Effects of different pulsed vacuum drying strategies on drying kinetics, phenolic composition, and antioxidant capacity of chrysanthemum (*Imperial chrysanthemum*)

Huihuang Xu[1], Min Wu[1*], Tong Zhang[1], Fei Gao[2], Zhi'an Zheng[1], Yang Li[1]

(1. *College of Engineering, China Agricultural University, Beijing 100083, China*;
2. *School of Food and Health, Beijing Technology and Business University, Beijing 100048, China*)

Abstract: This work aimed to discuss effects of pulsed vacuum drying (PVD) at different temperatures (45 ℃, 50 ℃, 55 ℃ and 60 ℃), vacuum durations (5 min, 10 min and 15 min) and multi-stage heating on drying kinetics, colour attributes, phenolic compounds and antioxidant capacity of chrysanthemum (*Imperial chrysanthemum*). Results indicated that successive temperature increase reduced the drying time and enhanced the drying rate and moisture diffusivity. Lower temperature (45 ℃) and the multi-stage (35 ℃-55 ℃-60 ℃) drying presented the superiority in the protection of color, preservation of phytochemical composition (chlorogenic acid, luteolin, total phenolic and total flavonoid content) and improvement of antioxidant capacity (DPPH and FRAP) of chrysanthemum, which was attributed to the low-oxygen drying environment and reduction of thermal degradation losses. Based on the results of drying efficiency and drying quality, the multi-stage heating (35 ℃-55 ℃-60 ℃) has the excellent potential to produce high-quality dried chrysanthemum on a commercial scale.

Keywords: pulsed vacuum drying, drying kinetics, chlorogenic acid, luteolin, antioxidant capacity
DOI: 10.25165/j.ijabe.20221504.7359

Citation: Xu H H, Wu M, Zhang T, Gao F, Zheng Z A, Li Y. Effects of different pulsed vacuum drying strategies on drying kinetics, phenolic composition, and antioxidant capacity of chrysanthemum (*Imperial chrysanthemum*). Int J Agric & Biol Eng, 2022; 15(4): 236–242.

1 Introduction

Chrysanthemum (*Imperial chrysanthemum*) is one of the traditional herbal and medicinal plants belonging to the *Compositae family*, and it is primarily cultivated in most areas of China, including Jiangxi, Anhui and Qinghai. Chrysanthemum is highly-favored by health-conscious consumers due to its active ingredients, such as volatile oils, various amino acids and vitamins[1]. Studies have reported that these active ingredients have anti-inflammatory, antineoplastic, antidiabetic, antibacterial, and lipid-lowering functions[2].

Traditionally, dried flower head of chrysanthemum is typical as a healthy drink, and it has been popular with the public for thousands of years. However, fresh flower heads of chrysanthemums have high moisture content (MC), approximately 80% (wet basis, w.b.), which may lead to some adverse effects on product quality, such as rot and pest problems, enhancement of enzymatic and non-enzymatic reactions[3]. Drying is a fundamental way to prolong the storage mean while reducing the packaging and transportation costs of chrysanthemum by decreasing the moisture content to a safety limit.

Hot air drying (HAD) is a conventional method that has been extensively used in the food industry due to its advantages of simple operation and low investment[4]. However, many studies showed that it led to colour deterioration, radial and axial shrinkage, loss of active ingredients, and poor water holding capacity, which was due to the drying condition under high intensity for a long time[5]. Shi et al.[6] found that hot air drying resulted in the loss of active ingredients, such as total flavonoid, vitamin C and amino acid content, especially at high temperatures (75°C). Freeze-drying (FD) had the superiority of retaining active ingredients, colour, flavor and rehydration characteristics of dried products. However, its high energy consumption and production costs have restricted its development in agricultural product processing[7]. Therefore, freeze-drying cannot be considered a suitable choice for drying chrysanthemums with specific seasonal availability.

With the development and innovation of drying technology, pulsed vacuum drying (PVD) has become popular in the field of agricultural product processing. PVD exhibits a pulsating pressure until the safe moisture content is obtained under infrared heating[8]. Pulsed pressure causes a tunnelling effect that facilitates the formation of porous structures in the product matrix, which is conducive to moisture migration. In addition, during the vacuum stage of PVD, low pressure leads to a decrease in boiling point of the water. Furthermore, an oxygen-deficient environment prevents colour deterioration and the loss of bioactive components by inhibiting oxidative reactions[9]. Therefore, PVD can improve product quality and enhance drying efficiency. Study reported that porous structures were formed on the cross-section of red pepper dried by PVD, which appeared to improve the rehydration

Received date: 2022-01-20 Accepted date: 2022-04-25
Biographies: Huihuang Xu, Master candidate, research interests: agricultural engineering, Email: xhhhenan@163.com; **Tong Zhang**, Master candidate, research interests: agricultural engineering, Email: ziktlqh@163.com; **Fei Gao**, PhD, Lecturer, research interests: food science and engineering**; Zhi'an Zheng**, PhD, Associate Professor, research interests: agricultural engineering, Email: zhengza@cau.edu.cn; **Yang Li,** Master candidate, research interests: Mechanical engineering, Email: 905391412@qq.com.

