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Drying Characteristics and Product Quality of Lemon Slices Dried with Hot Air Circulation Oven and Hybrid Heatpump Dryers

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Abstract - In this research, drying characteristics and product quality of Coulomb-force-assisted heatpump and oven dried lemon slices were studied. Lemon slices with 3 mm thickness each, were dried using oven and Coulomb-force-assisted-heatpump dryer with and without auxiliary heater at different drying conditions. It was found that the drying rate of the lemon slices dried by all drying methods showed only falling rate states, which indicates the drying kinetics were controlled by internal moisture diffusion. Oven drying of lemon slices at 60°C showed the highest drying rate among all, followed by oven dried slices at 50°C, Coulomb-force-heater-assisted-heatpump (CF-HT-HP) dried slices at 31°C, Coulomb-force-assisted-heatpump (CF-HP) dried slices at 22°C, oven dried slices at 40°C and heatpump dried slices at 22°C. The average effective moisture diffusivity value for the slices dried with these drying methods was found in the range of 16.2 to 63.8 × 10⁻⁴ mm²/min⁻¹. In terms of quality assessment, CF-HP dried lemon slices retained the highest amount of Vitamin C as compared to the lemon slices dried by other drying methods. However, it retained relatively lower amount of total phenolic content (TPC) as compared to oven dried products. Among of all, CF-HP drying method produced dried lemon slices with the highest Vitamin C (6.74 mg AA / g dry weight) whereas oven dried lemon slices at 50°C preserved most of the TPC in the dried slices, which recorded as 13.76 mg GA / g dry weight.

Keywords: Hybrid heat pump drying; Coulomb force; drying rate; Vitamin C; Total Phenolic Content (TPC)

1. INTRODUCTION

Lemons (Citrus Limon) are nutritious fruit with a myriad of health benefits. They boast one of the nature’s highest Vitamin C concentrations, total phenolic content (TPC) and a unique flavour and aroma (Santos and Silva, 2008). Vitamin C is essential for a strong immune system and hence prevents the illness such as flu, colds and recurrent ear infections (Santos and Silva, 2008). Moreover, researches have shown that consumption of fruits with high Vitamin C can lower one’s risk of heart disease and strokes and reduce the symptoms of arthritis (Frei et al, 2012). On the other hand, TPC in lemons demonstrates plenty of biological properties including anti-allergenicity, anti-mutation effects, anti-atherogeneity, anti-inflammatory, anti-microbial, anti-thrombotic, cardioprotective and vasodilatory actions (Ramful et al., 2011). In addition, phenolic acids such as chlorogenic acid, syringic acid and vanillic acid are also claimed to comprise cytoprotective
ability in the prevention of diabetic neuropathy complication (Galuppo et al., 2014).

Ripe lemons contain 90% of water. However, high water content causes the growth of microbial which eventually shortens the overall shelf life of the lemons. Therefore, drying works as a preservation method by removing the water from the lemons in order to extend its shelf life. Thus far, there are various drying methods which have been used for drying of lemon peels and slices such as closed type solar drying, infrared drying, open sun drying, hot air drying, and microwave drying (Chen et al., 2005; Swanson, 2009). Yet, studies conducted to investigate the appropriateness of these drying methods in drying of lemon slices are limited. The quality attributes of dried lemon slices in term of bioactive ingredient were not investigated in detail although the drying characteristics were determined.

Lemon slices dried by conventional methods such as open sun drying and hot air drying tends to produce dried products with low quality especially in term of active ingredients (Chen et al., 2005; Chin et al., 2009). Open sun drying required relative low operating cost. Yet, it is weather dependent and tends to produce low quality products due to stretched drying time. Contamination and losses caused by insect infection or diseases may occur as well (Chen et al., 2005). Alternatively, hot air drying is typically operated at an elevated drying temperature to enhance the drying rate. Nevertheless, this drying method requires great amount of energy for drying process and tends to produce low quality dried products especially in terms of the retention of heat-sensitive bioactive ingredients (Chin et al., 2009).

