rate of *Ailanthus excelsa* leaves under different drying methods was calculated from the following equation [Wankhade *et al*., 2012] [20],

$$\text{Drying rate} = \frac{M_i - M_d}{t}$$

Where,
M_i = Mass of sample before drying, g
M_d = Mass of sample after drying, g
t = Drying time, min.

2.4 Moisture ratio

The moisture ratio of *Ailanthus excelsa* leaves for different drying methods was calculated by using the following equation [Wankhade *et al*., 2012] [20],

$$\text{Moisture ratio} = \frac{M_t - M_e}{M_o - M_e}$$

Where,
M_t = Moisture content at a given time, t (%, d.b.)
M_e = Equilibrium moisture content (%, d.b.)
M_o = Initial moisture content (%, d.b.)

2.5 Mathematical modelling for drying of *Ailanthus excelsa* leaves in different drying methods

The drying models *viz*., Newton, Page, Modified Page, Henderson and Pabis, Modified Handerson and Pabis, and Logarithmic were selected for mathematical modelling of drying of *Ailanthus excelsa* leaves [Pin *et al*., 2009] [13] and are as follows,

- Newton: MR = exp(-kt)
- Page: MR = exp(-kt^n)
- Modified Page: MR = exp(-kt)^n
- Henderson and Pabis: MR = a*exp(-kt)
- Modified Henderson and Pabis: MR = a*exp(-kt)+b*exp(-gt)+c*exp(-ht)
- Logarithmic: MR = a*exp(-kt)+c

Where,
MR = Moisture ratio
t = Drying time, h
k, n, a, b, c, g and h = Drying constants

The parameters of all the models were estimated by using 'MATLAB' version 7.0 software. The best fit of the proposed models on the experimental data was evaluated using linear regression analysis using curve fitting tool in MATLAB. The statistical parameters, standard square error (SSE) and root mean square error (RMSE) were calculated by employing the following equations.

$$P = \frac{100}{N}\sum_{i=0}^{N}\frac{MR_o - MR_P}{MR_o}$$

$$\mathrm{RMSE} = \sqrt{\frac{\sum_{i=0}^{N}\left(\mathrm{MR_o} - \mathrm{MR_p}\right)^2}{\mathrm{df}}}$$

$$\mathrm{SSE} = \frac{1}{\mathrm{N}}\sum_{i=1}^{N}(\mathrm{MR_o} - \mathrm{MR_p})^2$$

Where,
MR_o = Observed moisture ratio
MR_P = Predicted moisture ratio
df = Degrees of freedom
N = Number of data points

2.6 Milling of dried *Ailanthus excelsa* leaves
The grinding equipment used in the investigation were normal pulverizer, water cooled pulverizer and liquid nitrogen (LN_2) cooled pulverizer.

2.6.1 Normal pulverizer
Dried *Ailanthus excelsa* leaves were ground in impact type normal pulverizer (Sriram Machinery Works; SRM-108, Andalpuram, Tamil Nadu, India) of 25 kg.h^{-1} capacity for the ambient grinding pr°C ess. In this normal pulverizer, swinging hammer heads were attached to a rotor that rotated at high speed inside a hardened casing. Dried leaves were put into the hopper of the normal pulverizer and were ground with the help of rotor. The fine ground sample of *Ailanthus excelsa* powder was collected from the outlet of the pulverizer.

2.6.2 Water cooled pulverizer
Water cooled pulverizer (Sriram Machinery Works; SRM-108, Andalpuram, Tamil Nadu, India) was used for grinding of dried *Ailanthus excelsa* leaves (Sankalpa *et al*., 2016) [15]. The capacity of the water cooled pulverizer was 25 kg.h^{-1}. The flow of material from the feeding hopper was regulated by means of a vertical adjustable slide to suit the load. Dried *Ailanthus excelsa* leaves were fed through the hopper of the grinder and were ground with the help of rotor inside the grinder. Temperature during grinding was reduced in the grinder with the help of water circulation through it. After grinding, the material was passed through the screen fitted inside the lower discharge end of the grinding chamber.

2.6.3 Liquid nitrogen cooled pulverizer
The dried *Ailanthus excelsa* leaves were ground using liquid nitrogen cooled pulverizer. The low temperature grinding was done by passing the liquid nitrogen (LN_2) at -196 °C during grinding (M/s. Spectra Cryogenic Systems, Kota, Rajasthan) (Sankalpa *et al*., 2016) [15]. From the hopper, the dried *Ailanthus excelsa* leaves to be ground were metered at a specific rate into the grinding mill. The liquid nitrogen was injected into the feeder which combined with the samples leading to cooling and embrittling. The cryo material was transported along with the cold nitrogen to the grinding mill where it was pulverized. The milled powder was passed through the screen provided at the bottom of the grinding chamber.