***Corresponding author: Min Wu**, PhD, Assistant Professor, research interests: agricultural engineering, drying technology and engineering. College of Engineering, China Agricultural University; Engineering Research Center for Modern Agricultural Equipment & Facilities. P.O. Box 50, No.17 Qinghua East Road, Beijing 100083, China. Tel/Fax: +86-10-62736883, Email: minwu@cau.edu.cn.

capability and water holding capacity[10]. However, few reports focus on the application of vacuum pulse drying technology in the field of chrysanthemum processing.

In the current work, effects of drying temperature, vacuum duration and multi-stage heating on the drying kinetics and quality attributes of chrysanthemums were experimentally evaluated. These findings in present work would contribute to a better understanding of the effects of different drying technologies on drying kinetics, physicochemical properties of chrysanthemum, which are necessary to optimize the chrysanthemum drying process.

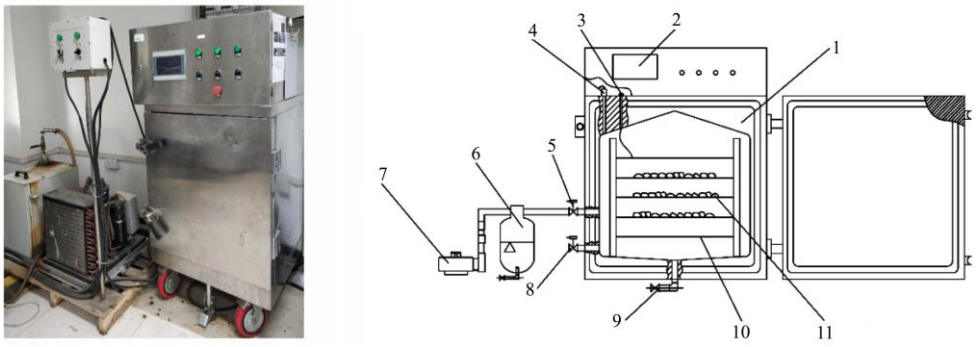
2 Materials and methods

2.1 Materials

Fresh chrysanthemum was obtained from Lushan, Jiangxi province of China and stored in a refrigerator at –4 ℃ before drying. Chrysanthemums with uniform colour and size, and no insects to rot, were manually selected as dry samples. The average diameter, weight, and thickness of chrysanthemum were approximately 50 mm, 80 g and 30 mm, respectively. The initial moisture content of flower heads was 82.86% (w.b.), measured by oven-drying at 105 ℃ for 24 h following the AOAC standard method[11].

2.2 Pulsed vacuum drying (PVD)

A laboratory-scale pulsed vacuum dryer was used in this research, located in the College of Engineering of China Agricultural University, Beijing, China (Figure 1). The equipment mainly consists of three parts: (1) a vacuum system composes of vacuum pump, condenser, air solenoid value to keep the pulsed vacuum condition; (2) a heating system which includes multiple carbon fiber heating infrared panels with a rated power of 250 W and a surface area of 50×50 cm^2; (3) an electronic control system used to adjust the temperature of heating panel and chamber pressure. A PID controller (Omron, model E5CN, Tokyo, Japan) is used to maintain the set chamber pressure and drying temperature with an accuracy of ±0.1 ℃.



a. The pulsed vacuum dryer b. Schematic diagram

1. Drying chamber 2. Touch screen 3. Electric heating panel temperature sensor 4. Pressure sensor 5. Vacuum valve 6. Air tight condenser 7. Vacuum pump 8. Air solenoid valve 9. Drain solenoid valve 10. Electric heating panel 11. Chrysanthemum

Figure 1 Physical and schematic diagram of the pulsed vacuum dryer

During the drying process (Figure 2), air is discharged from the drying chamber to a specified vacuum pressure (a) and maintained at this level for a set time (b). Immediately followed by pressure recovery (c) and was maintained for the expected time (d). The parameters set by the program depend on the material characteristics and operating conditions. P_1 and P_2 represent the highest and lowest pressures in the drying chamber, respectively.