Furthermore, volumetric heating from microwave drying allows the transmitted heat energy to be absorbed by the whole lemon slices instead of its surface. This causes the great improvement in drying rate and effective moisture diffusivity and thus, shortens the total drying time. However, the drawback of this method is the loss of the precious bioactive ingredients such as Vitamin C and TPC of the lemon slices. The heat energy generated within the drying sample will greatly deteriorate the Vitamin C and TPC in dried lemon slices as they are heat sensitive and volatile bioactive ingredients (Darvishi et al., 2012; Darvishi et al., 2014; Ghanem et al., 2012).

In this research, Coulomb-force-assisted-heatpump (CF-HP) drying is proposed as an alternative drying strategy for the drying of lemon slices. In CF-HP drying, a high voltage plate (15 kV, 50 Hz) is incorporated in the heatpump dryer in order to enhance the removal rate of bound moisture, which in turn counteracts the long drying time required by heatpump dryer due to mild temperature drying. The working principle of the microwave drying shows that water dipoles are capable to re-orient themselves in the rapid reversal of electromagnetic field (the frequency of electromagnetic field can be ranged from 300 MHz to 3000 MHz) (Bradshaw et al., 2011). As CF-HP drying was conducted at low frequency; water molecules could be polarized almost instantly and heat generation due to rapid re-orientation of water molecules could be minimized. Owing to the bipolar property of moisture content inside the lemon slices, a positive net force (Coulomb force) can be induced when the lemon slices are placed near to the high voltage (15 kV), but low frequency (50 Hz) plate. The generated force stimulates the moisture diffusion in the lemon slices which consequently dried by convective air flow produced by heatpump system. The quality of the drying material can be preserved as this CF-HP drying operates at mild drying temperature, low relative humidity and relatively high drying rate. Hence, CF-HP drying appears to be an advanced drying technology which is envisaged to enhance the overall drying kinetics of lemon slices, while at the same time producing dried lemon slices with high retention of antioxidants.

2. MATERIALS AND METHODS

2.1 Sample Preparation

Fresh Eureka lemons were sliced crosswise into circular slices of thickness 3.0 ± 0.5 mm and initial weight of 8.200 ± 0.100 g. A sharp knife was used so that lemons could be sliced evenly. The seeds were removed prior to the drying process.

2.2 Drying Procedures

Hot air circulation oven (Memmert 100-800) and hybrid heatpump dryer were employed to study the drying process of the lemon slices. Hybrid heatpump drying system was constructed as shown in Fig. 1. The drying performance of heatpump dryer was evaluated before it was integrated with high voltage plate and auxiliary heater to form a hybrid drying system.

2.2.1 Oven Drying

Oven was switched on and adjusted to the selected drying temperature for 30 minutes prior to the start of the experiments in order to produce drying air with stable temperature. The slices were dried at three different temperatures which were 40.0 ± 0.5 °C, 50.0 ± 0.5 °C and 60 ± 0.5 °C, at the corresponding relative humidity (RH) of 33.4 %, 20.2 % and 12.6 %, respectively. Lemon slices were placed on aluminium foil before being loaded into the oven.

2.2.2 Hybrid Heatpump Drying

Hybrid heatpump dryer was switched on for 30 minutes prior to the start of the experiments to produce drying air with stable drying condition. The average drying temperature, relative humidity, and air velocity inside the drying chamber were recorded as 22.0 ± 1.0 °C, 34.0 ± 0.5 %, and 1.1 ± 0.1 ms⁻¹, respectively when the auxiliary heater was turned off. When the auxiliary heater was turned on, the average drying temperature and relative humidity inside the drying chamber were recorded as 31.0 ± 1.0 °C and 24.0 ± 0.5 %, respectively with average air velocity remained the same. The lemon slices were placed on string mesh before being rested on the high voltage plate.
in the drying chamber. The thickness of the string meshes used was 3.0 mm.

In both drying processes, the weight of the slices was recorded using precision balance (A&D-GX1000, with precision of 0.001 g) at 5 minutes interval for the first 30 minutes, 10 minutes interval for the next hour, 30 minutes interval for the following 3 hours, and 1 hour interval for the rest of drying period. The drying processes were terminated when the slices achieved equilibrium moisture content (EMC), which is indicated by constant weight of the dried slices. The drying experiment was conducted in three replicates. The bone dry weight of the samples was determined by subjecting the dried samples to oven at 105°C for 24 hours.