3. Results and Discussion
The drying kinetics of *Ailanthus excelsa* leaves was studied by analysing the reduction in moisture content, drying rate and moisture ratio during drying period. Irrespective of drying methods for *Ailanthus excelsa* leaves, an increasing trend was observed in the reduction of moisture content in the beginning of the drying process. As the drying process proceeded, the loss of moisture in *Ailanthus excelsa* leaves decreased with drying time due to the evaporation of water with respect to time. The moisture content of the fresh leaves of *Ailanthus excelsa* was found to be 238.64% (d.b.). The results of the effect of drying methods on moisture content, drying rate and moisture ratio is discussed under the following headings.

3.1 Effect of drying methods on moisture content of *Ailanthus excelsa*
The results of effect of drying methods *viz.* shade drying, tray drying (40±1 °C) and dehumidified air drying (40±1 °C and 15±1% RH) on moisture content of *Ailanthus excelsa* leaves are presented and discussed.

3.1.1 Shade drying
In shade drying, the moisture content of *Ailanthus excelsa* leaves was decreased from 238.64% (d.b.) to 4.28% (d.b.) in 28.00 h. After 28.00 h, constant moisture content was obtained for another one hour of drying. Hence, the drying time was considered as 28.00 h. The average temperature and humidity in shade drying was found to be 29.14 °C and 35.45%, respectively.

3.1.2 Tray drying
In tray drying, the moisture content of *Ailanthus excelsa* leaves was decreased from 238.64% (d.b.) to 4.19% (d.b.) in 9.00 h. After 9.00 h, constant moisture content was obtained for another one hour of drying. Hence, the drying time was considered as 9.00 h. The average temperature and humidity in tray dryer was found to be 40 °C and 31.07%, respectively.

3.1.3 Dehumidified air drying

In dehumidified air drying, the moisture content of *Ailanthus excelsa* was decreased from 238.64% (d.b.) to 4.22% (d.b.) in 6 h at 40±1 °C and 15±1% RH. After 6.00 h, constant moisture content was obtained for another one hour of drying. Hence, the drying time was considered as 6.00 h.

The effect of drying methods on moisture content of *Ailanthus excelsa* leaves is depicted in Fig. 1. It could be observed that shade drying took highest drying time followed by tray drying and dehumidified air drying methods. This might be due to the lower drying air temperature prevailed during the drying process and more relative humidity fluctuations in the atmosphere in case of shade drying, similar result was recorded for shade drying of *Isparta* rose flowers [Boyar *et al*., 2013] [4] and shade drying of green tea leaves (36 h) under natural air flow at 25 °C temperature [Roshanak *et al*., 2016] [14]. Less time of drying was observed in tray drying as compared to shade drying because of change in relative humidity, similar result was observed for *purslane leaves* (420±30 min.) at 50 °C [Youssef and Mokhtar, 2014] [21]. Shorter drying period in dehumidified air drying might be due to less relative humidity of air maintained inside the drying chamber, similar results were recorded for drying of whole leaf aloe vera which required 11 h at 55±1 °C and 18±1% RH [19] and sweet orange peel in 3.50 h at 45 °C and 15% RH [18] in dehumidified air dryer.