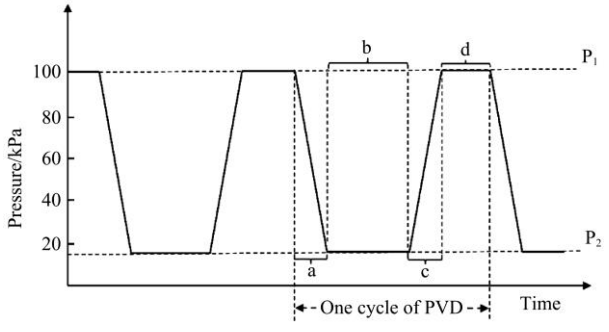


Figure 2 Schematic diagram of the PVD: air pressure (kPa) vs. time (min)

2.3 Drying experiment

The PVD drying experiments were conducted at different temperatures (45 ℃, 50 ℃, 55 ℃, and 60 ℃) and the pulse ratios (5:3, 10:3 and 15:3), respectively. In addition, a multi-stage heating test was carried out to study the drying behavior of the chrysanthemum. The chrysanthemum was spread into a single layer on four stainless steel trays with an initial weight of (1000 ± 50) g for each layer. The drying chamber door was opened at atmospheric pressure and the sample was weighed within 60 s. Drying experiments were finished until the sample moisture content decreased under 10% (w.b.). The experiments were performed in triplicates. Before each test, the dryer ran for 30 min to achieve steady-state conditions.

2.4 Drying characteristics analysis

2.4.1 Moisture ratio

The moisture ratio (*MR*) of chrysanthemum during drying was calculated using the following equation[12]:

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

where, M_t is the moisture content at a particular time *t*; M_o indicates the dry-based initial moisture content of chrysanthemum; M_e indicates the dry-based equilibrium moisture content.

When the values of M_e are comparatively small compared to M_t or M_o Equation (1) could be expressed in a simpler form as:

$$MR = \frac{M_t}{M_o} \quad (2)$$

2.4.2 Drying rate

The drying rate (*DR*) was used to indicate the speed of drying under different drying conditions, which could be calculated according to Equation (3):

$$DR = \frac{M_{t1} - M_{t2}}{t_2 - t_1} \quad (3)$$

where, *DR* indicates the drying rate, %/min; M_{t1} and M_{t2} are the dry-based moisture content, g/(g·min) at the drying times of t_1 and t_2, respectively, and $t_1 < t_2$, min.

2.5 Effective moisture diffusivity (D_{eff})

The drying process of the chrysanthemum could be described by the second Fick's law of diffusion, and the effective moisture diffusivity (D_{eff}) also was calculated by the following Equation (4):

$$MR = \frac{Mt}{Mo} \approx \frac{8}{\pi^2}\exp(-\frac{\pi^2 Deff}{H^2}t) \quad (4)$$

where, D_{eff} is the effective moisture diffusivity coefficient, m^2/s; H is the thickness of the chrysanthemum, with 30 mm as its value; t represents the time required for the drying process, s.

2.6 Color measurement

The surface colour of fresh and dried chrysanthemum was measured by a handheld colour reader (WR-10, Shenzhen Wave Optoelectronics Technology Co., Ltd, China). Colour was expressed in CIELAB, i.e., L^* (whiteness or brightness), a^* (redness or greenness), and b^* (yellowness or blueness) coordinates. Whiteness index and yellowness index are widely used to characterize the degree of discoloration during food thermal treatment. WI indicates the degree of whiteness, which can be calculated by Equation (5)[13]:

$$WI = \sqrt{(100 - L^*)^2 + a^{*2} + b^{*2}} \quad (5)$$

The yellowness index (YI) indicates the degree of yellowness.

$$YI = \frac{142.86b^*}{L^*} \quad (6)$$

2.7 Phytochemical contents determination

2.7.1 Determination of chlorogenic acid and luteolin

The determination of chlorogenic acid and luteolin were performed using a UPLC system consisted of an auto-injector and a binary bump. A column (2.1 × 100 mm, 1.8 μm, Waters Corp., Milford, USA) was used for chromatographic separation, with a mobile phases of solvents A (acetonitrile) and B (0.05% formic acid water, v/v), and the flow rate was set as 0.3 mL/min. The program of elution gradient was carried out as follows: 10% A-90% B at 0-0.5 min, 10% A-90% B at 0.5-5 min, 30% A-70% B at 5-9.5 min, 90% A–10% B at 9.5-11 min. the injection volume was 5 μL and the column temperature was 30 ℃.

MS was conducted as follows: the capillary voltage was +2900 V (positive mode) and –3100 V (negative mode), the ion source and the desolvation gas (N_2) were 120 ℃ and 400 ℃, with the flow rate of 50 L/h and 600 L/h, respectively. The flow of collision gas (Ar) was 0.07 mL/min, using a water triple quadrupole mass spectrometer with electrospray ionization (ESI) in multiple reaction mode (MRN). The of in positive and negative ESI mode were 2900 and 3100 V, respectively. The collected data was further used for quantification analysis, and expressed as mg per g of dry weight (DW).

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

Dried sample powder (100 mg) was blended with 10 mL of 80% methanol (v/v), followed by treating at 50 ℃ for 30 min in an ultrasonic water bath (40 kHz and 200 W). The suspension was then centrifuged at 10000 ×g for 15 min, the supernatant obtained was collected to determine TPC and TFC using the methods of Hodaei et al.[14]

For TPC, 0.1 mL extract was mixed with 1 mL Folin-Ciocalteau reagent and 2 mL Na_2CO_3 solution (15%, w/v). The obtained reaction was incubated for 30 min in a water bath at 50 ℃, then the absorbance was read at 750 nm using a UV-spectrophotometer (TU-1810, Beijing Puekinje General Instrument Co., Ltd, Beijing, China). TPC was expressed as mg equivalents of gallic acid, mg GAE/g DW.