2.3 Drying Kinetics

The drying kinetics of thin layer samples is often used to describe the heat and mass transfer during the drying process and it is affected by the drying condition. Single layer semi-empirical expressions are sufficient to describe the drying kinetics when external resistance to the heat and the mass transfer are minimized (Midilli et al., 2002; Rayaguru and Routray, 2012). The initial moisture content (M0), moisture content at a given drying time t (Mi), and EMC (M∞) of the lemon slices were calculated by equation (1), (2) and (3), respectively. These equations were expressed in dry basis with the unit of gram water per gram dry sample. Wd refers to bone dry weight of the sample.

\[ M_0 = \frac{W_o - W_d}{W_d} \]
\[ M_i = \frac{W_i - W_d}{W_d} \]
\[ M_eq = \frac{W_eq - W_d}{W_d} \]

2.4 Drying Rate

The drying rate, R at constant drying condition is given by equation (4). \( F_i \) (g H2O / g dry weight) is the free moisture content of the sample at time \( t_o \) as shown in equation (5) (Geankoplis, 2003; Chin et al., 2009).

\[ R = \frac{W_d}{A} \frac{F_{t+1} - F_i}{t_{t+1} - t_i} \]
\[ F_i = M_i - M_eq \]

Equation (5) can be rewritten in terms of predicted moisture ratio, as shown in equation (6). Thus, the drying rate for each experiment was calculated using equation (7) where \( MR_{pre} \) is the predicted moisture ratio at a given drying time t.

\[ F_i = MR_{pre,t} (M_0 - M_eq) \]

\[ R = \frac{W_d}{A} \left( \frac{MR_{pre,t+1} - MR_{pre,t}}{t_{t+1} - t_i} \right) \] (7)

2.5 Effective Moisture Diffusivity

According to Fick’s second law of diffusion, the drying behaviour of lemon slices could be described by the simplified mathematical Fick’s second model, as shown in equation (8) (Crank, 1975; Aktaş et al., 2009; Kaya et al., 2010). It was used to calculate the effective diffusivity, \( D_{eff} \) (mm²/s) at different moisture contents of lemon slices along the drying process. The values of \( D_{eff} \) were obtained by fitting the moisture ratio calculated from equation (8) to the experimental data. L is the thickness of samples (mm), n is the positive integer, and t is the drying time (s).

\[ MR_t = \frac{8}{\pi^2} \sum_{n=0}^{L} \frac{1}{(2n+1)^2} \exp \left( \frac{- (2n+1)^2 \pi^2 D_{eff} t} {L^2} \right) \]

………………….. (8)

2.6 Vitamin C Analysis

For vitamin C analysis of dried samples, metaphosphoric acid-acetic acid (HPO2.CH3COOH) extracting solution and indophenols standard solution (IS solution) were needed as described in AOAC Official Method 967.21, 45.1.14. Dried lemon slices (at equilibrium moisture content) were cut into small pieces of equivalent size and subjected to extraction process for 24 hours using 50 ml of HPO2.CH3COOH. After removal of solid slices by vacuum filtration, the supernatants were titrated into 5 ml of IS solution. The amount of supernatants, \( V_s \) used to decolourize the IS solution was recorded.

The standard curve for quantitative analysis of vitamin C content in the dried lemon slices was obtained using Ascorbic acid as standard solution. Ascorbic acid solutions (AAS) were prepared by dissolving the L(+)-Ascorbic acid into extracting solution to obtain five solutions with different concentrations (C), which were 0.1, 0.2, 0.3, 0.4, and 0.5 mg of Ascorbic acid per ml of extracting solution. Subsequently, these solutions were titrated into 5 ml of IS solution. The amount of AAS used, \( V_s \) to decolourize the IS solution for each concentration was recorded. The graph of \( V_s \) against 1/ C was plotted as shown in Fig. 2. The concentration of Vitamin C in each dried sample extract can be determined based on the volume of supernatant (V) used to decolourize the IS solution (equation 9). Vitamin C in the dried samples was expressed in mg of Ascorbic Acid / g dry weight according to equation (10). V is the volume of the extracting solution used.

\[ C_{AAS} = \frac{0.310 \, \text{mg}}{V_s} \] 

………………….. (9)

\[ \text{Vitamin C} = \left( \frac{C_{AAS} \times V}{W_d} \right) \] 

