Dual Impact of Different Drying Treatments and Ethanol/Water Ratios on Antioxidant Properties and Colour Attribute of Jackfruit Leaves (*Artocarpus heterophyllus* Lam.) Mastura Variety (J35)

Haswani Maisarah Mustafa,[a] Nor Amaiza Mohd Amin,[a,*] Rabitah Zakaria,[a] Mohd Shamsul Anuar,[a] Azhari Samsu Baharuddin,[a] Halimatun Saadiah Hafid,[a,b] and Farah Nadia Omar [a]

Artocarpus heterophyllus (jackfruit) leaves (JL) are a waste product that is commonly used as livestock feed. Jackfruit leaves have been revealed to possess many medicinal values such as antioxidant and anti-inflammatory properties. In this study, different drying treatments (shade (SD), sun (SN), and oven (OV)) and ethanol/water ratios (E/W) were investigated to evaluate the impact on drying kinetics, color, and antioxidant properties of jackfruit leaves. Results showed that the Newton model was the best fitted mathematical model for the JL drying kinetics. The moisture effective diffusivities ranged from 2.920×10^{-10} to 6.814×10^{-10} m^2/s over the temperature range studied. Shade drying was able to preserve the green pigment better than OV and SN drying treatments. Treatment with ethanol/water ratio at 80% and oven-dried (OV80) revealed the highest phenolic content (195.05 ± 1.21 mg gallic acid equivalent (GAE)/g extract weight (EW)), flavonoid content (11.02 ± 0.17 mg artocarpin equivalent (AE)/g EW), and antioxidant activities (90% scavenging activity and reducing power of 1043.84 ± 5.28 µM trolox equivalent (TE)/g EW) compared to SD and SN treatments. The OV80 also possessed the highest artocarpin, squalene, and β-sitosterol contents determined. The OV80 was selected for improving antioxidant and colour stability, and has the potential to be developed into functional biopolymer production.

Keywords: Artocarpus heterophyllus leaves; Drying; Newton model; Antioxidant; Aartocarpin

*Contact information: a: Department of Process and Food Engineering, Faculty of Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor; b: Graduate School of Life Science and System Engineering, Kyushu Institute of Technology, 2-4 Hibikino, Wakamatsu-ku, Kitakyushu, 808-0196, Japan; * Corresponding author: myza@upm.edu.my*

INTRODUCTION

Jackfruit, which is scientifically known as *Artocarpus heterophyllus* Lam., is a tropical fruit that is popular in Malaysia. Originated from India, it is widely cultivated in other countries such as Thailand, Vietnam, Indonesia, the Philippines, Sri Lanka, China, and Brazil (Zheng *et al*. 2014). Jackfruit has been reported to have both flesh and seeds customarily eaten, boiled, or fried; and the non-edible part, such as leaves, has been revealed to possess medicinal values such as antioxidant, anti-inflammatory, antimicrobial, anticancer, hypoglycemic, and hypolipidemic activities (Jagtap and Bapat 2010; Baliga *et al*. 2011).

Jackfruit leaf extract was also reported to contain high antioxidant properties in the aqueous fraction (294.5 mg/g), ethyl acetate extract (361.2 mg/g), both of which exhibited high scavenging activities (219.9 and 235.8 μg/mL, respectively) (Loizzo *et al*. 2010).

Upon detachment from the twigs after harvesting, jackfruit leaves may deteriorate, thus reducing their shelf life. Therefore, there is a need for preservation to be able to prolong their shelf life. Drying is a preservation method commonly applied to prevent microbial growth and reduce enzymatic activity, thus extending the shelf life of plant material at room temperature. However, during the drying process, deterioration and changes in properties lead to the undesirable impact on the quality of the product (Babu *et al*. 2018).

Changes in color, aroma, and degradation of bioactive compounds serve as an indicator of the oxidation process. Exposure to exhaustive long hours of drying time and high temperature could result in reduced antioxidant properties. Drying methods that are commonly used include convection drying, shade drying, and sun drying. Previous studies exposed jackfruit leaves to various drying methods, particularly to evaluate the effect of drying methods on the cellular constituent that can release the phenolic compound from the plant material (Roshanak *et al.* 2016). Prakash *et al*. (2017) reported on the shade-dried jackfruit leaves for wound healing properties, focusing on the excision model and phytochemical screening for the presence of flavonoids. Ojwang *et al*. (2017) reported that the air-dried drying method of jackfruit leaves still contain an amount of antioxidant activities (65% to 78%).

The air-dried under the shade of jackfruit leaves collected from different districts in Uganda were also revealed to present an abundant amount of total phenolic content and total flavonoid content (ranging from 37.39 to 30.92 mg/g and 5.02 to 6.70 mg/g, respectively).

For many years, different drying methods of leaves have been adopted to reach a compromise between quality and efficiency. Options included the commonly used method, the shade drying. Donkor *et al*. (2016) reported that natural drying may result in a slight decrease of flavonoid compound with the temperature increased. In contrast, a higher temperature (60 to 70 °C) leads to an increase in carvacrol content but causes darkening of the leaf color (Rahimmalek and Goli 2013; Roshanak *et al*. 2016). However, the leaf color may retain the color quality demonstrated by lowering drying temperature to 40 °C (Chua *et al*. 2019a).

Conventional drying process always suffers from a long drying time with high energy consumption. A co-drying technique with solvent was introduced to increase the drying rate with better process control without reducing the quality of bioactive compounds, colour, and antioxidant properties. Thus, it could lead to better nutrient preservation. To the best of the author's knowledge, there has been a lack of studies on the effect of drying characteristics and solvent ratios on the quality attributes of jackfruit (*Artocarpus heterophyllus*) leave extract. The aim of the present work is to determine the impact of shade, sun, oven treatments, and ethanol/water solvent ratios on colour and antioxidant properties of jackfruit (*Artocarpus heterophyllus* Lam. var. *Mastura*) (J35) leaves extract.