For TFC, 1 mL of extract was added 0.5 mL of $NaNO_2$ (5%, w/v) and incubated for 5 min at room temperature, followed by adding the 0.5 mL of Al $(NO_3)_3$ (10%, w/v), then incubated for another 6 min. After that, 4 mL of 4% NaOH was added to the reaction and incubated for 15 min at room temperature. The reading of absorbance was performed at 510 nm using a UV-spectrophotometer. The data was expressed as mg equivalent of rutin, mg RE/g DW.

2.7.3 Determination of antioxidant capacity

The antioxidant capacity of samples was evaluated by DPPH and FRAP assays[15,16], and all results were expressed mg Trolox equivalent antioxidant capacity mg/g DW.

For DPPH assay, 0.8 mL extract diluent was mixed 3.2 mL DPPH solution (40 mg/L) and incubated in the dark for 30 min. The absorbance was read at 517 nm using UV spectrometer.

For the FRAP assay, 0.1 mL of extract diluent was blende with 3.9 mL FRAP solution and incubated for 10 min. The absorbance was determined using a spectrophotometer.

2.8 Statistical analysis

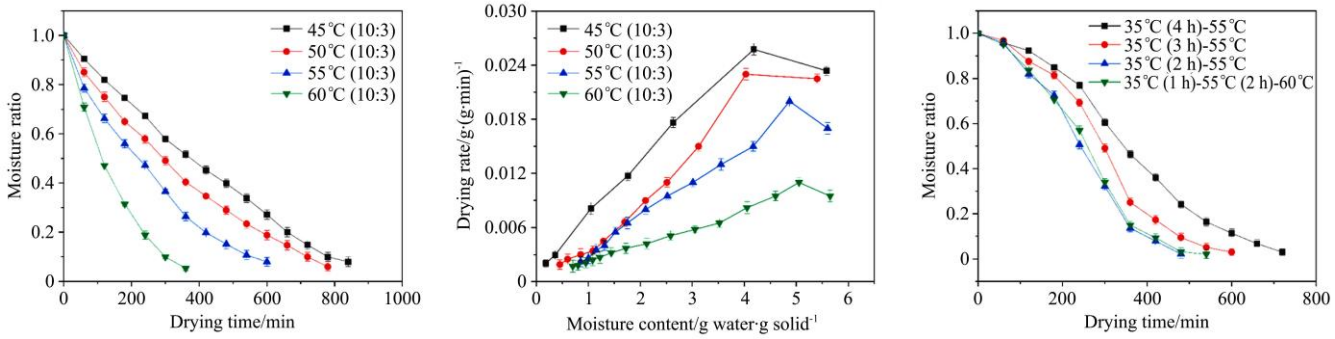
The experiment under different conditions was repeated three times, and results were expressed as the mean of three determinations ± standard deviation. The experimental data was processed and plotted using Origin 2021 software (Origin Lab, USA). The statistical analysis was completed by ANOVA and Duncan test (differences considered significant at p<0.05) using SPSS 20.0 (SPSS Inc., Chicago, IL, US).

3 Results and discussion

3.1 Drying characteristics

3.1.1 Drying kinetic curves

The changes in the moisture ratio of chrysanthemum under different drying conditions are depicted in Figures 3a, 3c and 3e. The time required for moisture ratio of the chrysanthemum from 82.86% (w.b.) to target moisture content (below 10%) gradually decreased with an increase in drying temperature at any drying conditions. For instance, the drying time of the chrysanthemum dropped from 840 min to 360 min when the drying temperature increased from 45 ℃ to 60 ℃. The time needed for sample dried at 60 ℃ was the shortest, reduced by approximately 55% compared to that at 45 ℃, suggesting drying temperature has a significant (p< 0.05) effect on shortening drying time, which can be attributed to the enhancement of the driving force for moisture migration[17]. Similar results were also observed from Figure 3c. The shorter the duration of the first heating stage (35 ℃), the shorter the time required for drying to the target moisture content. It was worth noting that the third stage of heating at 60 ℃ has no positive effect on the removal of moisture, compared to the two-stage heating (35 ℃ (2 h)-55 ℃), which can be because excessively high temperature has aggravated the hardening of the material surface and hindered the migration of moisture[18]. Furthermore, the atmospheric duration was set to 3 min, the moisture ratio of chrysanthemums with different vacuum durations (5, 10, and 15 min) varied with drying time, as shown in Figure 3e. As the vacuum duration increased from 5 to 15 min, the time required for chrysanthemums to reduce to the final moisture content decreased from 540 to 300 min. The time needed for chrysanthemums drying decreased with increasing vacuum duration. The time required for a vacuum duration of 15 min was the shortest, indicating that the increase in vacuum duration had a positive effect on the reduction in drying time.



