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Drying behaviour of *Andrographis paniculata* in vacuum drying

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Abstract

Hempedu bumi (*Andrographis paniculata*, AP), also known as the king of bitters, is an herb commonly found in Asian communities for medicinal usage. The drying behaviour of AP at temperatures of 40, 50, and 60°C with vacuum pressures of 10 and 30 kPa was investigated in this study. The data were then fitted with semi-theoretical and theoretical thin-layer drying models. The results reveal that the drying time is significantly ($p<0.05$) affected by temperature and pressure. A two-term, thin-layer model was determined as the most suitable model to fit the drying behaviour of AP. The effective diffusivity and active energy for moisture diffusion were 10-13 m^2/s and 33.4 kJ/mol, respectively.

Introduction

Herbs are gaining popularity worldwide for their medicinal use, as "going natural" is on an upward trend. It is believed that herbs, which are natural substances, are safer than synthetic products. Hussin (2001) estimates that approximately 80% of the world population uses some form of herbal medicine. It has been reported that Malaysians spent approximately RM2.0 billion on herbal products in 1997(Hussin, 2001). These data demonstrate that herbs are widely accepted and have great market potential.

Hempedu bumi (*Andrographis paniculata* (AP)) is an herb from the family of Acanthaceae, which grows in India, China, and southeast Asia. It is well known as the 'king of bitters'. AP has shown beneficial effects on hyperdipsia, burning sensations, wounds, ulcers, chronic fever, malarial and intermittent fever, inflammations, diarrhoea, cough, and skin diseases (Warrier *et al*., 1993); anti-angiogenic activity (Sheeja *et al*., 2007); hepatoprotective activity and anti-platelet activity (Nagalekshmi *et al*., 2011); and anti-diabetic potential (Reyes *et al*., 2006). Kumar *et al*. (2004) suggested that AP contains anticancer and immunostimulatory compounds. In Malaysia, AP has been widely used for diabetes and hypertension. Andrographolide is the major constituent extracted from the plant and is responsible for the bitter taste and functional properties of the plant (Parasher *et al*., 2011). Andrographolide is a diterpene lactone and is insoluble in water. The maximum level of andrographolide is found in the leaves of the plant (Kurian *et al*., 2007).

Herbs have a moisture content as high as 80%. Removal of the moisture from herbs is necessary to prevent microbial contamination and to allow for further processing. Drying is one of the most ancient, but effective, preservative methods that can be used to extend an herb's shelf life. Conventional drying methods, such as sun drying, have been used for many years, but the drying conditions are uncontrollable and the process is time-consuming. Oven drying is also commonly used in the dehydration process; however, this method results in a loss of flavour, colour, and nutrients due to the high temperature used (Sharma and Prasad, 2001). To prevent the loss of sensory and nutritive qualities of herbs, different types of advanced drying techniques have been developed. Freeze drying is able to maintain many of the bioactive components in herbs, but the drying rate is slow (Krokida and Philippopoulos, 2006). Microwave drying can retain much of the quality of the herbs and is efficient (Zhou *et al*., 2011). However, a disadvantage of microwave heating is non-uniformity within the drying cavity, which can lead to scorching of the drying material despite the presence of a turn-table (Mermelstein, 1998).

Vacuum drying is a method of removing water from moist materials at low pressures. The vacuum expands the air and water vapour in food products and creates a frothy structure. This method offers advantages such as a higher drying rate, a lower drying temperature, an oxygen-deficient processing environment, and higher quality products compared with those derived from conventional atmospheric drying. Ah-Hen *et al*. (2013) report that vacuum

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drying manages to shorten the drying time to 800 min rather than 1500 min at atmospheric drying. As this method uses low temperatures, it is suitable for heat-sensitive materials including fruits, vegetables, and herbs. Studies of vacuum drying of eggplants, carrots, apples, and pumpkins have been conducted (Arevalo-Pinedo *et al.*, 2004; Arévalo-Pinedo and Murr, 2006; Wu *et al.*, 2007). However, there is a lack of reporting on the vacuum-drying kinetics of AP. Therefore, the aim of this study was to fill this gap in knowledge. This information is very useful for designing a dryer and better understanding the drying behaviour of AP.