EXPERIMENTAL

Materials

Fresh jackfruit leaves (*Artocarpus heterophyllus* Lam. var. *Mastura*) were harvested from a jackfruit plantation owned by Farmers' Organisation, Maran, Pahang Darul Makmur, Malaysia (3.5829° N, 102.7748° E). The leaves were labelled as JL. Upon arrival, the leaves were washed with clean water to remove surface dirt and any debris. Then, it was packed in average weight of 500 g of JL in polyethylene bag prior to drying. Moisture content of the sample was measured using a digital moisture analyzer (MB45; Ohaus Corporation, Parsippany, NJ, USA) in triplicate. The leaves were weighed to (2.0 ± 0.01 g) and heated at 103 ± 0.1 g until a constant weight was achieved.

Chemicals and reagents

Ethanol was purchased from John Kollin Corporation (Midlothian, Scotland). Folin-Ciocalteu reagent, sodium carbonate (Na_2CO_3), and ferric chloride hexahydrate ($FeCl_3 \cdot 6H_2O$) were purchased from Merck, Darmstadt, Germany. Chemicals of 2,4,6-tris-2,4,6-tripyridyl-s-triazine (TPTZ), sodium acetate trihydrate, 1,1-diphenyl-2-picrylhydrazyl (DPPH), and Trolox (6-hydroxy-2,5,7,8-tetramethylchromane-2-carboxylic acid, 97%) as well as 5-α-cholestane, squalene, and β-sitosterol standards were purchased from Sigma Chemical Co. (St. Louis, MO, USA). Artocarpin standard was purchased from Chemfaces (Wuhan, China). Methanol used for high-performance liquid chromatography (HPLC) analysis was purchased from Merck (Darmstadt, Germany). All reagents used were of analytical grade unless otherwise stated.

Methods

Drying of jackfruit leaves

Shade drying: Jackfruit leaves were spread evenly on a net in a single layer under shade with average temperature and relative humidity of approximately 25 °C and 58%, respectively. Samples were dried and the moisture content measurements were taken on an hourly basis until they reached constant moisture content. Moisture content was analyzed using the digital moisture analyzer.

Sun drying: Approximately 30 g of fresh JL were weighed in different batches. Every batch was evenly distributed and exposed to sunlight with average temperature readings and relative humidity of approximately 33 °C and 45%, respectively. All samples were allowed to dry from 8:00 h to 18:00 h and collected in the evening, and the moisture content reading was taken on an hourly basis. Samples were immediately put in desiccator for 15 minutes before measuring the moisture content using the digital moisture analyzer in triplicate until constant weight was achieved in percentage (%) unit.

Oven drying: Approximately 30 g of jackfruit leaves were dried at 40 °C using an automatic electric oven (Memmert GmbH + Co. KG, Büchenbach, Germany) in triplicate. The oven was pre-heated for 30 minutes to condition the temperature at 40 °C. Then, the jackfruit leaves were evenly spread on the stainless steel grids to avoid stacking on each other to achieve maximum drying surface area. Moisture content of samples was checked using the digital moisture analyzer.

Mathematical modeling of drying kinetics

Table 1 shows the mathematical models to describe the drying kinetics of jackfruit leaves for each method. The moisture ratios (MR) of JL were calculated using Eq. 1,

$$MR = \frac{M_t - M_e}{M_0 - M_e} \quad (1)$$

where MR is the moisture ratio, M_t is the moisture (%) at time t, M_0 is the initial moisture (%), and M_e is the moisture at equilibrium (%). Assuming the values of M_e equal to zero are negligible, due to M_e being relatively small compared to M_t or M_i (Doymaz and Kipcak 2018).

Problem solver Microsoft Excel (Microsoft Corp., v. 2010, Redmond, WA, USA) was used to solve the seven nonlinear mathematical models (Table 1). Chi square (χ^2) and root mean square error (RMSE) were used to evaluate the relationship of the experimental and predicted results in addition to the correlation coefficient (R^2). The following equations were employed to calculate χ^2 and RMSE,

$$\chi^2 = \frac{\sum_{i=1}^{n} (MR_i - \overline{MR})^2}{n-1} \quad (2)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n} \left(MR_{i\ experimental} - MR_{i\ calculated}\right)^2}{n}} \quad (3)$$

where $MR_{i\ experimental}$ is the concentration of experiment I, and $MR_{i\ calculated}$ is obtained from the model equation, and n represents the number of experiments.

Table 1. Mathematical Model Applied for Drying Curves

No.	Model Name	Model Equation
1.	Newton	*MR* = exp(-k*t*)
2.	Logarithmic	*MR* = a exp(-k*t*) + c
3.	Verma	*MR* = a exp(-k*t*) + (1 - a) exp(-g*t*)
4.	Two term	*MR* = a exp(−k$_0$*t*) + b exp(−k$_1$*t*)
5.	Midilli	*MR* = a exp(-k*t*^n) + bt
6.	Page	*MR* = exp(-k*t*^n)
7.	Henderson and Pabis	*MR* = a exp(-k*t*)

Determination of effective diffusivity

Drying characteristics of biological products can be described by Fick's law diffusion model as represented by Eq. 4,

$$\ln(MR) = \left(\frac{6}{\pi^2}\right) - \left(\frac{D_{eff}}{r^2}\right) x\ t \quad (4)$$

where D_{eff} is the effective diffusivity of moisture (m^2/s), t is the time for drying (s), and r is the radius of the plant ($r = 7.8 \times 10^{-4}$ m). The form of Eq. 4 can be applied for particles with slab geometry assuming uniform initial moisture distribution. The diffusivity equation provides an approximate method to present a common quantitative comparison of various products in the aspect of the diffusion coefficient in the process of drying.

Color determination

The dried leaves were crushed and ground to a fine powder using a hammer mill (Perten Laboratory Mill 120; Perten Instruments, Hägersten, Sweden). Evaluations of colour were tested on a colorimeter (Konica Minolta Color Reader CR10; Konica Minolta Sensing Americas, Inc., Ramsey, NJ, USA). Measurement of color was determined in L^*, a^*, and $\underline{b}^*$ coordinates, where L is the lightness (0 = black, 100 = white). The L^*, a^*, and b^* values are the averages of three readings. The chromaticity coordinate a^* measures red (+) and green (-), and the chromaticity coordinate b^* measures yellow (+) and blue (-). The L^* and $a^*/\underline{b}^*$ values are commonly used as an index to report the color quality.