a. Drying curves for chrysanthemum under different drying temperature

b. Drying rate for chrysanthemum under different drying temperature

c. Drying curves for chrysanthemum under different heating strategies

d. Drying rate for chrysanthemum under different heating strategies

e. Drying curves for chrysanthemum under different pulsed ratios

f. Drying rate for chrysanthemum under different pulsed ratios

Figure 3 Drying curves (a, c and e) and drying rate curves (b, d and f) under different drying conditions

3.1.2 Drying rate

From drying rate curves (Figures 3b, 3d and 3f), there was no constant rate drying stage during the entire drying process, and the overall trend was falling rate drying, except for the short-term rise in the early stage of PVD, because the material was in a preheated state. This indicated that moisture movement from the inside to the surface of chrysanthemum was governed by internal diffusion of moisture that was the dominant physical mechanisms[19]. Considering the effect of drying temperature on the drying rate, with the increasing temperature, the drying rate of PVD increased, which could be attributed to the promotion of moisture diffusion inside the material by high temperature. In addition, there were two inflection points in the drying rate curve of stage-heating drying, which may be linked to the enhancement of the partial pressure of water vapor inside the materials caused by the temperature increase. Furthermore, increasing the vacuum duration reduced the drying time. This phenomenon can be explained by the assumption that frequent pressure changes in the drying chamber may result in pulsating temperature changes that were not conducive to material heating, leading to a reduction in drying efficiency.

3.1.3 Effective moisture diffusivity

It is widely accepted that the effective moisture diffusion coefficient is one of the most significant parameters to describe a variety of mass transfer mechanisms[20]. Therefore, the effective moisture diffusion coefficient is closely bound up with the dehydration ability of the materials, which is crucial for the experimental design and optimization of the drying process. Table 1 lists the calculated diffusivity values of the practical moisture of chrysanthemums under different drying conditions. The drying conditions had a noticeable effect on the D_{eff} values. The effective diffusivity values of chrysanthemum varied in the range of (2.65-11.32)×10^{-7} m^2/s. As expected, the increase in temperature caused an increase in D_{eff} values due to the enhanced movement of water molecules. Moreover, the D_{eff} value of chrysanthemums dried by PVD at 60 ℃ (15:3) was the highest. This result agreed that higher temperature and longer vacuum duration could improve the drying rate and reduce the drying time. Therefore, it can be concluded that an appropriate temperature and vacuum duration can effectively improve the drying efficiency.

Table 1 Effective moisture diffusivity values of chrysanthemum under different drying conditions

Temperature/ ℃	Pulsed ratio	Linear regression equation	R^2	D_{eff} /10^{-7} m^2·s^{-1}
45	10:3	ln*MR* = -0.0029*t* + 0.2430	0.9437	2.65
50	10:3	ln*MR* = -0.0033*t* + 0.1750	0.9869	3.01
55	10:3	ln*MR* = -0.0042*t* + 0.1134	0.9868	3.83
60	10:3	ln*MR* = -0.0094*t* + 0.2609	0.9613	8.58
60	5:3	ln*MR* = -0.0057*t* + 0.1948	0.9892	5.20
60	15:3	ln*MR* = -0.0124*t* + 0.0660	0.9978	11.32
35 (4 h)-55	10:3	ln*MR* = -0.0041*t* + 0.4299	0.9150	3.74
35 (3 h)-55	10:3	ln*MR*= -0.0056*t* + 0.5097	0.8944	4.56
35 (2 h)-55	10:3	ln*MR*= -0.0076*t* + 0.6597	0.9062	6.94
35 (1 h)-55(2 h)-60	10:3	ln*MR*= -0.0076*t* + 0.6238	0.8596	6.94

3.2 Color change

For product quality evaluation, colour is always considered an essential sensorial indicator, which influences the choice of appropriate drying approaches and determines the economic value of the products. As shown in Table 2, five parameter values of samples dried by a variety of conditions varied from approximately 71.45 to 81.12, 0.61 to 3.2, 36.05 to 40.33, 40.72 to 49.44 and 63.49 to 80.49, respectively. Compared with freeze-dried samples, the effect of thermal treatment on the color parameters was reflected in the decrease of L^* values, and increases of a^* and b^* values. Considering lightness (L^*) value showed a trend of rising first and then falling with increasing temperature. This phenomenon may have resulted from the increasing temperature reducing the drying time, thus inhibiting colour deterioration.

However, excessive temperature enhanced the generation of browning reactions or degradation of pigments during drying[21]. Similar results also are observed during different vacuum durations, samples dried by PVD with a pulsed ratio of 15:3 presented the highest lightness value (79.52±0.96). In addition, stage-heating drying with a shorter drying time yielded brighter samples.