………………….. (10)
2.7 TPC Analysis

Similar to Vitamin C analysis, dried lemon slices (at equilibrium moisture content) were cut into small pieces of equivalent size and subjected to extraction process for 24 hours using 50 ml of solvent which made up of acetone, distilled water and hydrochloric acid in volume percentage of 75%, 22% and 3%, respectively. After removal of solid slices by vacuum filtration, 1 ml of supernatants were titrated into 5 ml of 0.2 N Folin-Ciocalteau reagent and held for 3 minutes. Then, 4 ml of 7.5% sodium carbonate solution (Na₂CO₃) was added into the mixture and held for 30 minutes incubation in dark at room temperature to allow colour development. Subsequently, the extract was adjusted with 40 ml of distilled water and absorbance of the extract was measured at 765 nm in single beam spectrophotometer (JENWAY 6320D).

The standard curve for quantitative analysis of TPC in the dried lemon slices (Fig. 3) was obtained using Gallic acid (0.02 - 0.08 mg/ml) as standard solution. TPC of the dried samples was determined based on equation (11) and it was expressed in mg of Gallic acid / g dry weight according to equation (12). DF is dilution factor and V is the volume of the extracting solution used.

\[
C_{GA} = \frac{A}{9.02 \text{ ml mg}^{-1}} \quad (11)
\]

\[
\text{TPC} = \left( \frac{C_{GA} \times DF \times V}{W_d} \right) \quad (12)
\]

2.8 Statistical Analysis

The results of experiments were analyzed in triplicate by using completely randomized design. Analysis of variance was performed by SAS statistical analysis package (SAS institute Inc, SAS /STAT 9.2). Mean were compared by Tukey’s Studentized Ranged test at 95% confidence level.

3. RESULTS AND DISCUSSIONS

3.1 Drying Kinetics

Fig. 4 showed the graph of moisture ratio against drying time of thin layer drying kinetics of lemon slices under different drying methods. Among all tested drying methods, oven drying at 60°C showed the fastest drying rate with total drying time found to be 48.6% to 93.3% shorter as compared to those oven and hybrid heat pump dried slices at lower drying temperature. Drying at elevated temperature intensifies the drying rate by enhancing the vapour pressure within the lemon slices which stimulates the diffusion of moisture to the product surface and consequently evaporated by convection. However, the drying kinetics of CF-HP and CF-HT-HP dried lemon slices are comparable with oven drying at elevated temperature (40°C and 50°C) within the first 500 minutes. Low relative humidity condition in hybrid heat pump dryer stimulates the moisture evaporation of the sample although it was conducted at mild drying temperature. Nevertheless, when moisture content of the sample further decreases along the drying process, drying temperature dominates the drying process and prolongs the total drying time required as compared to oven drying at elevated temperature. Mild temperature drying of hybrid heat pump system prolongs the total drying time especially at the late phase of drying process, due to slow internal diffusion of bound moisture in the lemon slices which limits the drying rate. Unlike other porous drying materials, the moisture content was encapsulated in the semi-permeable membrane of lemon pulp. Thus, diffusivity of moisture to the surface of the lemon slices was totally depended on moisture transportation through the semi-permeable membrane (Geankoplis, 2003). Similar behaviour of drying process was reported by Chen et al., (2005). For heat pump and hybrid heat pump drying, total drying time required for CF-HT-HP dried lemon slices was the shortest, which was found to be 24.0% and 34.5% shorter than CF-HP and HP dried lemon slices, respectively. The assistance of auxiliary heater as well as Coulomb force intensified the drying rate by drawing out the vapour within the lemon slices which in turn expedited the diffusion of moisture to the product surface. This shortened the total drying time required by hybrid heat pump drying as compared to the heat pump drying alone.

3.2 Drying Rate

Variation of drying rate with free moisture content of lemon slices subjected to different drying methods was illustrated in Fig. 5. Highest drying rate was found for oven drying of lemon slices at 60°C, which recorded at 13.1 g H₂O / m²•min, followed by oven drying at 50°C and 40°C, CF-HT-HP, CF-HP and HP drying recorded at 8.4, 4.9, 4.0, 3.5, and 3.2 g H₂O / m²•min, respectively. Only two falling rate periods were found in the drying of lemon slices; first falling rate and second falling rate, which designates the entire drying process was dominated by the internal moisture diffusion. The turning points of two falling rate periods were determined to be at free moisture content of 1.5, 4.2, 5.0 and 5.6 g H₂O / g dry weight for hybrid heat pump, oven 40°C, 50°C, and 60°C dried lemon slices, respectively. The turning point of the first falling rate was found at high free moisture content of the lemon slices drying at elevated drying temperatures, as high initial rate of unbound moisture evaporation of the slices occurred before the bound moisture vaporized at the later stage of drying.