Extraction of jackfruit leaves

Shade-dried (SD), sun-dried (SN), and oven-dried (OV) samples were extracted using the maceration method. The sample was dissolved in ethanol/water ratio of 1 : 20 with modification (Perva-Uzunalic *et al*. 2006) as described in Table 2. Samples were left in the incubator shaker for 3 d with continuous stirring at 200 rpm. Samples were filtered and the solvent was evaporated to dryness using a rotary evaporator to afford a thick green mass. All experiments in this study were performed in triplicate.

Table 2. Ranges of Experimental Variables

Drying Method	Ethanol Percentage (%)	Acronym
Shade (SD)	100	SD100
	80	SD80
	50	SD50
	0 (Water only)	SD-W
Sun (SN)	100	SN100
	80	SN80
	50	SN50
	0 (Water only)	SN-W
Oven at 40 °C (OV)	100	OV100
	80	OV80
	50	OV50
	0 (Water only)	OV-W

Spectrophotometric Analysis

Total phenolic content (TPC) and total flavonoid content (TFC)

Total phenolic content and total flavonoid content were analyzed spectrophotometrically (Shimadzu UV1800; Shimadzu Scientific Instruments, Columbia, MD, USA) as described by Mustafa *et al*. (2016). Gallic acid was prepared according to the Folin–Ciocalteu assay as standard solution, and the results were prepared with gallic acid as standard with calibration curve (0.0, 0.2, 0.4, 0.6, 0.8, and 1 mg/mL) and expressed as gallic acid equivalent (mg GAE/g extract). Absorbance was read at 725 nm against ethanol as blank. The total flavonoid content was prepared with artocarpin as standard with the calibration curve (0.0, 0.2, 0.4, 0.6, 0.8, and 1 mg/mL). The absorbance was measured spectrophotometrically at 513 nm. Results were expressed as artocarpin equivalents (mg AE/ g extract).

Antioxidant activities

The ferric reducing antioxidant power (FRAP) assay was performed according to the method described by Liu *et al*. (2009) with modifications of the adjusted volume of FRAP reagent. The FRAP reagent contained 20 mM $FeCl_3 \cdot 6H_2O$, 10 mM TPTZ (2,4,6-tripyridyl-s-triazine) solution in 40 mM HCl, and 0.3 M acetate buffer with pH 3.6, and incubated at 37 °C for 10 min in an incubator (Memmert GmbH + Co. KG, Schwabach, Germany). The FRAP reagent was mixed in the ratio of 1:1:10. Aliquot of 100 µL sample was mixed with 2.9 mL of FRAP reagent. The absorbance was measured spectrophotometrically at 593 nm after incubation at room temperature for 1 h. Trolox (1000 µM) was used for the calibration curve, and the results were expressed as µM of Trolox equivalents per gram extract weight.

The antioxidant activity was carried out through evaluation of free radical scavenging effect on 1,1–diphenyl-2-picrylhydrazyl (DPPH). The determination was based on the method described by Yamaguchi *et al*. (1998) with some modification on sample volume added. An aliquot (600 µL) sample was added to 4.5 mL of 0.1 mM DPPH ethanolic solution. The mixture was then thoroughly vortexed and incubated for 20 min in dark conditions at room temperature. The absorbance was measured at 517 nm against ethanol as blank. Results were expressed as a percentage of inhibition of the DPPH radical and calculated according to Eq. 5,

$$\% \text{ inhibition of DPPH} = \frac{(\text{Abs control} - \text{Abs sample})}{\text{Abs control}} \times 100 \quad (5)$$

where *Abs control* is the absorbance measured as absorbance unit (AU) of DPPH without sample.

Quantification of Compounds

Bioactive compound by HPLC

The sample with the highest amount of antioxidant properties from each treatment was further analyzed for artocarpin content. Artocarpin in samples was analyzed using a Shimadzu HPLC system (Shimadzu SPD-10AV, Shimadzu Scientific Instruments, Columbia, MD, USA), which was equipped with a ultraviolet/visible (UV/VIS) detector (SPD-10AV) (i.d. 4.6 × 250 mm, 5 µm column). The analysis was performed according to Septama and Panichayupakaranant (2016) with modification of the adjusted mobile phase. A mobile phase was prepared that consisted of methanol and water (85% methanol and 15% deionized water), at a flow rate of 1 mL/min. Detection of artocarpin using HPLC was coupled using a diode array detector (DAD) at 285 nm. All volume samples were injected at 20 µL. The identification of the compound was based on the comparison with external standards. The signals and area of each peak were processed using Class VP software (Shimadzu LabSolutions, v.6.14, Kyoto, Japan). The amount of sample was expressed as milligram per gram of extract weight (mg/g).

Volatile compounds by gas chromatography (GC)

The sample with the highest amount of antioxidant properties from each treatment was further analyzed for volatile compounds. Samples were prepared by adapting the standard method Cd 11b-91 (Bruschweiler and Dieffenbacher 1991) using Shimadzu GC-2010 Plus equipped with split/splitless injector and flame ionization detector (FID) (Shimadzu, Kyoto, Japan). Approximately, 0.02 g of the sample was weighed and added

with 5-α-cholestane serving as an internal standard (ISTD), 0.3 mL pyridine, and 0.15 mL N-trimethylsilyl-N-methyl-trifluoroacetamide (MSTFA). The sample solution was mixed well and heated at 40 °C for silylation purposes. The GC analysis was performed using a capillary column, HT5 (12 m × 0.32 mm, i.d. 0.1 m, Scientific Glass Engineering Analytical Science (SGE), Melbourne, Australia). The temperatures of the flame ionization detector (FID) and injector were set at 370 °C and 360 °C, respectively. The oven was set initially at 80 °C, held for 1 min, and then increased from 10 °C /min to 360 °C /min, followed by maintaining at 360 °C for 15 min. Hydrogen was used as a carrier gas at a flow rate of 30 mL/min. For the GC analysis, a split ratio of 1:10 was set, and a 1 µL of sample solution was injected into the GC system.