Table 2 Color parameters (L^*, a^* and b^*), white index (*WI*), yellowness index (*YI*) of chrysanthemum at different drying conditions

Drying method	Temperature/ ℃	Pulsed ratio	Colour parameters			*WI*	*YI*
			L^*	a^*	b^*		
Freeze-drying	-	-	81.12±1.65^{a}	1.57±0.21^{g}	36.05±0.67^{c}	40.72±1.02^{f}	63.49±0.57^{g}
Pulsed vacuum drying	45	10:3	74.09±0.28^{cd}	1.82±0.26^{f}	39.6±0.39^{a}	47.36±0.77^{bc}	76.36±0.29^{c}
	50	10:3	77.2±0.15^{b}	1.68±0.4^{g}	39±0.45^{ab}	45.21±1.12^{de}	72.17±0.2^{de}
	55	10:3	74.98±0.56^{c}	3.2±0.17^{a}	37.15±0.18^{c}	44.9±0.97^{e}	70.78±0.5^{e}
	60	10:3	74.42±0.78^{cd}	2.57±0.19^{e}	38.8±0.47^{b}	46.54±1.08^{cd}	74.48±0.89^{cd}
	60	5:3	73.58±0.84^{d}	2.76±0.13^{d}	39.5±0.24^{ab}	47.6±0.33^{bc}	76.69±0.17^{bc}
	60	15:3	79.52±0.96^{a}	0.61±0.06^{h}	36.63±0.37^{c}	41.97±0.59^{f}	65.81±0.35^{f}
	35 (4 h)-55	10:3	71.45±1.43^{e}	3.05±0.2^{g}	39.96±0.28^{ab}	49.21±0.82^{a}	79.9±2.15^{a}
	35 (3 h)-55	10:3	72.17±0.79^{e}	2.926±0.11^{c}	39.87±0.19^{ab}	48.71±1.2^{ab}	78.92±1.19^{ab}
	35 (2 h)-55	10:3	73.87±1.34^{cd}	2.49±0.23^{e}	39.34±0.17^{ab}	47.29±0.9^{bc}	76.08±2.01^{c}
	35 (1 h)-55(2 h)-60	10:3	71.58±0.88^{e}	3.11±0.24^{b}	40.33±0.11^{a}	49.44±0.73^{a}	80.49±0.26^{a}

Note: Values with the different letters within each column indicate significant differences (p<0.05).

In terms of redness and yellowness, drying conditions had a significant (p<0.05) influence on the a^* values but had little impact on the b^* values. As presented in Table 2, the red colour deepens as the temperature increases, which can be related to the Maillard reaction, because the chrysanthemum was packed with biomolecules such as amino acids and reducing sugars. They were sensitive to the moisture content and temperature. Thus, the thermal process contributed to a series of chemical reactions, resulting in a reddish-brown product.

Table 2 shows that the whiteness index values decreased while the yellowness index increased, indicating that chrysanthemum was sensitive to temperature. A similar phenomenon was observed by Kotwaliwale et al.[22] They found that the whiteness of mushrooms decreased while yellowness increased during drying, which may be related to the pigmentation in the products resulting from high temperature. Therefore, temperature was the critical element affecting chrysanthemum colour retention. In addition, the chrysanthemum dried at 60 ℃ with a pulse ratio of 15:3 had the lowest WI value, which illustrated that the reduction in drying time brought about minor thermal damage to chrysanthemum.

3.3 Effect of drying on phytochemicals content

3.3.1 Effect of drying on chlorogenic acid and luteolin

Chlorogenic acid and luteolin are important phenolic compounds in chrysanthemum, which can perform a variety of physiological functions in the human body. The content of chlorogenic acid and luteolin in samples dried under different conditions were showed in Figure 4, ranging from 36.47 to 53.34 mg/100 g DW and 4.83 to 22.51 mg/100 g DW. The PVD samples exhibited higher retention levels of bioactive compounds than freeze-dried samples. Furthermore, the increase in drying temperature caused a decrease in chlorogenic acid and luteolin content, which may be linked to the thermal degradation induced by the high temperature. Significantly, the multi-stage drying productively improved the retention level due to the phasic temperature rising from 35 ℃-60 ℃, especially obtained the highest chlorogenic acid content. For luteolin, the extension of the vacuum duration effectively increased the luteolin content, this may be ascribed to the reduction of pressure change frequency in the drying chamber. This contributed to establishing a relatively stable hypoxic environment, which inhibited the transformation, degradation, and hydrolysis[23]. Indeed, samples dried at a pulsation ratio of 15:3 possessed the highest luteolin content. In summary, the differences in chlorogenic acid and luteolin contents in the samples were associated with effect of drying conditions on the metabolic activity of chrysanthemum causing the accumulation or loss of phenolic compounds.