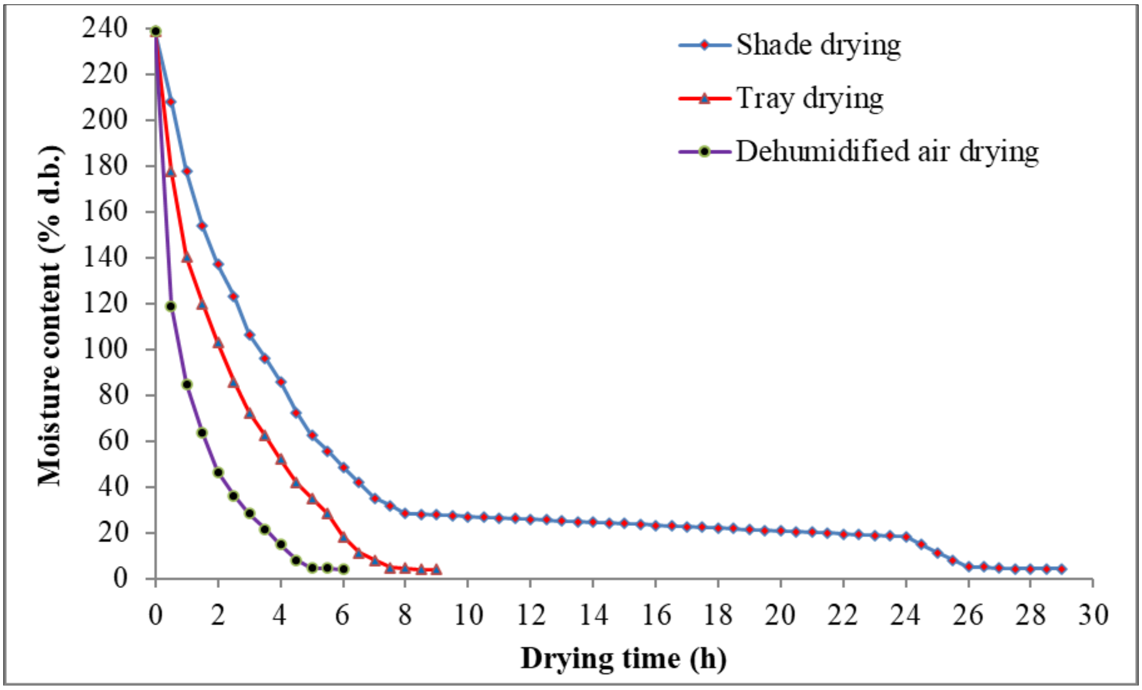

Fig 1: Effect of drying methods on moisture content of *Ailanthus excelsa* leaves

3.2 Effect of drying methods on drying rate of *Ailanthus excelsa* leaves

The drying rate of fresh *Ailanthus excelsa* leaves dried in shade drying ranged from 0.0034 g.g^{-1} dm. min^{-1} to 0.0001 g.g^{-1} dm. min^{-1}. In case of tray drying, drying rate ranged from 0.0069 g.g^{-1} dm. min^{-1} to 0.0001 g.g^{-1} dm. min^{-1}. Here the temperature varied according to the climatic condition. The drying rate of fresh *Ailanthus excelsa* leaves dried under dehumidified air dryer ranged from 0.0135 g.g^{-1} dm. min^{-1} to 0.0001 g.g^{-1} dm. min^{-1}. In all the drying method, the drying rate was mainly dependent on drying temperature.

From the Fig. 2, it is noticed that the drying rate was significantly influenced by the drying methods. The lowest drying rate was noticed in case of shade drying followed by tray drying and dehumidified drying. This might be due to the lower drying air temperature prevailed during the drying process and more relative humidity fluctuations in the atmosphere in case of shade drying, similar result were recorded for shade drying of Isparta rose flowers [Boyar *et al*., 2013] [4]. The tray drying of *Ailanthus excelsa* leaves took lesser time in relating to shade drying this might be due to the higher temperature in the tray drying and less relative humidity might have absorbed larger quantity of moisture in lesser time resulting in higher drying rate. Whereas, shorter drying period in dehumidified air drying might be due to less relative humidity of air maintained inside the drying chamber, similar result were recorded for drying of whole leaf aloe vera in dehumidified air dryer [Pattali *et al*., 2015] [11].