Statistical analysis

All experiments were performed in triplicate, and the results were given as means ± standard deviation. Differences among the treatments were determined using an analysis of variance (ANOVA) and a Tukey test with a confidence level of $P < 0.05$ to indicate significant differences. The analyses were made using Minitab 16 software (Minitab Inc., State College, PA, USA).

RESULTS AND DISCUSSIONS

Impact of Drying Treatments

Heat-treated jackfruit leaves resulted in losses in moisture content under shade, sun, and oven, as depicted in Fig. 1. The initial moisture content of jackfruit leaves was 60.4 to 62.7%. The drying time for sun treatment was 5.4 times and 1.6 times faster than shade and oven conditions, respectively. The reduced drying time distributed by the external parameters that may affect the drying process include wind speed, humidity, temperature, air stream velocity, heat supply, the contact between hot surfaces and the wet solid, and solar radiation (Sodha *et al*. 1985; Jain and Tiwari 2003; Prasad 2009). In addition, open space under sun drying allowed higher diffusion coefficient of product per unit exposed area while oven treatment had a slower drying rate due to lower diffusion coefficient of the oven resulting in an increase of relative humidity in the oven chamber (Kumar and Tiwari 2007; Arslan and Özcan 2010). The highest temperature that was recorded during sun drying the sample was approximately 38 °C during mid-day. The heat penetrates inside the sample, producing a moisture gradient in the sample, and water vapor starts to form. Diffusion of moisture and water vapor takes place from the interior replacing the loss of moisture by evaporation on the surface. Higher drying rates occur in the beginning and become gradually reduced through the end of drying process. Similar results have been obtained by other researchers in relation to other fruits and vegetables (Alara *et al*. 2018; Tellez *et al*. 2018). Although the sun drying method showed the shortest time compared to shade and oven methods due to fluctuating sunlight, the drawbacks of this treatment are the unhygienic conditions, such as dust particles, microbial attack, prone to damage, and loss to nutritional attributes, at the peak of daytime. Whilst shade drying is able to prevent damage and loss of compound of interest, the drawback of this shade treatment is that it requires the longest time to achieve its constant weight. Furthermore, natural drying treatments (shade and sun drying) are also

unable to achieve consistent quality standards (Roshanak *et al*. 2016; Babu *et al*. 2018). Despite many drawbacks highlighted for natural drying treatments, these methods are accessible and free source. Although oven drying was operated at a cost estimated at USD 80, this method is preferable as it is more efficient in terms of producing optimum quality product.

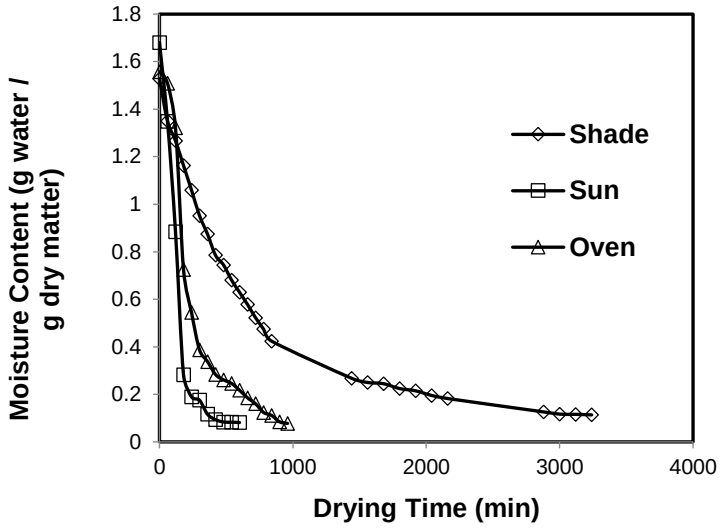

Fig. 1. Drying curves for jackfruit leaves and data given are the mean values of three replications

The predicted data by the Newton model and experimental data moisture ratios *versus* drying time for jackfruit leaves are shown in Fig. 2. The results showed that drying air temperature was an effective parameter for the drying of jackfruit leaves. Seven different drying models of MR were used to predict the moisture content as a function of drying time (Table 1).

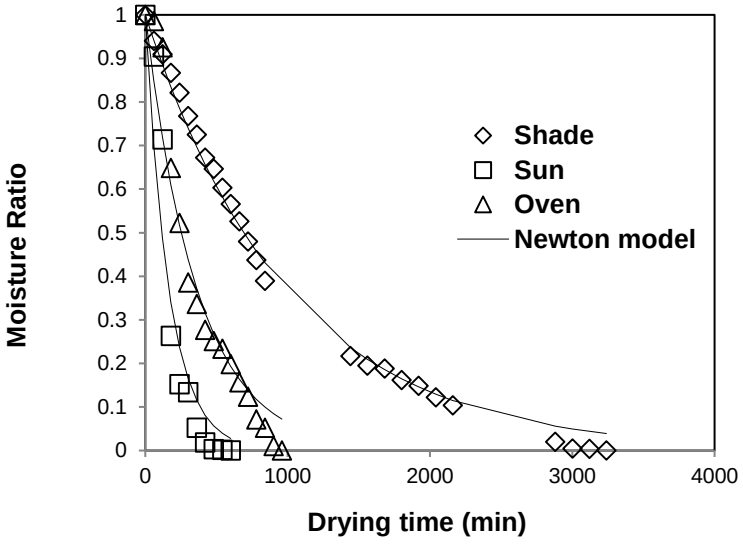

Fig. 2. Variation of moisture ratio of experiment and predicted by Newton model of various drying conditions for jackfruit leaves and data given are the mean values of three replications

