Investigation of hot-air assisted continuous radio frequency drying for improving drying efficiency and reducing shell cracks of inshell hazelnuts: The relationship between cracking level and nut quality

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ABSTRACT
Hot-air assisted continuous radio frequency (HARF) heating was utilized to dry inshell hazelnuts with high initial moisture content (25.5%, MCwb) for achieving higher drying efficiency and lower shell cracks. Effect of electrode gap and hot-air (HA) velocity at 40 °C on drying efficiency, heating uniformity, and shell cracking ratio were investigated. The ideal electrode gap and hot-air (HA) velocity were identified as 14 cm and 2.0 m/s, respectively, to provide shorter drying time (3.5 h) compared to HA drying (22 h), and less (36%) level 3 cracked nuts (cracks with width >0.3 mm and length >10 mm, CR3). The two-step HA-HARF drying using 19% MCwb as an intermediate MCwb resulted in dried nuts with lower CR3 (27%). Shell cracking ratio highly correlated with the size and weight of inshell nuts and kernels, kernel weight percentage, and air-gap volume between shell and kernel. Unexpectedly, shell cracks to level 3 influenced kernel color, but not lipid oxidation. Level 3 cracked nuts dried by the two-step HA-HARF exhibited much less lipid oxidation than that of HA dried non-cracked nuts during accelerated storage at 35 °C, indicating that two-step HA-HARF has great potential for drying hazelnuts with high quality and storability.

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1. Introduction

The state of Oregon produces 99% of the total USA hazelnut (Corylus avellana L.). To prevent mold and aflatoxins contamination and ensure food safety, fresh hazelnuts are dried to reduce moisture content (wet basis, MCwb) to an upper limit of 10% for inshells and 6% for kernels according to the industry standard and federal regulation (USDA, 2016). The corresponding kernel water activity (aw) is less than 0.65 (Ozay et al., 2008). Hot-air (HA) drying using convective hot-air has been applied by the hazelnut industry for few decades. Our previous study identified HA drying at 43 °C/40% relative humidity (RH) as an optimal condition of inshell nuts for achieving relatively higher retention of bioactive compounds and lower lipid oxidation and enzyme activity compared to other HA conditions (Wang et al., 2018a). However, HA drying took long time (12–24 h) depending on MCwb of fresh nuts, which not only induced high energy cost because of insufficient heat transfer and low thermal conductivity of voids between shell and kernel, but also adversely impacted the quality of hazelnuts (Wang et al., 2018a).

Radio frequency (RF) dielectric heating alone or in combination with HA (for improving heating uniformity and moisture diffusion) as an advanced thermal treatment technology has been studied to shorten the drying times for nuts including macadamia nuts (Wang et al., 2014), walnuts (Zhang et al., 2016a; Zhou et al., 2018), and peanuts (Harraz, 2007; Zhang et al., 2016b) owing to its high drying efficiency and low energy cost. We also investigated the effect of electrode gap and HA temperature in continuous hot-air assisted radio frequency (HARF) drying on the drying efficiency and heating uniformity of inshell Barcelona, a typical cultivar of Oregon hazelnuts, and found that HARF drying had
much higher drying rate and produced dried kernels with lower lipid oxidation compared to HA drying (Wang et al., 2020b). However, the continuous HARF drying led to higher cracking ratio (66%) of inshell nuts, probably due to excessive moisture vapor accumulating between shell and kernel caused by high RF energy input. It was generally believed that shell cracks could potentially influence nut quality during storage owning to exposure to oxygen. A two-step HA-HARF drying method using HA drying to reach 12% MCwb, following with an intermittent or continuous HARF drying mode (with or without pausing of RF power during the HA circulation) to finish the process was then studied with a goal of reducing shell cracking ratio to < 15% for Jefferson nuts, the cultivar mostly liable to cracking (Wang et al., 2020a). Nevertheless, depending on the weather condition of the harvesting season, MCwb of fresh inshells and kernels could go up to 25–35%, much higher than that of the nuts used in our previous study (21% MCwb of inshells and 16% MCwb of kernels for fresh Jefferson). With such a high MCwb, shell cracks and lipid oxidation because of long time exposure to high temperature and energy input can become a serious issue. Therefore, it is necessary to investigate the behavior of high MCwb nuts when subjecting to HARF and two-step HA-HARF drying and develop strategies to achieve high drying efficiency without affecting nut quality.

