Hot Air Drying Combined with Intermediate Microwave Treatment Improving Production Efficiency and Product Quality of Jin Yin Hua

Lifen Li, Zhihua Liu, Xuesen Wen * School of Pharmaceutical Sciences Shandong University Jinan 250012, China

Abstract—The flower of Lonicera japonica Thunb., Jin Yin Hua in Chinese, is a commonly used health food and herbal medicine in many Asian countries. In this paper, the better time for microwave treatment combined with hot air drying was investigated to overcome the drawbacks of traditional drying process based on the appearance, color, drying characteristic, chlorogenic acid content, PPO activity and antioxidation of the resulting flowers. The results showed that the flowers with good appearance, high chlorogenic acid content and antioxidation were obtained by 2-min microwave processing after drying at 45°C to 50% weight loss, and then drying at 80°C, in addition, this process could also save more than 65% drying time comparing with drying at 45°C alone. Therefore, this drying process was suggested for large-scale drying of Jin Yin Hua.

Keywords—Lonicera japonica, hot air drying microwave, chlorogenic acid, antioxidation

I. INTRODUCTION

The dried flower of *Lonicera japonica* Thunb. (Caprifoliaceae) is called Jin Yin Hua in Chinese. It has been used as a traditional herbal medicine and health food in China and other Asian countries for more than 3000 years with prominent anti-inflammatory, antipyretic, and bactericidal effects[1].

Traditionally the material is dried under sun or in the shade, however, these methods are outdated for low efficiency and poor product quality. There are several reports on Jin Yin Hua drying in literatures, such as hot air drying, vacuum drying and freeze drying, or hot-air drying associated with pretreatment by microwave, steaming or baking[2-4]. Among them, Hot air drying is still popularly applied in large-scale production of Jin Yin Hua recently when whole equipment cost, drying efficiency and product quality are taken into account. However, hot air drying will still take a long time to avoid phenolic acid oxidation by its endogenous polyphenol oxidases [5]. Previously, we found that the flower must be dried at a temperature below 55°C to avoid browning, this will take more than 20 h, in addition, once became damp during storage, the resulting dried flowers would turn brown rapidly [6].

National Key Technology Research and Development Program from the Ministry of Science and Technology of China (2011BAI06B01). Microwave drying has already been applied to many vegetables and fruits for the benefit of short drying time, high active constituent and inactivating polyphenol oxidase[4], however, microwave drying alone has some major drawbacks including uneven heating, possible textural damage, and limited product penetration of the microwave radiation into the product [7-9].

In order to improve the quality of Jin Yin Hua and enhance its drying productivity, both steam and microwave blanching were applied to quickly inactivate polyphenol oxidase, and then a higher temperature drying were followed to shorten the drying time in our preliminary experiment. However, the dried flower lost its natural shape and color, and becoming slender and stiff due to the collapse and shrinkage of cells, thus the product was rejected by the market although it has a high level of chlorogenic acid. To overcome some drawbacks of hot air drying, microwave drying or drying combined with steam or microwave pretreatment, intermediate microwave treatment during hot air drying may be promising, which could accelerate drying process while presenting quality[9]. To our knowledge, there was no such research on microwave combined hot air drying of Jin Yin Hua. In this paper, the right time of microwave application during the hot air drying was investigated.

II. MATERIALS AND METHODS

A. Materials and Chemicals

Fresh flowers of *L. japonica* Thunb. were picked up from medicinal plant garden of Shandong University in Jinan, China, and conserved at 4 °C for subsequent experiments. The average dry matter content was $20.0 \pm$ 1.0 % determined by drying at 105°C until a constant weight obtained.

Chromatographically pure methanoic acid, acetonitrile and methanol were obtained from Kemiou Chemical Reagents Development Center (Tianjin, China). Catechol was purchased from Guangfu fine chemical research institute (Tianjin, China). Diphenyl bitter benzene hydrazine (DPPH) was bought from Aigma-Aldrich. Co (USA). The other chemicals of analytic grade were obtained from local commercial source.

B. Drying Apparatus

A hot air drying apparatus was used, which was consisted of four parts, an axial fan, a heating chamber, a drying chamber and a weighing system [10]. Sample weight was automatically recorded by a computer through communication with an electronic balance at an interval of 1min. A stainless steel mesh sample basket (25 cm \times 25 cm) was suspended from the balance (0.01 g). Air velocity was measured using an anemometer and adjusted by an electron speed regulator. Drying temperature was automatically controlled by a PID controller.