Table 3. Curve Fitting for Shade, Sun, and Oven Drying

Model Name	Drying Condition	Constants	R^2	RMSE	χ^2
Newton*	Shade	k = 0.0010	0.9948	0.1676	0.3480
	Sun	k = 0.0059	0.9999	0.1002	0.2450
	Oven	k = 0.0027	0.9999	0.0974	0.2208
Logarithmic	Shade	a = 0.9909 k = 0.0011 c = 0.1206	0.9460	0.1515	0.3147
	Sun	a = 0.0009 k = 1.0000 c = 0.3015	0.9963	0.0644	0.1459
	Oven	a = 0.8840 k = 1.0000 c = 0.3014	0.9998	0.0974	0.2208
Verma	Shade	a = 1.0700 k = 0.0012 g = 1.3309	0.9906	0.1662	0.3451
	Sun	a = 1.6497 k = 0.0089 g = 0.7500	0.9772	0.0619	0.1404
	Oven	a = 1.2200 k = 0.0033 g = 0.5000	0.9847	0.0945	0.2141
Two Term	Shade	a = 0.5510 k_0 = 0.0010 b = 0.4700 k_1 = 0.0012	0.9934	0.1671	0.1431
	Sun	a = 0.5000 k_0 = 0.0085 b = 0.5799 k_1 = 0.0033	0.8718	0.0493	0.1117
	Oven	a = 0.5500 k_0= 0.0030 b = 0.5760 k_1 = 0.0033	0.9692	0.0915	0.2074
Henderson and Pabis	Shade	a = 1.0490 k = 0.0010	0.9935	0.1600	0.3471
	Sun	a = 1.0987 k = 0.0059	0.9292	0.0560	0.1269
	Oven	a = 1.2000 k = 0.0034	0.9620	0.0901	0.2043
Midilli	Shade	a = 0.9359 k = 0.0018 n = 0.9040 b = 0	0.9681	0.1587	0.0684
	Sun	a = 0.9965 k = 0.0060 n = 0.9990 b = 0	0.9168	0.0545	0.1236
	Oven	a = 1.0997 k = 0.0031 n = 0.9990 b = 0	0.9687	0.0914	0.2073
Page	Shade	k = 0.0010 n = 1.0010	0.9945	0.1677	0.3481
	Sun	k = 0.0035 n = 1.0999	0.9357	0.0567	0.1287
	Oven	k = 0.0025 n = 1.0249	0.9563	0.0890	0.2019

*Best fitted model

The statistical analyses of curve fitting coefficient determination (R^2), reduced root mean square error (RMSE), and reduced chi-square, characterized for jackfruit leaves dried under shade, sun, and oven conditions are shown in Table 3. Newton, Logarithmic, and Verma models exhibited high values of R^2 for all drying treatments ranging between 0.9460 and 0.9999 with slightly low RMSE values ranging from 0.0644 to 0.1676.

Effective Diffusivity

The effective diffusivity (D_{eff}) values (m^2/s) of dried jackfruit leaves for shade, sun, and oven drying treatments were 2.9203×10^{-10}, 6.8141×10^{-10}, and 6.5707×10^{-10}, respectively. Sun drying possessed the highest D_{eff} values, which were approximately 2.3 fold than that of shade drying. For shade and sun drying, it was expected that D_{eff} values increased with the increase in temperature. The D_{eff} value for the sun drying was 1.04 fold greater than that for oven, which revealed that the process of open sun drying had better mass transfer efficiency than that of oven drying treatment studied. This can be associated with the free air circulation during open drying process. In general, D_{eff} values fall within the range of 10^{-11} to 10^{-9} for dried food materials (Arslan and Özcan 2010).

Colour

Drying treatments had a significant effect on the color changes of jackfruit leaves. Therefore, colour assessment is another parameter that was observed during the drying of leaves. The colour measurement of dried jackfruit leaves revealed that the L^* value of shade drying treatment was the highest among all dried leaves (15.8), followed by oven drying (13.4), while sun drying revealed the lowest L^* value (11.1) among dried treatments ($P < 0.05$). Exposure to direct sunlight is the most effective factor in color damage during drying (Rahimmalek and Goli 2013). Similar results were obtained for b^* values. Sun drying treatment showed the highest b^* value (39.5) in comparison to others. For a^* value (greenness), shade drying treatment had the highest value (more negative), while sun drying showed the most discoloration of green pigment. Sun drying caused a more significant increment of a^* values than the other treatments, which suggested that the degradation of green color (Fig. 3) in the final product occurred in a greater ratio. This could be due to the loss of chlorophyll content that is responsible for the green color of plant leaf and enzymatic browning.

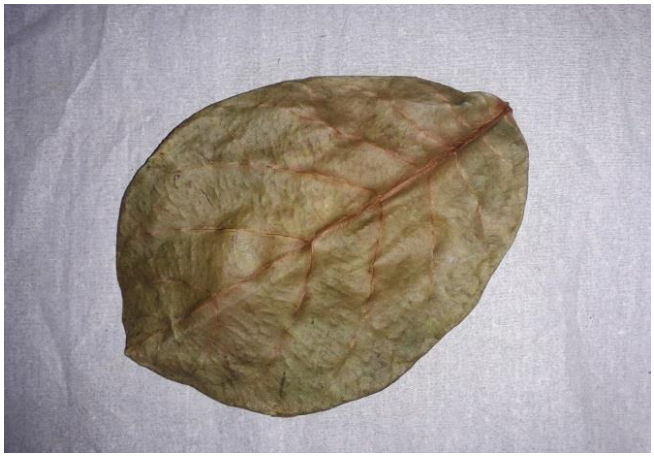

Fig. 3. Degradation of green colour of sun-dried jackfruit leaf

The degradation of chlorophyll could be due to the chemical nature of pigments that are responsible for the green colour in leaves. Plant leaves containing chlorophyll A and B will degrade to pheophytin A and B, respectively when exposed to the heat. During the process of exposure to heat, pheophytin is produced when chlorophyll containing magnesium was replaced by two hydrogen atoms in the presence of mild acids (Izli *et al*. 2017; Roshanak *et al*. 2016). The possibility of discolouration of green colour was expected to occur in sun-dried jackfruit leaves. Therefore, sun drying is known to be the least desirable for the final dried product. Overall, sun and shade treatments were considered the least and the most desirable drying methods, respectively, regarding the final colour of jackfruit leaves.