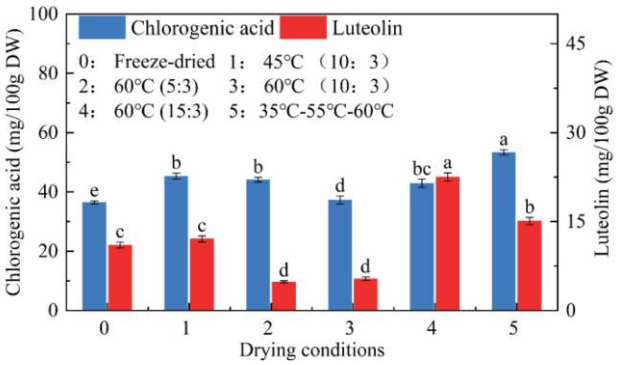


Figure 4 The content of chlorogenic acid and luteolin in samples under different drying conditions

3.3.2 Effect of drying on total phenolic content (TPC) and flavonoid content (TFC)

Table 3 lists the effects of the drying conditions on the TPC and TFC. Compared to freeze-dried samples, the TPC and TFC of dried chrysanthemum increased remarkably by 144.87% and 136.84%. TPC of dried samples at drying temperature of 45 ℃-60 ℃ ranged from 24.43 to 49.91 mg GAE/g DW. Vacuum duration time range of 5-15 min, caused TPC to increase from 40.07-45.03 mg GAE/g DW. Furthermore, considering the multi-stage drying, the reduction in the duration of the first stage resulted in an increase in TPC from 38.67-53.75 GAE/g DW. In addition, the drying treatment significantly increased the retention of TFC compared to the freeze-dried treatment. Particularly, at drying temperature of 40 ℃-60 ℃, TFC increased by 50.9%-136.84%. In contrast, with the increase in vacuum duration time, TPC exhibited an initial rise followed by a decrease. A similar phenomenon was observed in the effect of the reduction in the duration of the first stage on the change in TFC.

The distinct increase in TPC may be attributed to the change in pulse pressure leading to the mechanical rupture of cell membranes and loosening of the pectin network in the cell wall, which increases extractability[24]. Similarly, pulsed pressure can also lead to the breakdown of covalent bonds, which is conducive to increasing the accessibility of flavonoids. Furthermore, TPC and

TFC did not exhibit regular trends with an increase in drying temperature. This phenomenon may be attributed to two factors. On one hand, increasing the drying temperature can result in the thermal degradation of heat-liable phenolic compounds, but on the other hand, which also shortened drying time and improved the retention of phenolic compounds. Therefore, a moderate temperature, such as 55 ℃, will induce higher TPC and TFC compared with 45 ℃, 50 ℃ and 60 ℃. Overall, it is essential to consider the synergistic effect of temperature and drying time to minimize the loss of phenolic compounds.

Table 3 Total phenolic content (TPC), total flavonoid content (TFC), DPPH and FRAP in chrysanthemum at different drying conditions

Drying method	Temperature/ ℃	Pulsed ratio	TPC/mg GAE (g DW)$^{-1}$	TFC/mg RE (g DW)$^{-1}$	DPPH/mg Trolox (g DW)$^{-1}$	FRAP/mg Trolox (g DW)$^{-1}$
Freeze-drying	-	-	21.95±0.62^{f}	27.88±0.43^{g}	2.18±0.08^{f}	7.24±0.72^{e}
Pulsed vacuum drying	45	10:3	49.91±0.68^{b}	66.03±0.49^{a}	9.87±0.28^{a}	18.69±0.26^{a}
	50	10:3	24.43±2.66^{f}	54.30±1.71^{c}	9.09±0.32^{b}	14.90±0.05^{d}
	55	10:3	43.15±0.06^{cd}	55.98±0.06^{bc}	7.96±0.22^{c}	18.88±0.05^{a}
	60	10:3	40.23±1.70^{de}	51.84±0.67^{d}	7.23±0.01^{de}	14.71±0.51^{d}
	60	5:3	40.07±1.58^{de}	54.56±0.61^{c}	7.57±0.22^{cd}	17.63±0.29^{ab}
	60	15:3	45.03±1.47^{c}	56.07±1.52^{bc}	7.23±0.05^{de}	15.89±0.72^{cd}
	35(1 h)-55(2 h)-60	10:3	53.75±0.11^{a}	51.97±0.85^{d}	9.34±0.10^{b}	18.00±1.15^{ab}
	35 (2 h)-55	10:3	45.31±1.75^{c}	56.59±0.06^{b}	9.93±0.30^{a}	17.32±1.30^{abc}
	35 (3 h)-55	10:3	42.95±0.00^{cd}	46.41±0.06^{f}	7.37±0.30^{de}	16.70±0.29^{bc}
	35 (4 h)-55	10:3	38.67±0.85^{e}	48.31±0.06^{e}	7.02±0.31^{e}	14.25±1.01^{d}