Shell cracks during a drying process may be attributed to the structural and morphological changes of the shell, and excessive moisture vapor accumulating in the air-gap between shell and kernel (Rahman, 2001). HA velocity was found more important for controlling shell cracks than HA temperature during HA drying of hazelnuts (Wang et al., 2018b). In RF heating, a smaller electrode gap resulted in an elevated RF power, thus elevating excessive moisture vapor accumulation (Jiao et al., 2016). Besides, intermediate MCwb for HA pre-drying in the two-step HA-HARF affected cracking ratio of Jefferson nuts with moderate MCwb (21% MCwb of inshells and 16% MCwb of kernels) (Wang et al., 2020a). Therefore, electrode gap and HA velocity in the HARF drying and intermediate MCwb in the two-step HA-HARF drying should be precisely controlled for drying high MCwb hazelnuts to reduce these potential consequences. In addition, the relationship between cracking level and physical properties of hazelnuts in HARF drying was unknown. Furthermore, the influence of shell cracking level on nut quality and storability remains controversial without solid data support.

The overall goal of this study was to investigate the principles and strategies to reduce shell cracks of high MCwb inshell hazelnuts in HARF drying while retaining high drying efficiency and nut quality and storability. Specific objectives were to (1) evaluate the effect of electrode gap (13, 14, 15 cm) and HA velocity (1.5, 2.0, 2.5 m/s) in HARF drying and intermediate MCwb (19, 16, 13%) in two-step HA-HARF drying on heating uniformity and shell cracks of Jefferson hazelnuts with high inshell and kernel MCwb (25.5% and 32.6%, respectively), (2) explore the correlation of cracking ratios with physical properties of hazelnuts, and (3) compare quality and storability of dried hazelnuts with different cracking levels and from three different drying methods (HA, HARF, two-step HA-HARF). Results from this study would provide valuable information to develop a practical protocol of HARF drying on hazelnuts with improved drying efficiency and better nut quality and storability.

2. Materials and methods

2.1. Materials and chemical reagents

Oregon Jefferson hazelnuts were provided by the Oregon Hazelnut Marketing Board and harvested by machine in late September, 2019. Washed and cleaned nuts were primarily packed in woven sack, and secondarily packed by low-density polyethylene bag (0.127 mm thickness) (PlasticMill, New Jersey, USA) to prevent moisture losses during storage at 1.7 °C before treatments.

Folin-Ciocalteu (FC) reagent, 1,1-diphenyl-2-picrylhydrazyl (DPPH), gallic acid, and phenolthalein were obtained from Sigma-Aldrich (St. Louis, MO, USA); chloroform and diethyl ether from Fisher Scientific (Hampton, NH, USA); cumene hydroperoxide from TCI America (Portland, OR, USA); sodium hydroxide, hexane, methanol and L-ascorbic acid from MACRON, Avantor Performance Materials (Center Valley, PA, USA).

### 2.2. Three drying methods

MCwb of fresh hazelnut inshells and kernels were 25.5% and 32.6%, respectively, measured by the gravimetric oven drying method (AOAC, 2000). Drying was terminated when MCwb of inshells reached <10%.

Three drying methods including HA, HARF, and two-step HA-HARF drying were applied in this study. HA drying process was conducted at 43 °C using a forced-air dryer (MP-2000, Enviro-Pak, Clackamas, OR, USA) with horizontal airflow at ~1.0 m/s and 40% RH according to our previous study (Wang et al., 2018a).