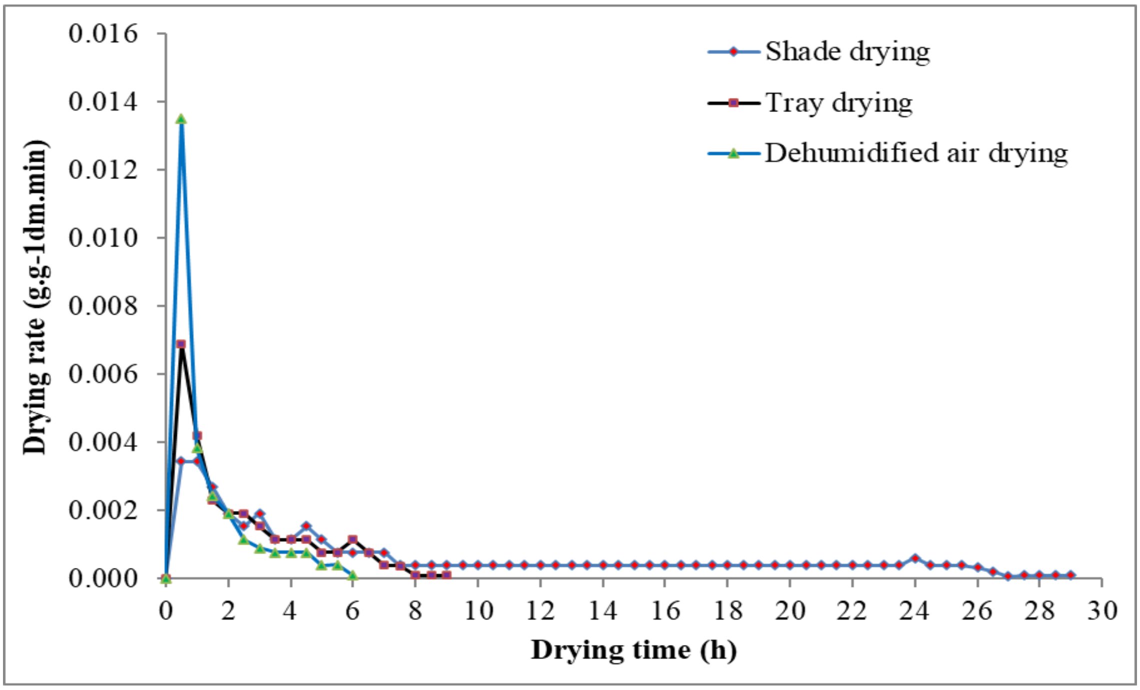

Fig 2: Effect of drying methods on drying rate of *Ailanthus excelsa* leaves

3.3 Effect of drying methods on moisture ratio of *Ailanthus excelsa* leaves

The variations in moisture ratio of *Ailanthus excelsa* leaves dried under shade drying, tray drying and dehumidified air drying methods are depicted in Fig. 3. From the figure, it could be observed that the moisture ratio was significantly influenced by the drying methods. From the figure, it is noticed that the moisture ratio of *Ailanthus excelsa* dried in shade drying, tray drying and dehumidified air drying ranged from 0.872, 0.744 and 0.498, respectively in the initial stage of drying to 0.018 at the final stage of drying.

The lowest moisture ratio was noticed in case of shade drying followed by tray drying and dehumidified drying methods. This might be due to the lower drying air temperature prevailed during the drying process and more relative humidity fluctuations in the atmosphere in case of shade drying, similar result were recorded for tray drying and shade drying of drumstick leaves [Satwase *et al*., 2013] [16] and shade drying of Isparta rose flowers [Boyar *et al*., 2013] [4] and tray drying for purslane leaves [Youssef and Mokhtar, 2014] [21]. Shorter drying period in dehumidified air drying might be due to less relative humidity of air maintained inside the drying chamber, similar result were recorded for drying of whole leaf aloe vera [Pattali *et al*., 2015] [11] and sweet orange peel [Sankalpa *et al*., 2016] [15] in dehumidified air dryer.