Total Phenolic Content and Total Flavonoid Content

The impacts of drying treatment on TPC and TFC are shown in Fig. 4. It was found that there was a significant increase in total phenolic content and total flavonoid content observed over the extraction drying treatments (SD, SN, and OV), and the total phenolic content and total flavonoid content reached maximum values around 195.05 mg GAE/g and 11.02 mg AE/g of jackfruit leave extracts, respectively, both dried in the oven. At different drying treatments, there were significant percentage increments for the OV80 that were 1.33 and 1.28 fold higher than that for shade and sun. The TFC observed the highest value of 11.02 mg AE/g of jackfruit leave extracts dried in the oven, 1.48 fold and 1.20 fold significantly greater than the shade and sun, respectively.

The increase in TPC and TFC as affected by drying treatment could be due to the liberation of the cell constituents from the plant cell that may accelerate the movement of bound phenolic and flavonoid compounds due to the thermal treatment as the heat exerts adjustments to the plant tissue microstructure, thus rupturing the structure of cell integrity, thereby allowing migration of phenolic compounds from the plant cells (Chua *et al.* 2019b). The longer time required for shade drying and the exposure to UV under open sun may affect the sensitive phytochemical compounds toward breakdown under various factors of chemical reaction comprising of light, oxygen, and enzymes (Martinez-Las Heras *et al*. 2014).

The effect of ethanol concentrations on the extraction of TPC and TFC in jackfruit leaves extracts are shown in Fig. 4. At different ratios of ethanol/water from 0% to 80% for the same drying treatment, results showed an increase in TPC and TFC values. There were significant increments at 80% E/W, which possessed 2.67 fold and 5.68 fold higher values than water extraction for TPC and TFC assays. However, when ethanol concentration reached 100% (v/v), the TPC and TFC values in jackfruit leave extracts decreased. The decline in values for the jackfruit leaves extract could be due to some lipid components that were also extracted, which may hinder the extraction of phenolic compounds in jackfruit leaves extracts. While the extraction with water alone may not allow the optimum extraction of phenolic and flavonoid compounds, water extraction exhibited the lowest amount of phenolic and flavonoid content in this result. However, the addition of certain percentage of ethanol into water could improve extraction efficiency as the solubility of carbohydrates and proteins in ethanol is higher than water (Alara *et al*. 2018). In this study, it showed that JL contained more semi-polar constituents than polar ones.

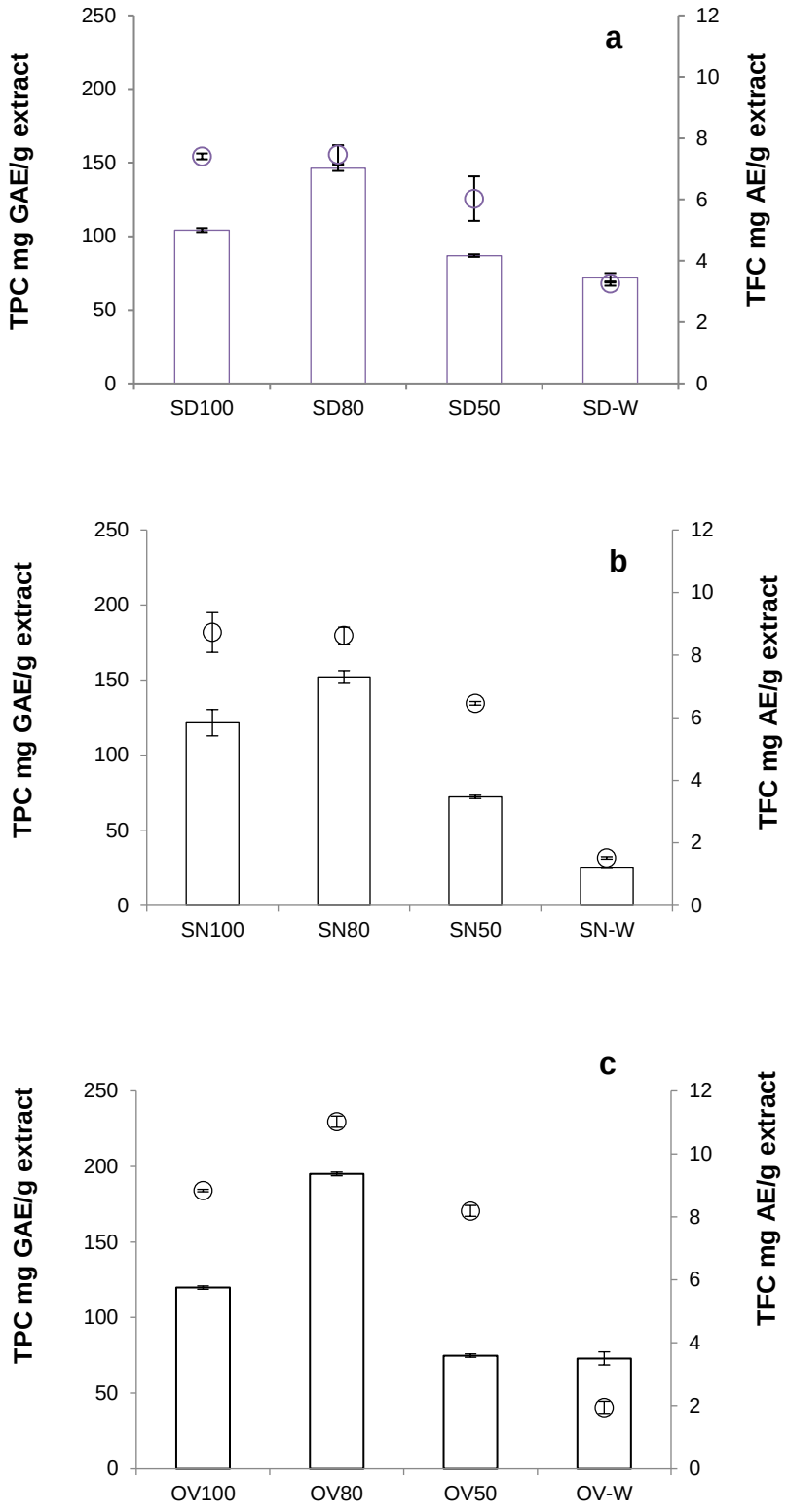

Fig. 4. Effect of different drying treatments and ethanol/water ratios on total phenolic content and TFC of jackfruit (*A. heterophyllus*) leaves extract: a: shade, b: sun, and c: oven; Legend: - □ – TPC, - ○ – TFC