HARF drying process was conducted in a 6-kW, 27-MHz pilot-scale RF unit (COMBI 6-S, Strayfield International, Wokingham, U.K.) with a customized auxiliary hot-air system to provide air circulation at 40 °C (Wang et al., 2020b). Jefferson inshell nuts (1.2 kg) were loaded in a rectangular container with inner dimensions of 25.5 cm × 15.5 cm × 8 cm, and placed at a fixed position above the bottom electrode. The container consisted of Teflon wall and 12.7 mesh nylon screen as the cover at the bottom and sides. Three electrode gaps (13, 14, 15 cm) were chosen based on our previous study for achieving desired heating uniformity and drying efficiency, and HA with three air velocities (1.5, 2.0, 2.5 m/s, measured from the air outlet) was applied vertically blowing from the perforated bottom electrode. HA temperature (~ 40 °C) was recorded by a fiber optic temperature sensor (TempSens, Opsens Inc., Sainte-Foy, Quebec, Canada) located above the sample container. Four other fiber optic temperature sensors were inserted into pre-drilled holes in the nuts placed in the center of the top layer, the corner of top layer, the center of the bottom layer, and the corner of bottom layer, respectively for recording kernel temperatures. During HARF drying, samples were taken out at 15 min, 30 min, and then every 30 min to take thermal image photos by a digital infrared camera (FLIR T400, FLIR Systems, Inc., North Billerica, MA, USA) and weigh within 30 s to calculate MCwb. Samples were then placed back into the RF cavity for continuous heating under the same conditions. The ideal electrode gap and HA velocity were identified from above experiments by achieving high drying efficiency, but low cracking ratio and lipid oxidation. MC in dry basis (MCdb) shown in Fig. 1 was calculated as:

\[
MC_{db} = \frac{100MC_{wb}}{100 - MC_{wb}}
\]  (1)

The 16% MCwb of inshells was found to be a critical MCwb dividing HARF drying of Jefferson nuts into two stages: stage I (fast drying rate with rapid removal of free water from wet nuts) and stage II (moisture started to transfer through the shell inducing most of cracks) (Wang et al., 2020a). Therefore, for two-step HA-HARF drying, nuts were dried by HA first to reach a given MCwb of 19, 16, and 13%, called intermediate MCwb, then continued for HARF drying till reaching inshell MCwb of ≤ 10%. The ideal intermediate MCwb was identified using the same criteria as stated above.

The above stated HA, HARF and two-step HA-HARF testing conditions were repeated twice and each drying method
Fig. 1 – Drying curve and drying rate of inshell hazelnuts by hot-air associated radio frequency (HARF) heating. (a) and (b) Hot-air (HA) velocity effect at electrode gap of 14 cm; (c) and (d) electrode gap effect at HA velocity of 2.0 m/s; (e) and (f) intermediate moisture content (MC) effect in the two-step HA-HARF drying. Note: HA-19MC-HARF drying meant to use HA drying to reach 19% MC following with HARF drying to finish the process; HA-16MC-HARF drying meant to use HA drying to reach 16% MC following with HARF drying to finish the process; HA-13MC-HARF drying meant to use HA drying to reach 13% MC following with HARF drying to finish the process. Red line in (a), (c), and (e) indicated ~19% MC dry basis (16% MCwb), a critical MC found in our previous study.

with identified ideal conditions was then repeated four times to get enough nut samples for storage test. All dried nuts were cooled down to room temperature. Nuts from two replicates of each drying method were packed into woven polypropylene bags and stored at 35 °C (25% RH) for a 6-month accelerated storage test. Dried nuts from the other four replicates of each drying method were classified for cracking levels as described in Section 2.3, and then packed into woven polypropylene bags. In the typical commercial storage, inshell hazelnuts are stored at ambient temperature (10–26 °C) for < 1 year. Q10 factor, representing the factor by which the rate of a reaction increases for every 10 °C, was reported as 1.87 for hazelnut kernels in polyethylene packaging (Shafei et al., 2020) and 1.13 for inshells using nitrogen packaging (Dong et al., 2012). To accelerate the lipid oxidation, all packed nuts were stored at 35 °C and 25% RH for a 6-month accelerated storage test that was equivalent to 1-year storage at 20 °C based on an estimated Q10 of 1.50. Samples were analyzed at month 0, 3 and 6.

2.3. Classification of cracking levels of dried inshell nuts

Dried inshell hazelnuts from all three drying methods were sorted on cracking levels after drying and cooling to room temperature, and classified into five cracking levels (Table 1). Level 0 represented the nuts without cracking, level 1 was nuts cracked during drying process but the cracks were closed after cooling down to room temperature, level 2 was nuts showing a crack of a width < 0.3 mm or length < 10 mm during drying and the crack still existed after cooling down, and level 3 was nuts with one or more cracks of a width > 0.3 mm and length > 10 mm. Severely cracked nuts had damaged shell during drying and cooling, which met the criteria for level 3 and accounted as a part of level 3 cracked nuts, but also recorded individually. The quantitative ratio of non-cracked nuts was recorded as CR0, representing level 0 nuts. Cracking ratio (%) of the nuts at each level was calculated as a percentage of cracked nuts at each level to the total number of nuts at each replicate, and
2.4. Heating uniformity