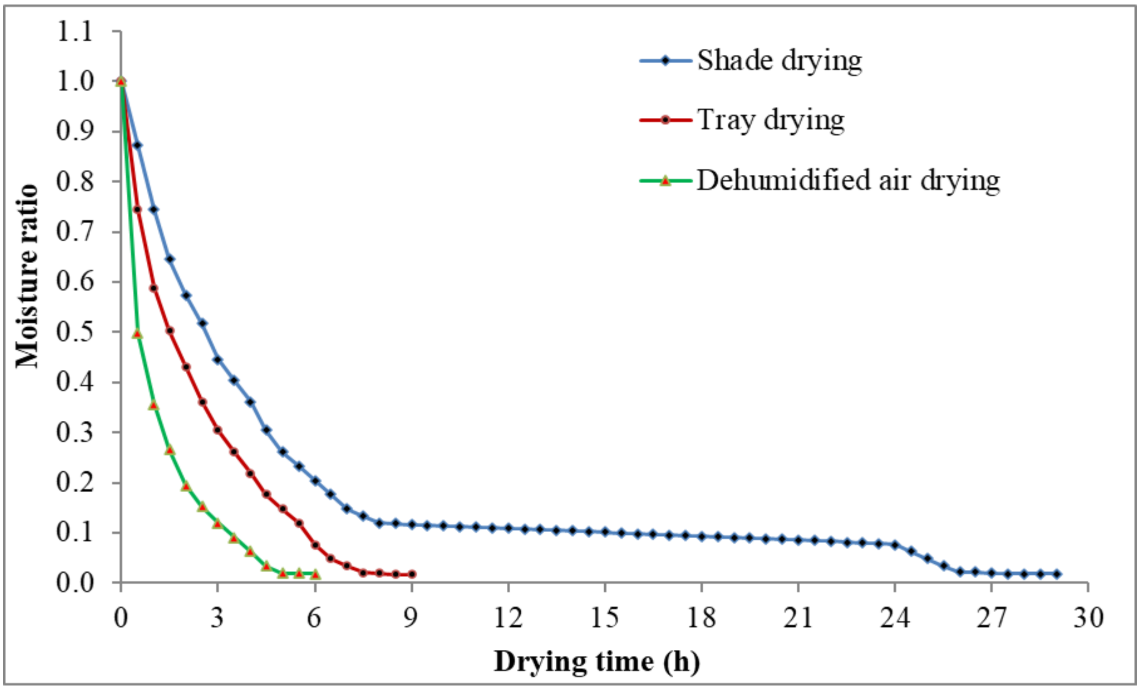

Fig 3: Effect of drying methods on moisture ratio of *Ailanthus excelsa* leaves

3.4 Mathematical modelling for drying of *Ailanthus excelsa* leaves in different drying methods

Different drying models namely, Newton, Page, Modified Page, Henderson and Pabis, Modified Henderson and Pabis and Logarithmic models were selected and fitted to the drying data based on their ability to model the drying phenomenon of fresh *Ailanthus excelsa* leaves. The drying constants and the statistical validity of models was evaluated in MATLAB 2007 software and compared by means of coefficient of determination (R^2). In order to select the best model, this factor was used as the main selection criteria, but also the goodness of the fit was determined using additionally using other statistical parameters such as reduced sum square error (SSE), adjusted R^2 and root mean square error (RMSE) values. For best fit, R^2 value should be higher and SSE and RMSE values should be lower. The constants of various drying models for different drying methods are shown in Table 1 and 2. The tables also present the estimated values of statistical parameters of various models for different drying methods.

Table 1: Effect of drying methods on drying constants of drying models used for *Ailanthus excelsa* leaves

Sl. No.	Model	Constant	Drying method		
			SD	TD	DD
1	Newton	k	0.2743	0.4212	0.9292
2	Page	k	0.2618	0.4738	1.0300
		n	1.0310	0.8954	0.6801
3	Modified Page	k	1.1400	0.6763	0.9206
		n	0.2406	0.6227	1.009
4	Henderson and Pabis	a	1.001	0.9519	0.932
		k	0.2745	0.4006	0.8585
5	Modified Henderson and Pabis	a	10.2000	1.1450	0.675
		b	-9.2870	12.8400	-2.094
		c	0.1090	-13.030	0.4213
		g	0.2324	0.5820	0.3583
		h	0.8357	0.5776	4.327
		k	0.2346	0.3983	0.3908
6	Logarithmic	a	1.0250	0.9608	0.9136
		c	-0.0441	-0.0167	0.04523
		k	0.2421	0.3798	1.0450

Table 2: Effect of drying methods on estimated values of statistical parameters for different drying models used for drying of *Ailanthus excelsa* leaves

Sl. No.	Parameter	Drying method	Models					
			Newton	Page	Modified Page	Henderson and Pabis	Modified Henderson and Pabis	Logarithmic
1	R^2	SD	0.9960	0.9963	0.9960	0.9960	0.9947	0.9984
		TD	0.9887	0.9925	0.9887	0.9916	0.9920	0.9920
		DD	0.9656	0.9996	0.9656	0.9711	0.9974	0.9801
2	SSE	SD	0.008141	0.007539	0.008141	0.00814	0.01088	0.003268
		TD	0.01581	0.01038	0.01581	0.01172	0.01113	0.01109
		DD	0.03164	0.000369	0.03164	0.02659	0.0024	0.01828
3	Adjusted R^2	SD	0.9960	0.9961	0.9958	0.9958	0.9933	0.9983
		TD	0.98887	0.9921	0.9888	0.9911	0.992	0.991
		DD	0.9656	0.9993	0.9624	0.9684	0.9971	0.9761
4	RMSE	SD	0.01805	0.01772	0.01842	0.01842	0.02332	0.01192
		TD	0.02964	0.02471	0.03049	0.02626	0.02926	0.02633
		DD	0.05135	0.007265	0.05363	0.04917	0.01485	0.04276

The values of moisture ratio were determined for all drying methods such as shade drying, tray drying and dehumidified air drying. Among the different drying methods, the Page model was best fitted with the samples dried under dehumidified air dryer with the highest R^2 value of 0.9996. The variations of experimental and predicted moisture ratio values with drying time are depicted in Fig. 4.