Antioxidant Activities

The impact of drying treatment on DPPH and FRAP is shown in Fig. 5. Based on Fig. 5(a to c), the results showed that the free radical scavenging activities (DPPH) and ferric reducing absorption power (FRAP) of jackfruit leaves extract were highest attained in oven treatment, with values of 90.1% and 1043.80 μM TE/g EW, respectively. Meanwhile, samples dried by shade drying and sun drying showed the lowest DPPH and FRAP, which were 58.8% and 197.50 μM TE/g EW, respectively.

Temperature and drying time were considered as two crucial parameters for interpreting sample extract yield variation when employing different techniques to analyze (Hamrouni-Sellami *et al*. 2013). Higher reducing capacity of an extract serves as an indicator of the ability of its higher potential antioxidant activity. The oven drying treatment at 40 °C may still be able to preserve the heat sensitive bioactive compound present in JL extract. In previous studies, the antioxidant activities and polyphenol contents in heat-treated various herb leaves increased, indicating that phenolic compounds at a partial state of oxidation are exposed to oxygen and may exert a high antioxidant activity (Yi and Wetzstein 2011; Chua *et al*. 2019a).

However, under shade treatment, the lengthy process of exposure to oxygen may imply a greater exposure of the material to enzymatic and oxidation reactions, which thus accelerate the degradation of active compounds. Meanwhile, the antioxidant potential in sun treatment was lower than the oven treatment, which could be due to the direct exposure to sunlight that might degrade the compounds. Evidently, the shorter time in sun treatment did not compensate the deterioration of the antioxidant compounds that could be beneficial to human health (Martinez-Las Heras *et al*. 2014; Barimah *et al*. 2017).

At different solvent ratios, it was found that extraction using 80% ethanol/water (SD80, SN80, and OV80) exhibited significantly higher ($P < 0.05$) FRAP (1043.84 μM TE/g extract weight) and DPPH (90% scavenging effect) values compared to other ratios. The OV80 were found to possess 2.85 fold (FRAP value) and 1.43 fold (free radical scavenging effect) higher jackfruit leaves extract than that of water extraction (365.72 μM TE/g extract weight and 64.7%, respectively). This could be explained by the tendency of phenols to dissolve in a wide range of mixtures of aqueous ethanol compared to water extraction alone. Solvent polarity played an important role in the extractability and solubility of bioactive compounds from plant materials. Several studies have been reported on extracting bioactive compounds using different single solvent, solvent mixtures, and solvent ratios (Pin *et al*. 2010; Dube *et al*. 2017). Alothman *et al*. (2009) reported the highest antioxidant capacities of FRAP at 70% ethanol-water mixtures by increasing the effectiveness of the swelling power of plants with the presence of water and providing high surface area of solute-solvent contact. The addition of water will lower the mixture viscosity and thus will ameliorate the mass transfer of extract (Hemwimol *et al*. 2006).

Correlation of Total Phenolic Contents and Their Antioxidant Activities

It was demonstrated that SD80, SN80, and OV80 exhibited the highest TPC and TFC values compared to other ethanol aqueous compositions. The FRAP and DPPH assays showed the same orientations. In all treatments studied, the phenolic content and its related antioxidant activities exhibited high coefficient of determination (R^2) values at 0.9441 and 0.6700, and were significant at $P < 0.05$.