Materials and Methods

Materials and equipment

Fresh Andrographis paniculata was obtained from MARDI, Serdang, and collected randomly to ensure that the sample was homogenised. The drying equipment used in this study consisted of a vacuum oven (WTC Binder VDV53, Germany) and a double-stage high-vacuum pump (Javac, Australia).

Drying experiment

The experiments were conducted with 3 drying temperatures, 40, 50, and 60°C, at absolute pressure of 10 and 30 kPa, with 3 replicates for each treatment. The vacuum-drying oven was preheated to the desired temperature. The leaves and stems of the herb were cut into small pieces of approximately 1 cm in length. Then, 30 g of sample was spread in a single layer on an aluminium tray and put in the drying chamber under the selected drying conditions. The vacuum oven's door was opened and the sample weight was recorded every 15 minutes during the initial drying period. When a decrease in weight was not obvious, the drying period was extended to 30 minutes or 1 hour as necessary. The samples were dried until their moisture content reached less than 0.11 kg water/ kg solid.

Mathematical modelling

The moisture ratio (MR) and drying rate (DR) of AP during the thin-layer drying were determined as

$$MR = \frac{M_t - M_e}{M_o - M_e} \quad (1)$$

$$DR = \frac{M_{t+dt} - M_t}{dt} \quad (2)$$

where M_t is the moisture content of the sample at any time, Me is the equilibrium moisture content, M_o is the initial moisture content, and t is the drying time. However, the MR was simplified to M_t/M_o (Kayisoglu and Ertekin, 2011; Mitra *et al.*, 2011)because M_e is relatively smaller than M_t and M_o.

Table 1. Selected semi-theoretical and theoretical models for thin-layer drying

Model name	Model	Reference
Newton	$MR = \exp(-kt)$	(O'Callaghan *et al.*, 1971)
Page	$MR = \exp(-kt^n)$	
Henderson and Pabis	$MR = a\exp(-kt)$	(Henderson and Pabis, 1961)
Logarithmic	$MR = a\exp(-kt) + c$	(Lee and Kim, 2009)
Two-term	$MR = a\exp(-kt) + b\exp(-k_o t)$	(Henderson, 1974)
Two-term exponential	$MR = a\exp(-kt) + (1-a)\exp(-kat)$	(Sharaf-Eldeen *et al.*, 1980)
Fick's second law	$MR = \frac{6}{\pi^2}\sum_{i=1}^{i=\infty}\frac{1}{i^2}\exp\left[-i^2\pi^2\left(\frac{D_{eff}t}{L^2}\right)\right]$	(Crank, 1975)

a, b,c= coefficient, k, k_o= drying constant, *L*= half thickness of slam (m), D_{eff}= effective diffusivity (m^2/s)

Several semi-theoretical and theoretical thin-layer drying models (Table 1) were tested to select the best model for describing the drying kinetics of AP under the vacuum conditions. By using SOLVER, an optimisation tool in Microsoft Excel 2007, the parameters of the different models were determined. The lowest sum of the square error (SSE) difference between the experimental and calculated moisture ratio was the criterion for choosing the best model to describe the drying curve. In addition, the root mean square error (RMSE) was used to determine the quality of the fit.

$$RMSE = \left[\frac{1}{N}\sum_{i=1}^{N}(MR_{cal,i} - MR_{exp,i})^2\right]^{\frac{1}{2}} \quad (3)$$

where $MR_{cal,i}$ is the ith calculated moisture ratio and $MR_{exp,i}$ is the *i*th experimental moisture ratio.

The effective diffusivity was related to temperature by Arrhenius' equation:

$$D_{eff} = D_{\infty} e^{\left(-\frac{E_a}{RT}\right)} \quad (4)$$

where D_{∞} is the pre-exponential factor of the Arrhenius equation, E_a is the activation energy (kJ/mol), R is the universal gas constant (8.314 J/mol K), and T is the absolute temperature (K). The activation energy was calculated by plotting the *ln* D_{eff} versus *1/T*.

Moisture content determination

The moisture content of the samples was determined using a moisture analyser (XM 120, Precisa Instruments Ltd, Switzerland). For each moisture content determination, a sample of approximately 1.0 g was placed in the sample pan of the apparatus. This method was validated with the oven method by

Table 2. Drying time required to achieve 0.11 kg water/ kg solid for various drying conditions

Pressure (kPa)	Drying time (0.11 kg water/ kg solid) (minutes)		
	40°C	50°C	60°C
10	$255 \pm 26^{b,x}$	$270 \pm 15^{b,x}$	$140 \pm 10^{a,x}$
30	$320 \pm 37^{b,y}$	$320 \pm 20^{b,y}$	$195 \pm 30^{a,y}$

Different letters (a, b, c) within each row indicate significant differences ($p<0.05$).
Different letters (x, y, z) within each column indicate significant differences ($p<0.05$).