At free moisture content higher than 3.0 g H₂O / g dry weight, CF-HT-HP and CF-HP drying of lemon slices showed relatively high drying rate as it overtook the drying rate of oven dried slices at 40°C. Whereas, when the free moisture content of the slices reduced from 4.5 g H₂O / g dry weight to 2.0 g H₂O / g dry weight, the drying rate of hybrid heat pump dried slices was found to become comparable with the drying rate of oven dried slices at 50°C. For free moisture content below 2.0 g H₂O / g dry weight, drying rate for each drying methods was almost equal to each.
other except for oven drying at 60°C. The contribution of Coulomb force for hybrid heatpump drying process at mild temperature drying was significant during the drying process. The induced Coulomb force in hybrid heatpump drying could overcome the adhesion force between the bound moisture and the interior surface of the lemon pulp to enhance the moisture transportation through the semi-permeable membrane. As a result, there was an improvement in terms of drying rate for CF-HP and CF-HT-HP drying of lemon slices as compared to heatpump drying alone which in turn shortened the total drying time required as discussed in the previous section.

3.3 Effective Diffusivity

Generally, moisture diffusivity of drying material is affected by its moisture content, drying condition, as well as its composition and porosity (Luikov, 1970; Chin et al., 2009). However, for drying of lemon slices, the effective moisture diffusivity was investigated based on the effect of drying conditions and its moisture content when dried under different drying methods. The average effective diffusivity value was found in the range of 21.1 to 63.8 × 10⁻⁴ mm²/min⁻¹ for oven drying and 16.2 to 20.5 × 10⁻⁴ mm²/min⁻¹ for heatpump and hybrid heatpump drying. As shown in Fig. 6, the effective diffusivity of each drying method increased to maximum as moisture content decreased to about 3.0 g H₂O / g dry weight for heatpump and hybrid heatpump dried lemon slices and 4.6 g H₂O / g dry weight for oven dried lemon slices. The profile of effective diffusivity of all drying methods was similar to Region I and Region II of the diffusion model which was proposed by Luikov (1970), which indicated that vapour phase diffusion dominated the mechanism of internal diffusion. During oven drying, as the temperature of the lemon slices rose at early stage of drying process, the water vapour pressure inside the lemon slices increased. This resulted in rapid transportation of moisture content through the semi-permeable membrane and pressure induced opening of pores and thus enhanced the moisture diffusivity of lemon slices (Darvishi et al., 2012). For mild temperature drying such as heatpump and hybrid heatpump drying, the low relative humidity and induced Coulomb force in hybrid heatpump drying also stimulated the moisture diffusion by pulling out the moisture content towards the surface of lemon slices for vapour evaporation process.

Nevertheless, rapid declined in effective diffusivity was observed in both heatpump and hybrid heatpump drying methods when moisture content dropped below 3.0 g H₂O / g dry weight. This could be due to the shrinkage effect as the collapse of interior structure of lemon slices mitigated the moisture diffusion and only small amount of moisture was able to diffuse out to the surface when the moisture content of the slices was near to EMC (Chin et al., 2009). Meanwhile, effective diffusivity was found to drop mildly in all oven dried slices. This showed that drying with sufficient heat energy promotes greater vapour pressure inside the lemon slices for a better vapour phase diffusion and thus minimizes the shrinkage effect in mitigating the moisture diffusivity.