C. Drying Procedure

The flower buds were allowed to acclimate to room temperature before each drying experiment. At the beginning of each experiment, the dryer was adjusted to the indicated air velocity (0.5m/s) and temperature (45° C or 80° C). About 200 g samples were spread on the sample basket and firstly dried at a temperature of 45° C until lost 0, 10, 20, 30, 40, 50 or 60 percent of their initial weight, and then treated with microwave (G70D20Asp-DF Galanz microwave oven) for 2 min, finally dried at 80°C until the flower weight unchanged. Those dried at a constant temperature of 45 or 50°C, and the traditionally sun dried, were used as controls.

D. Drying Characteristics

Moisture ratio (MR) was used to describe the drying process, which were calculated as the following equation:

$$MR = \frac{M_t - M_e}{M_0 - M_e} \tag{1}$$

Where: M_t , M_0 , and M_e are the moisture content at the time of *t*, initial and finally equilibrium state (g water/g dry matter), respectively. The value of M_e is relatively small compared to M_t or M_0 for a long time drying; hence, the MR is simplified to be equal to M_t/M_0 [11].

E. Assay of PPO Activity

PPO activity was determined by an ultraviolet and visible spectrophotometer (Puxitongyong, Beijing, China) using catechol as substrate [5]. About 0.2 g powder of dried samples was mixed with 10 ml 0.1 M PBS, stored at 4° C for 4 h, then centrifuged at 1500 g under 4° C for 5 min. The supernatant was used for the PPO activity determination. A aliquot of 200 µl supernatant was mixed with 0.8 ml 0.1 M PBS and 1 ml 0.1 M catechol, then the absorbance at the wavelength of 410 nm was recorded at a 10 second interval from 20 to 60 second. One enzyme activity unit of PPO was defined as a 0.01 increase of the absorbance per one gram dried flower among one minute.

F. Determination of Chlorogenic Acid

HPLC method was applied to determine the content of chlorogenic acid, in accordance with our primary method[6]. The dried flower samples were ground into powder and pass through a 60 mesh. About 0.2 g fine powder was mixed with 10 ml 50% methanol and ultrasonically extracted for 20 min, then centrifuged at 4000 rpm for 5 min. The exaction was repeated for two times and the combined supernatant was diluted with water to 25 mL for HPLC determination. Chromatographic separation was achieved using an ODS C18 column (250×4.6 mm, 5 µm, Phenomenex, USA) on a Simadzu LC-10ATvp chromatographic system. Data were acquired and processed using N2000 chromatography software (Zhejiang University, China) [6].

G. Assay of Antioxidant Activity

The above extract was diluted with water for 50 times, which was mixed with equal volume of 0.048 g/L DPPH to evaluate the antioxidant activity of flower samples according to Yang etal. [13]

H. Colour Measurement

CIE colorimetric system was used to analyse the flower colour. The fine powers were scanned by a WF32 colorimeter (Shenzhen wave optoelectronics technology co. Ltd) to read three color parameters, Lightness (L), Redness-greenness (a) and Yellowness-blueness (b). The color alterations comparing with the samples dried under 45°C was estimated by the following equation:

$$\Delta E = \sqrt{(L_i^* - L_0^*)^2 + (a_i^* - a_0^*)^2 + (b_i^* - b_0^*)^2}$$
(2)

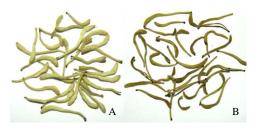
Where ΔE is color change, L_0^* , a_0^* and b_0^* are indexes of samples dried at 45°C, while the others are from different treated samples.

I. Statistical analysis

Results were expressed as mean \pm SD of values from three independent experiments.

III. RESULTS AND DISCUSSION

A. Appearance and Color Analysis of Dried Jin Yin Hua



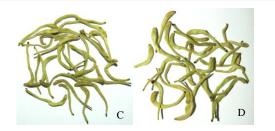


Fig. 1. Jin Yin Hua dried at different conditions. (A) dried at 45° C; (B, C and D) dried at 45° C to weight loss of 0%, 30% and 50%, respectively, followed by microwave processing for 2 min, and then dried at 80°C.