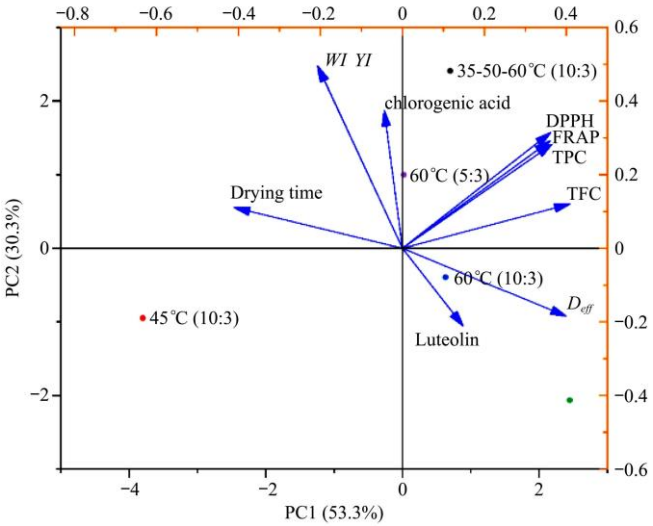
Note: Values with the different letters within each column indicate significant differences (p<0.05).

3.4 Effect of drying on antioxidant capacity

The antioxidant capacity of dried chrysanthemum evaluated using DPPH and FRAP assays is shown in Table 3. Drying significantly enhanced the antioxidant capacities of the samples. At temperature range of 45 ℃-60 ℃, the DPPH and FRAP values were 7.23-9.87 mg Trolox/g DW and 7.24-18.88 mg Trolox/g DW. The temperature increase had a negative effect on antioxidant capacity, which may be attributed to the oxidation and degradation of phenolic compounds with antioxidant potential caused by high temperatures. Furthermore, an increase in the duration of the first stage also resulted in a decrease in antioxidant capacity. This suggested high temperature and long drying time were not conducive to the improvement of antioxidant capacity. Unexpectedly, the extension of the vacuum duration had no positive effect on the enhancement of the antioxidant ability. This phenomenon can be ascribed to the fact that, the rapid removal of moisture reduces the accumulation of Maillard reaction products (MRPs), while most MRPs possessed strong antioxidant capacity. Similar results were reported by Manzocco et al.[25], who found that the antioxidant ability was enhanced due to the increase in MRP. Significantly, the antioxidant capacity (FRAP) of chrysanthemum was positively (p<0.05) correlated with TPC (r=0.79) and TFC (r=0.88) under different drying conditions, indicating that phenolic compounds played a key role in the improvement of antioxidant capacity.

3.5 Principal components analysis (PCA)

The current research showed that drying conditions have a significant influence on the quality of chrysanthemums. Thus, principal component analysis was carried out to further elucidate the relationship between drying conditions and the drying quality of dried chrysanthemum. Two main components with eigenvalues greater than 1, regarded as significant according to the Kaiser criterion were selected and are shown in Figure 5. From the PCA score plot, two principal components (PC1 and PC2) explained 83.6% of the total variance. PC1 was positively correlated with DPPH, FRAP, TPC, TFC D_{eff} and luteolin, accounting for 53.3% of the total variation. PC2 was positively correlated with *WI*, *YI*, drying time and chlorogenic acid, explaining 30.3% of the total variance. Drying at 60 ℃ for pulsation ratios of 10:3 and 15:3 showed positive PC1 indicating high values of D_{eff} and luteolin. Multi-stage heating (10:3) presented positive PC1 scores, suggesting higher DPPH, FRAP, TPC and TFC values. On the positive axis of PC2, drying at 60 ℃ for pulsation ratio of 5:3 showed a positive correlation with *WI*, *YI*, drying time and chlorogenic acid values, suggesting this condition was more conducive to improving their properties.



Note: TFC: total flavonoid content; TPC: total phenolic content.

Figure 5 Principal component analysis of the first two components (PC1 and PC2) based on drying kinetics, color attributes, and bioactive compounds of dried chrysanthemum obtained from different drying methods

4 Conclusions

The current work evaluated the effects of drying temperature, vacuum duration, and multi-stage heating on the drying characteristics, color attributes, phenolic compounds and antioxidant capacity of the chrysanthemum. Results indicated that the overall quality of dried chrysanthemum was governed by the combined effect of drying temperature and time. Increasing drying temperature and the vacuum duration or using multi-stage heating could effectively reduce drying time. For the protection of color attributes, higher temperature caused more severe color deterioration of chrysanthemum. Furthermore, considering the

retention level of bioactive composition, low temperature (45 ℃) and multi-stage heating (35 ℃-55 ℃-60 ℃) exhibited more significant superiority due to the reduction of thermal degradation losses, causing satisfying antioxidant capacity. Overall, this work provided an essential theoretical basis to optimize the drying process of chrysanthemum.

Acknowledgements

This work was supported by Chinese Universities Scientific Fund (2022TC079), China Agricultural Research System of MOF and MARA (CARS-21), National Natural Science Foundation of China (No. 32171912) and Beijing Municipal Natural Science Foundation (No. 6182022).

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