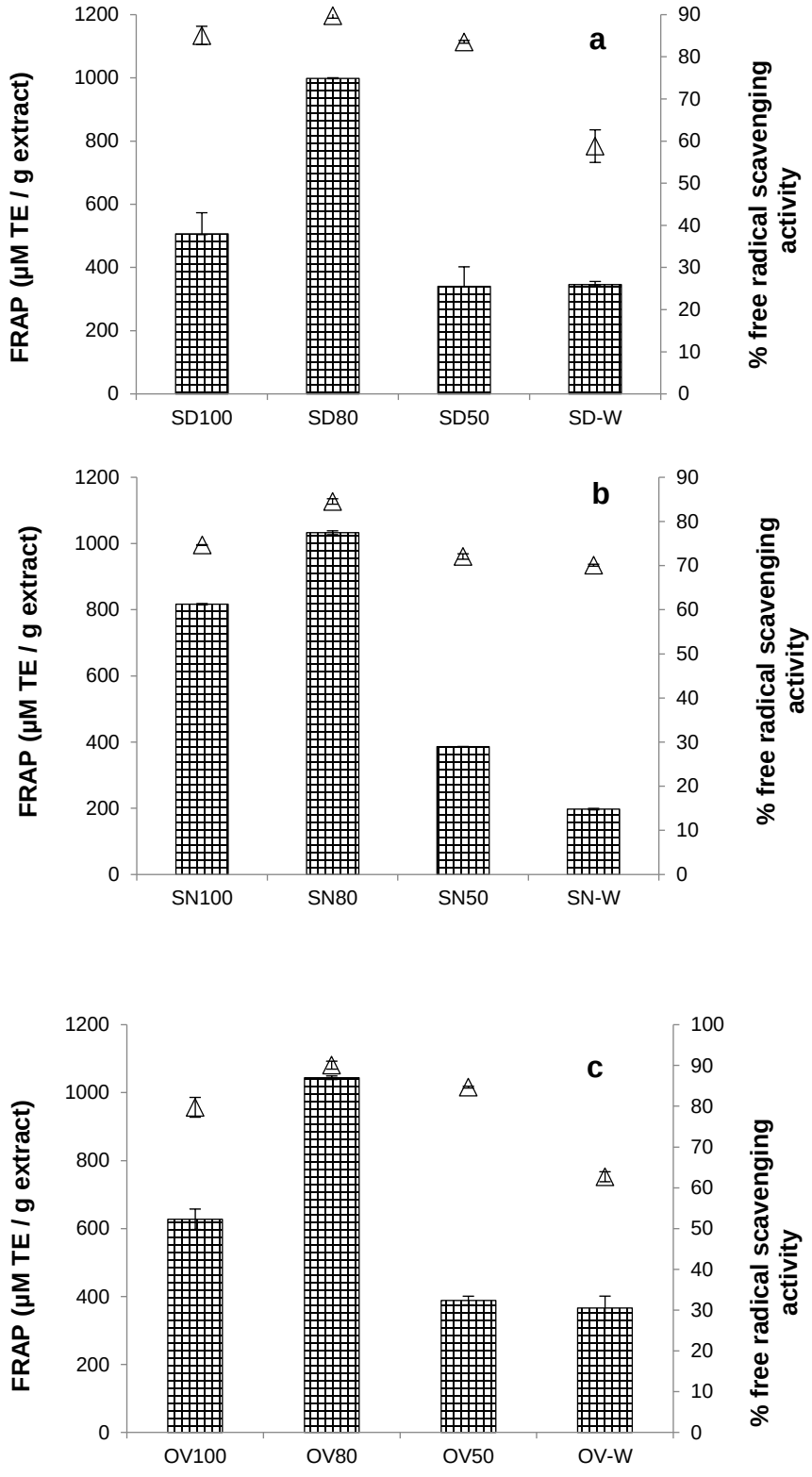

Fig. 5. Effect of different drying treatments and ethanol/water ratios on FRAP and DPPH of jackfruit *(A. heterophyllus)* leaves extract: a: shade, b: sun, and c: oven; - ▓ - FRAP, - Δ - DPPH

In accordance with TPC investigated, *i.e.*, the highest content (80% ethanol/water) demonstrated the highest in antioxidant activities in FRAP assay than any other compositions. However, as this correlation was inverted in the latter assay (DPPH), it was suggested that other constituents (*e.g.*, non-phenolics) could be attributed to the antioxidant ability by donating hydrogen atom of *A. heterophyllus* leaves extract (Catarino *et al*. 2018).

Meanwhile, the correlation between total flavonoid contents (TFC) and its associated antioxidant activities (FRAP and DPPH) yielded R^2 values of 0.7857 and 0.7472, respectively. These correlations exerted their different mechanisms and ability to reduce certain free radicals to form a stable compound and metal ions chelation (Maisarah *et al*. 2013). The mechanism of FRAP and DPPH employed the antioxidant ability to diminish certain radicals (ferric iron and DPPH radical). Another significant correlation between TPC and antioxidant capacity of plant extracts (FRAP and DPPH values) was attained. Based on these correlations, it can be assumed that the phenolic compounds were predominant in contributing to the antioxidant activities of these ethanolic compositions. Different antioxidant activities can be attributed to different mechanism of action either by electron transfer (ET) or hydrogen atom transfer (HAT) (Mustafa *et al*. 2016). Since FRAP values of SD80, SN80 and OV80 extracts were significantly higher ($P < 0.05$) than that scavenging activity, sample may act as HAT to stabilize lipid oxidation.

Artocarpin and Volatile Compounds of Jackfruit Leaves Extract

The artocarpin, squalene, and β-sitosterol content of shade, sun, and oven-dried treatments at 80% ethanol/water ratio (highest values of antioxidant properties) are given in Table 4. All crude samples assayed contained 4.71 ± 0.59 mg/g extract (SD80), 2.53 ± 0.29 mg/g extract (SN80), and 8.35 ± 1.78 mg/g extract (OV80), where OV80 showed significantly higher ($P < 0.05$) artocarpin content compared to other drying treatments. Meanwhile, the same results for volatile organic compounds were observed for squalene and β-sitosterol. Squalene content of OV80 (1.47 mg/g) was found higher than other drying treatments. However, the β-sitosterol content of SD80 and OV80 was found not statistically significant. Sun drying showed the lowest values of both volatile compounds. In this study, oven drying at low temperature with 80% ethanol/water ratio led to the bound phenolic and flavonoid contents being released from the plant matrix and recovered from the thick mass extract during the extraction. A previous study also reported that oven drying at low temperature is suitable in preserving the antioxidant values among other dried leave methods such as shade and sun drying (Babu *et al*. 2018). As far as the authors know, there is no report on profiling to detect the presence of bioactive compounds of interest in the effect of different drying methods and ethanol compositions on quality attributes on jackfruit leaves extract.

Table 4. Artocarpin, Squalene, and β-sitosterol Content in JL Extract

Sample	Content (mg/g)		
	Artocarpin	Squalene	β-sitosterol
SD80	4.71 ± 0.59[b]	1.12 ± 0.09[b]	0.32 ± 0.02[a]
SN80	2.53 ± 0.29[c]	0.55 ± 0.06[c]	0.11 ± 0.01[b]
OV80	8.35 ± 1.78[a]	1.47 ± 0.01[a]	0.28 ± 0.02[a]

Note: [abc] Significant difference ($p < 0.05$) of one sample over the other(s) if values in the same column carry different superscript

CONCLUSIONS

1. Different drying treatments and ethanol/water ratios on extraction resulted in differences in color, total phenolic content (TPC), total flavonoid content (TFC), and the antioxidant activities, which were investigated individually.
2. Newton model proved to be the best fit model for describing drying behavior of drying treatments of jackfruit leaves (JL). The moisture effective diffusivity calculated from the second Fick's law was within the range 2.920×10^{-10} to 6.814×10^{-10} m^2/s over the temperature range studied.
3. Oven-drying with 80% ethanol/water (OV80) ratio provided highest antioxidant properties (phenolic and flavonoid contents as well as antioxidant activities) and acceptable color properties better than the conventional drying treatments.
4. Degradation in green colour can be considered as an index for loss of quality in product. However, minimal colour losses in oven-dried leaves do not have an impact on the antioxidant performance in this study. Overall, oven drying is worth to be considered for drying jackfruit leaves.

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