The flowers dried at 45°C are the most popular form in Chinese market, especially used as a health tea, for the appearance like fresh ones, however, those pretreated by microwave became slender and stiff after dried at 80°C as shown in Fig 1A and B. While drying at 45°C to different degrees (weight loss from 10% to 60%) followed by microwave processing and drying at a higher temperature could substantially improve the appearance of the produce, especially those with weight lost up to 50% as shown in Fig 1D.

From the point of color analysis, the higher positive L* value tends to more white color, positive a* value means more red color and less green color, positive b* means more yellow color than blue color, and the greater ΔE value, the bigger difference against the flower dried at 45°C. The values of L*, a* and b* of flowers dried at 45°C is 8.854 ± 0.166 , -0.1 ± 0.069 and 7.398 ± 0.116 , respectively. The traditionally sun-dried sample was the worst one, by contrast, color indexes of those hot air dried combined with intermediate microwave processing greatly improved as shown in Fig 2. The better times for microwave processing seems when drying at 45°C to weight loss up to 40%-60% according to the results of color analysis.

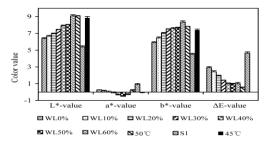


Fig. 2. Color analysis of various dried Jin Yin Hua (WL0%-WL60%: dried at 45°C to weight loss of 0% to 60%, respectively, followed by microwave processing for 2 min, and then dried at 80°C; 45°C and 50°C: dried at 45°C and 50°C; S1: dried in the sun.)

B. Drying Characteristics of Jin Yin Hua

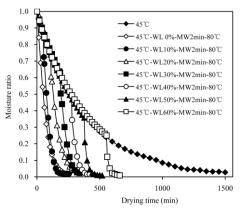


Fig. 3. Thin-layer drying curves of Jin Yin Hua (dried at 45°C to weight loss of 0% to 60%, respectively, followed by microwave processing for 2 min, and then dried at 80°C.)

As shown in Fig 3, drying at 45°C is a falling rate drying and takes a much long time. When combined with a microwave processing and then drying at 80°C, the process could substantially save the drying time. For example, more than 65% drying time could be saved when microwave applied at the time of weight loss 50%.

C. Microwave Processing Effect on Chlorogenic Acid Content in Jin Yin Hua

Chlorogenic acid content was determined to reflect the quality of dried Jin Yin Hua. The results are as shown in Fig. 4. Comparing with drying at 45°C alone, the drying protocol hot air drying combined with intermediate microwave processing can significantly increase the content of chlorogenic acid in Jin Yin Hua (p<0.001). As the result of color analysis, chlorogenic acid determination proved again that the quality of traditionally sun-dried Jin Yin Hua was poor.

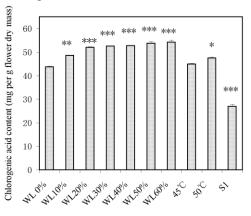


Fig. 4. The chlorogenic acid content of Jin Yin Hua dried under different conditions (WL0%-WL60%: dried at 45°C to weight loss of 0% to 60%, respectively, followed by microwave processing for 2 min, and then dried at 80°C; 45°C and 50°C: dried at 45°C and 50°C; S1: dried in the sun;, *, ** and *** indicate that the difference of chlorogenic acid content

comparing with that in samples dried at 45°C is significant determined by Student's t-test at the level of p<0.05; p<0.01 and p<0.001, respectively.).

D. Microwave Processing Effect on PPO Activity in Jin Yin Hua

PPO activity in Jin Yin Hua was largely inactivated by 2-min microwave processing regardless of its application moment, while most of its activity was reserved during drying at 45 or 50°C, or dried in the sun as shown in Fig. 5. PPO inactivation should be mainly, or at least partly, responsible for high content of chlorogenic acid and bright appearance in the flowers treated with microwave, and the processing will also make the produce stable during storage or extraction.

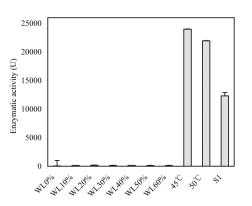


Fig. 5. PPO activity in Jin Yin Hua dried at different conditions (WL0%-WL60%: dried at 45°C to weight loss of 0%-60%, respectively, followed by microwave processing for 2 min, and then dried at 80°C; 45°C and 50°C; dried at 45°C and 50°C; S1: dried in the sun).