Table 3. Results of fitness of various thin-layer drying models at 10 kPa

Model	Temperature, °C	SSE	RMSE
Newton	40	0.012182	0.031861
	50	0.029717	0.04451
	60	0.005213	0.025528
Page	40	0.007808	0.025508
	50	0.013935	0.03048
	60	0.003871	0.021998
Henderson and Pabis	40	0.011578	0.031062
	50	0.025975	0.041613
	60	0.005101	0.025252
Logarithmic	40	0.002179	0.013476
	50	0.002566	0.01308
	60	0.001593	0.014111
Two-term	40	0.00099	0.00911
	50	0.00128	0.00924
	60	0.00058	0.0085
Term-term exponential	40	0.00551	0.02142
	50	0.01207	0.02837
	60	0.00281	0.01873
Approximation of diffusion	40	0.001416	0.010864
	50	0.001641	0.010459
	60	0.000755	0.009717

the AOAC standard moisture determination method 930.15 with a 1 to 2% deviation.

Statistical analysis

The results of the data analysis are presented as the mean values with standard deviations. Values were considered at 95% significance ($\alpha<0.05$), and a statistical program, Minitab 14, was used to perform the calculations.

Results and Discussion

Drying characteristics

Fresh *Andrographis paniculata* (AP), with an initial moisture content in the range of 2.41 to 2.88 kg water/ kg solid, was dried in a vacuum dryer until it reached a moisture content below 0.11 kg water/ kg solid. The drying times required for various conditions are shown in Table 2. From Table 2, we can see that the temperature significantly affects the drying time, where a higher temperature corresponds to a shorter drying time. This result may be because a larger driving force is implied for heat transfer and mass transfer during drying at higher temperatures, resulting in a higher moisture diffusivity (Nor *et al.*, 2009). This finding was similar to those in studies utilising eggplant (Wu *et al.*, 2007), sweet cherry (Doymaz and İsmail, 2011), coconut presscake (Jena and Das, 2007), Asian white radish slices (Lee and Kim, 2009), carrot (Arevalo-Pinedo *et al.*, 2004), and pumpkin (Arévalo-Pinedo and Murr, 2006).

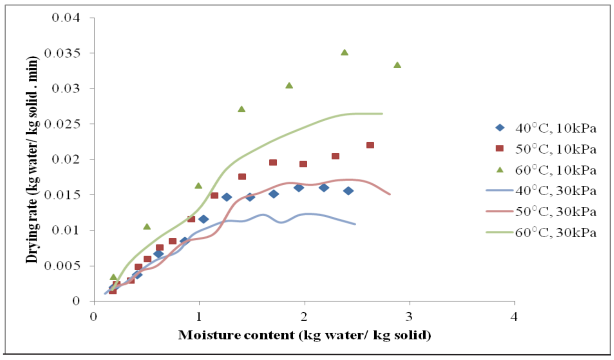

Figure 1. Drying rate of Andrographis paniculata under various drying conditions

In this study, pressure was also found to affect the drying time significantly (Table 2). Generally, a longer drying time was needed as the pressure increased from 10 to 30 kPa. This behaviour can be explained by the fact that a reduction of pressure results in a lower boiling point of water. At a lower temperature, the water more easily converts to its vapour form, and the driving force for the outward moisture diffusion process increases. More water vapour diffuses across the air-moisture interface and is available at the surface of the drying product. Thus, the moisture molecules can escape from the drying product more easily and more rapidly (Nor *et al.*, 2009). Similar behaviour has been reported for carrot(Arevalo-Pinedo *et al.*, 2004) and pumpkin (Arévalo-Pinedo and Murr, 2006).

Drying rate

From Figure 1, it can be seen that the drying rate was higher in the initial period of drying and decreased as the drying time increased. The initial drying rate was also relatively higher as temperature increased at low pressure.