The mean and standard deviation (SD) of the inshells surface temperature at 0 min and 30 min of HARF heating were analyzed from thermal image photos by ExaminIR (FLIR Systems, Inc., North Billerica, MA, USA). Heating uniformity index (λ) defined as the ratio of increase in SD of hazelnut surface temperature (°C) to increase in mean of hazelnut surface temperature (°C) using the following equation (Wang et al., 2005):

\[
λ = \frac{σ^2 - σ_0^2}{μ - μ_0}
\]  

(2)

where μ_0 and μ were the mean hazelnut surface temperature (°C) at 0 min and 30 min of HARF drying, and σ_0 and σ were SDs of hazelnut surface temperatures (°C) at 0 min and 30 min of HARF drying, respectively.

2.5. Physical and morphological properties of hazelnuts

Mass of inshell nuts and kernels (n = 20) for each level of cracked nuts were measured using a digital scale (Ohaus, Gold Series, Parsippany, NJ, USA). Inshell diameter and kernel diameter were measured by a caliper (Spi 2000, Swiss Precision Instrument Inc., Garden Grove, CA, USA). The density (g/mL) of shell (n = 10) was calculated by dividing mass (g) of shell with volume (mL) of shell determined using water displacement method in a graduated cylinder (Plasticic et al., 2006).

Correlations between data obtained and cracking levels were calculated using Pearson’s correlation coefficient by setting the data of cracking levels as 0, 1, 2, and 3, respectively.

For morphological properties, inshell hazelnuts were vertically cut by means of a handsaw, and the longitudinal section was observed by a stereomicroscope (Leica Microsystems AG, Heerbrugg, Switzerland).

2.6. Quality attributes of hazelnuts

2.6.1. Physicochemical properties of kernels

Kernels were obtained from shelled hazelnuts for quality analyses. MC_{wb} of kernels (%) was measured using the gravimetric oven drying method (AOAC, 2000) and a_{wb} of kernels was determined by an electronic hygrometer (Aqua Lab 3 TE, Decagon Devices Inc., Pullman, WA, USA) in triplicate. Kernels (n = 10) were cut into halves and the core of kernels were measured for L^* (lightness), a^* (redness), and b^* (yellowness) using a colorimeter (LabScan XE, HunterLab, Reston, VA, USA). Color difference (∆E = √[(L^* - L_0)^2 + (a^* - a_0)^2 + (b^* - b_0)^2]) was calculated from obtained L^*, a^*, and b^* values. L_0, a_0, and b_0 were the color index of fresh kernels.

2.6.2. Lipid oxidation of kernels

Lipid oxidation was measured by peroxide value (PV), free fatty acid (FFA) and K value (K232 and K270) in triplicate. PV presents primary hydroperoxide compounds from lipid oxidation, and FFA is formed from oxidative and enzymatic rancidity of the kernel oils. K232 and K270 indicate the conjugated diene and triene in oil samples from oxidation processes, respectively.

Oil extraction from kernels and analysis of PV and K value were done following our previous study (Wang et al., 2020b). Briefly, 0.1 g of oil sample was diluted in 10 mL of HPLC grade hexane, the absorbance was measured at 232 and 270 nm by the UV spectrophotometer, and reported as K232 and K270, respectively. FFA content was determined using NaOH titration method (AOCS, 1977). Briefly, 1.0 g of oil sample was mixed with 25 mL of ethanol-diethyl ether mixture (1:1, v/v). Four drops of 1% (w/v) phenolphthalein ethanol solution were added as a pH indicator. The mixture was titrated with 0.01 mol/L NaOH till the pink color appeared and lasted for at least 10 s with stirring. The FFA result was reported as % oleic acid.

2.6.3. Total phenolic content and DPPH antioxidant capacity of kernels

 Phenolic compounds extraction from kernels, determination of total phenolic content (TPC) and DPPH antioxidant capacity of kernels were conducted using the same methods in our previous study in triplicate (Wang et al., 2020b). TPC was analyzed according to the Folin-Ciocalteu (FC) colorimetric assay and reported as mg GAE/g dry kernel. DPPH antioxidant capacity was assayed using ascorbic acid as the calibration standard and reported as mg ascorbic acid equivalents (AAE)/g dry kernel.