E. Antioxidation of Jin Yin Hua

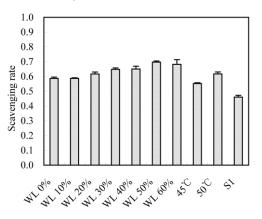


Fig. 6. Antioxidation of Jin Yin Hua dried under different conditions
(WL0%-WL60%: dried at 45°C to weight loss of 0% to 60%, respectively, followed by microwave processing for 2 min, and then dried at 80°C; 45°C and 50°C: dried at 45°C and 50°C; S1: dried in the sun).

Scavenging ability of organic free radical DPPH was used to reflect the antioxidation of the flowers dried at different conditions in this study. As shown in Fig 6, the DPPH scavenging rates of flowers with microwave treatment were all greater than that of flowers dried at 45°C, especially the flowers which lost 30%-60% of their initial weight when treated with microwave. The results shows a little difference from chlorogenic acid content in Fig 4, which indicates that there are still other active constituents related with anti-oxidative ability as Yang *et al.*, reported that flavonoids also accounted for its anti-oxidative ability[14]. The influence of hot air drying and microwave treatment on flavonoids remains further investigation

IV. CONCLUSIONS

In this study, microwave treatment in combination with hot air drying was applied to Jin Yin Hua drying, the right time for microwave application was optimized based on its effects on the appearance, color, drying characteristic, chlorogenic acid content, PPO activity and antioxidation of the resulting flowers. In summary, to enhance productivity and obtain Jin Yin Hua of high quality, 2-min microwave processing after drying at 45° C to 50% weight loss, and then drying at 80°C was suggested for large-scale drying.

REFERENCES

- X. Shang, H. Pan, M. Li, X. Miao, H. Ding, "Lonicera japonica Thunb.: ethnopharmacology, phytochemistry and pharmacology of an important traditional Chinese medicine," J Ethnopharmacol, 138, 1, 1-21 2011.
- [2] H. Qi, H. Sheng, C. Zhang, "Effects of Different Drying Methods on Quality of Flos Lonicerace," China Pharmaceuticals, 14, 024, 2010.
- [3] J. Li, J. J. Yang, "Determination of processing methods of Lonicerae Japonicae Flos in production place," Journal of Xinyang Agricultural College, 3, 041, 2011.
- [4] D. J. Chen, "Study on Effect of Drying Method on the Quality of Honeysuckle," Food Science, 11, 062, 2006.
- [5] N. N. Liu, W. Liu, D. J. Wang, Y. B. Zhou, X.J. Lin, X. Wang, S. B. Li, "Purification and partial characterization of polyphenol oxidase from the flower buds of Lonicera japonica Thunb.," Food Chemistry, 138, 478-483, 2013.
- [6] Z. H. Liu, X. S. Wen, G. D. Wei, Y. Q. Zhang, "Effect of steam blanching on drying characteristics of Jin Yin Hua, the flower bud of Lonicera japonica thunb," International Conference on Medicine Sciences and Bioengineering (ICMSB), 743-747, 2015.
- [7] M. Zhang, J. Tang, A. S. Mujumdar, S. Wang, "Trends in microwave-related drying of fruits and vegetables," Trends in Food Science & Technology, 17, 10, 524-534, 2006.
- [8] V. Sagar, P. S. Kumar, "Recent advances in drying and dehydration of fruits and vegetables: a review," Journal of Food Science and Technology, 47, 1, 15-26, 2010.
- [9] S. Chandrasekaran, S. Ramanathan, T. Basak, "Microwave food processing—A review," Food Research International, 52, 1, 243-261, 2013.
- [10] B. M. Zhu, X. S. Wen, G. D. Wei, "Effect of pre-treatments on drying characteristics of Chinese jujube (Zizyphus jujuba Miller)," International Journal of Agricultural and Biological Engineering, 7, 94-102, 2014.
- [11] Z. Erbay, F. Icier, "A review of thin layer drying of foods: theory, modeling, and experimental results," Critical Reviews in Food Science and Nutrition, 50, 5, 441-464, 2010.
- [12] H. Wang, X. D. Gao, G. C. Zhou, L. Cai, W. B. Yao, "In vitro and in vivo antioxidant activity of aqueous extract from Choerospondias axillaris fruit," Food Chemistry, 106, 3, 888-895, 2008.
- [13] X. J. Yang, S. Y. Duan, L. L. Song, "Extraction of Total Flavonoids from FLOS LONICERAE JAPONICAE and Its Oxidation Resistance," Journal of Anhui Agricultural Science, 40, 12, 7047-7049, 2012.