Drying model

Because a pressure of 10 kPa significantly shortened the drying time, the drying kinetics at this pressure were studied. The moisture content data

Table 4. Estimated values of the drying constants and coefficients for the two-term thin-layer drying model

Temperature, °C	Drying constant, min^{-1}		Coefficient	
	k	k_0	a	b
40	0.006063	-0.00951	1.005791	0.009191
50	0.008187	-0.0054	0.966439	0.048268
60	0.011004	-0.03092	1.010602	0.00083

Table 5. Effective diffusivity of AP at various temperatures and an absolute pressure of 10 kPa

Temperature, °C	D_{eff}, m^2/s	SSE	RMSE
40	3.1×10^{-13}	0.050361	0.064783
50	4.23×10^{-13}	0.039489	0.051309
60	6.72×10^{-13}	0.035967	0.067052

were converted into moisture ratios (MRs) and fitted with several selected thin-layer drying models, as shown in Table 1. The best model was selected based on the least SSE and RMSE, which represent the difference between the individual experimental and calculated data and the goodness of fit, respectively. As observed in Table 3, the lowest value of SSE and RMSE were found for the two-term model. Therefore, this model was selected as the best-fit model to represent the thin-layer drying behaviour of AP at 10 kPa. Table 4 presents the estimated values of the parameters in the two-term model for different drying temperatures. These estimated values were used to fit the experimental determined MR and are plotted in Figure 2. As seen, the two-term model provided conformity with the experimental data.

The results were also fitted with Fick's second law as a theoretical model with which to determine the effective diffusivity and activation energy. The obtained diffusivity values are shown in Table 5. The effective diffusivity increased as the temperature increased. These results are in agreement with the reported findings for Asian white radish (Lee and Kim, 2009) and black tea (Panchariya *et al.*, 2002). Arrhenius' model was then used to determine the relationship between effective diffusivity and temperature and is illustrated in Figure 3. The results show a linear relationship between ln Deff and 1/T. The activation energy was determined to be 33.4 kJ/mol. This value is lower than the activation energy of black tea drying (406.028 kJ/mol) (Panchariya *et al.*, 2002).

Conclusions

In this study, the temperature and pressure of drying significantly affected the drying process. The drying time was relatively shorter at high temperatures and low pressures. Based on a non-linear regression, a two-term model was chosen to describe the drying behaviour of AP. The value of the effective diffusivity was 10-13 m^2/s, and this value increased as temperature increased. The activation energy for moisture diffusion was determined to be 33.4 kJ/mol.

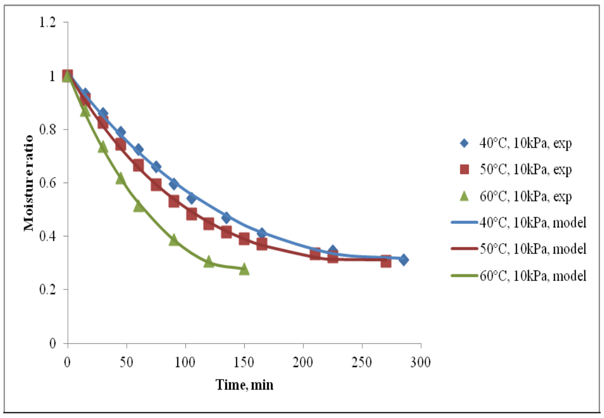

Figure 2. Experimental and two-term model drying curves at various drying temperatures and an absolute pressure of 10 kPa

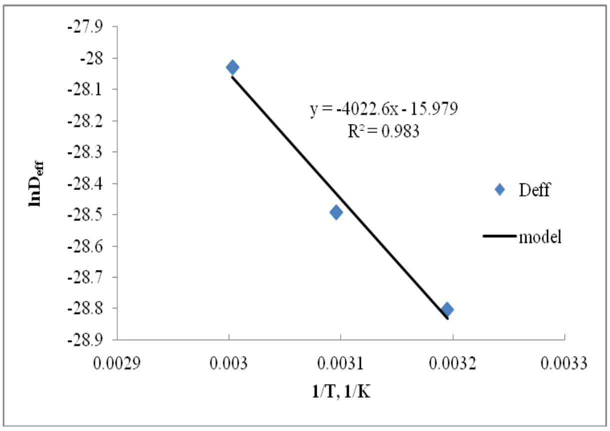

Figure 3. Relationship between effective diffusivity and temperature based on Arrhenius' model

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