This variation in R^2 might be due to the relative humidity and temperature of drying air influenced 'k' and 'n' values in the Page model. The result obtained was in good agreement with the results reported by [Gunhan *et al*., 2005] [5] for drying of bay leaves in laboratory dryer with R^2 value of 0.9993, [Upadhyaya *et al*., 2012] [18] for hot air drying of spinach leaves with R^2 value of 0.9911, [Ali *et al*., 2014] [1] for hot air drying of moringa leaves with R^2 value of 0.998 and [Pattali *et al*., 2015] [11] for dehumidified air drying of whole leaf aloe vera powder with R^2 value of 0.9764.

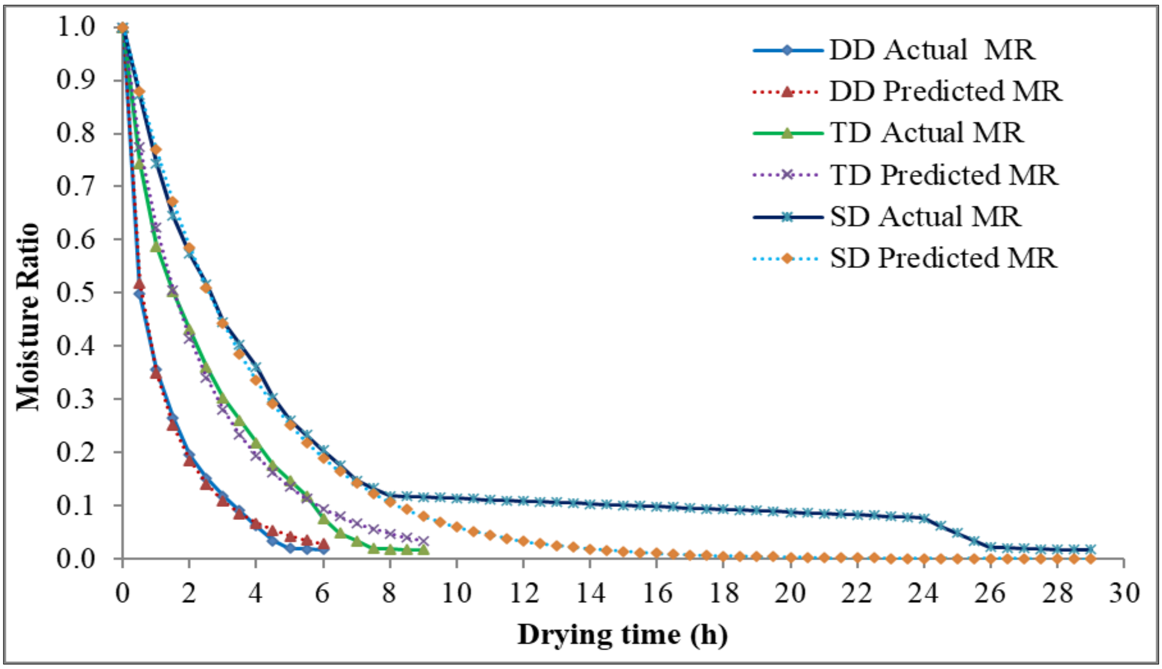

Fig 4: Effect of drying methods on experimental and predicted (Page model) moisture ratio of *Ailanthus excelsa* leaves

4. Conclusion

The effect of drying methods on *Ailanthus excelsa* leaves was investigated for drying kinetics. The reduction in moisture content of *Ailanthus excelsa* took less drying time of 6.00 h in dehumidified drying to dry the sample from an initial moisture content of 238.64% (d.b.) to final moisture content of 4.22% (d.b.) as compared to shade drying and tray drying. The drying rate was higher in the beginning of the drying processes and gradually reduced through the end of the drying process. Page model gave better fit to the experimental data with higher R^2 value of 0.9996 and lowest root mean square error (RMSE) and sum of square error (SSE) values of 0.007265 and 0.000369, respectively over the other two models.

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