3.4 Quality Analysis

3.4.1 Vitamin C Content of dried lemon slices

Vitamin C analysis of the oven and heat pump dried lemon slices at different drying conditions were conducted. The Vitamin C content in lemon slices was calculated using the standard curve in Fig. 2. Based on Fig. 7, CF-HP dried lemon slices showed the highest amount of Vitamin C among all samples with the value of 6.74 mg Ascorbic Acid/g dry weight. The Vitamin content of CF-HP dried slices is significantly higher (p<0.05) than oven dried slices although longer drying time is required. Similar to this, the amount of Vitamin C in heatpump and CF-HT-HP dried samples was found to be 2.4 % to 93.2% higher than oven dried samples. In other words, oven drying of lemon slices at elevated temperature degraded the Vitamin C content drastically as compared to heatpump and hybrid heatpump drying of lemon slices at mild temperature. This indicates drying of lemon slices at mild temperature could retain high amount of Vitamin C during the drying process (Kaya et al., 2010). Prominent degradation of Vitamin C in 50°C and 60°C oven dried lemon slices could be due to combination of thermal degradation and enzymatic oxidation of Vitamin C. Thus, heatpump and hybrid heatpump drying preserved most of the Vitamin C content in the dried lemon slices as mild drying temperature with relatively high drying rate could minimize the deterioration of Vitamin C through thermal degradation as well as enzymatic oxidation. In addition, integration of Coulomb force in heatpump drying (CF-HP) reduced the total drying time required as compared to HP drying which lead to a better Vitamin C retention in dried lemon slices. As compared with CF-HP and HP, the used of auxiliary heater in CF-HT-HP drying method which raised the temperature to 31°C further reduced the Vitamin C content of lemon slices to 5.77 mg Ascorbic Acid/g dry weight, which was insignificant difference (p>0.05) with the Vitamin C content of lemon slices dried at 40°C.

3.4.2 TPC Content of dried lemon slices

TPC analysis of lemon slices dried with different drying methods was conducted and the results were shown in Fig. 8. In contrast to the results shown in Vitamin C analysis of dried lemon slices (section 3.4.1), oven dried slices at elevated temperature (50°C) retained the highest amount of TPC, followed by oven dried slices at 60°C and 40°C, hybrid heatpump and heatpump dried slices. The result is in accordance with the findings of Vega-Galvez et al., 2009 and Moraes et al., 2013 for drying of pepper, where high retention of TPC in lemon slices dried at elevated temperature (50°C and 60°C) might be due to the availability of precursors of phenolic molecules by non enzymatic inter-conversion between phenolic molecules. Furthermore, fast drying rate of the lemon slices at 50°C and 60°C also prevents the degradation of TPC through
volatilization, oxidation and heat destruction process. The amount of TPC in 50°C and 60°C oven dried samples was significantly higher (p<0.05) than those in 40°C oven dried slices and both heatpump and hybrid heatpump dried slices. The effect of total drying time on the TPC of dried slices was further shown by the heatpump and hybrid heatpump dried slices. Among the heatpump and hybrid heatpump dried slices, the TPC in CF-HP and CF-HT-HP was significantly higher (p<0.05) than the TPC of HP dried slices (p<0.05). This could be due to shorter total drying time required by hybrid heatpump drying as compared to heatpump drying of lemon slices. The results show that drying duration significantly affects the total TPC of lemon slices as compared to drying temperatures.

4. CONCLUSIONS

The drying kinetics and quality evaluation of lemon slices dried by hot air circulation oven, heatpump and hybrid heatpump drying were investigated. Oven drying at various drying temperatures has high drying rate as compared to the heatpump and hybrid heatpump drying methods as the drying rate of lemon slices increases when the drying temperature increases. Nevertheless, integration of auxiliary heater and Coulomb force in heatpump drying improved the drying kinetics of lemon slices and it was found to be comparable with those lemon slices dried by hot air at 40°C and 50°C. This indicates a relatively high drying rate of hybrid heatpump drying method although it was conducted at mild drying temperature. Drying rate curves of lemon slices exhibited only falling rate periods for all drying methods, with average effective moisture diffusivity ranged from 16.2 × 10⁻⁴ to 63.8 × 10⁻⁴ mm²·min⁻¹. Quality analysis of lemon slices dried by oven and heatpump methods showed that TPC of the slices could be affected by total drying time whereas drying temperature significantly affects the Vitamin C content of the lemon slices. Drying of lemon slices at mild drying temperature and relatively high drying rate produced dried lemon slices with the highest content of Vitamin C among all dried products; whereas, the highest TPC content of dried lemon slices was found in oven dried lemon slices at 50°C. Coulomb-force-assisted-heatpump (CF-HP) is recommended for the drying of lemon slices as it demonstrated relatively high drying rate and produced the dried lemon slices with the highest Vitamin C content and relatively high amount of TPC among the dried slices.

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