2.6.4. Enzyme activity

Extraction and activity of peroxidase (POD) and polyphenol oxidase (PPO) were conducted and assayed according to Wang et al. (2020b) in triplicate. All enzyme activities were expressed
as μkat/L. One katal for PPO activity was defined as the amount of the enzyme converted one mol of catechol per second, and one katal for POD activity was defined as the amount of the enzyme converted one mol of o-dianisidine per second.

2.7. Experimental design and statistical analysis

Three specific experimental designs were applied in this study. First, a single-factor experiment was employed to investigate the effect of electrode gap (13, 14, 15 cm, HA velocity was fixed at 2.0 m/s) and HA velocity (1.5, 2.0, 2.5 m/s, electrode gap was fixed at 14 cm), respectively on HARF drying efficiency, shell cracking ratio, and heating uniformity of hazelnuts. Secondly, the identified ideal electrode gap and HA velocity from first study were used to investigate the effect of intermediate MCwb (19, 16, 13%) on two-step HA-HARF drying efficiency and shell cracking ratio using single-factor experiment. Thirdly, the effect of three drying methods (HARF, HA, two-step HA-HARF) and cracking levels on quality and storability of inshell nuts were studied using two-factor completely randomized design. One-way ANOVA coupled with post hoc least significant difference (LSD) was conducted for the multiple comparison of all experiments using SAS v9.2 (The SAS Institute, Cary, NC, USA) at significance level of 0.05.

3. Results and discussion

3.1. Effectiveness of HARF heating for improving drying efficiency and reducing shell cracks of high MCwb hazelnuts

3.1.1. Effect of HA velocity

Fig. 1(a) and (b) illustrate the drying curve and drying rate of inshell hazelnuts under HARF drying with different HA velocities. The drying rate decreased dramatically at the initial stage, but slightly at the later stage of HARF drying. This was because the higher dielectric loss factor of hazelnuts (a greater number of polar water molecules) at the beginning of HARF induced more RF power input (Wang et al., 2014). HA velocity of 1.5 m/s resulted in significantly (P < 0.05) longer drying time (4.0 h) than that of 2.0 and 2.5 m/s (3.5 h), while the average heating rate for the first 5 min of HARF, kernel average temperature, and heating uniformity were at the same level among three HA velocities (Table 2). This result was probably due to the relatively lower water evaporation rate induced by a slower HA velocity. After the kernel temperature exceeded the HA temperature (40 °C), the air mainly served as a medium to carry water vapor away from the shell surface and stabilize kernel temperature. Lower HA velocity could not sufficiently help promote water evaporation from the shell, thus extending the drying process. It was noteworthy that HARF drying with all three HA velocities resulted in non-detected PV, indicating low lipid oxidation generated during HARF drying. In comparison with HA velocity of 2.5 m/s, HA velocity of 2.0 m/s resulted in a lower CRb but similar CRc so that it was chosen to be the ideal HA velocity for HARF drying. The slight difference in shell cracking ratio between HA velocity of 2.0 m/s and 2.5 m/s might be caused by the higher drying rate at the initial stage (3.49 g water kg dry nut⁻¹ min⁻¹) at HA velocity of 2.5 m/s, leading to more structural change of shell and higher cracking ratio during the drying process.

3.1.2. Effect of electrode gap

As shown in Fig. 1(c) and (d), the smaller electrode gap of 13 cm significantly (P < 0.05) reduced the drying time (2.0 h), providing higher drying rate along with better heating uniformity (0.134) in comparison with larger electrode gaps (Table 3). This result could be attributed to the significantly higher heating rate (7.07 °C/min) and kernel temperature (84.63 °C) when using electrode gap of 13 cm, giving rise to faster moisture diffusion rate from kernels to the shell and air. Also, the heating uniformity calculated in this study was traced to the nut surface temperature data analyzed from the infrared images at heating time of 0 min and 30 min. Better heating uniformity found in HARF at electrode gap of 13 cm was because of a rapid increase in the mean hazelnut surface temperature from 0 min to 30 min, but similar increase in the standard deviation of nut surface temperatures compared to those at electrode gaps of 14 cm and 15 cm. However, HARF at electrode gap of 13 cm showed significantly higher CRb (14.72%) than that at electrode gap of 14 cm (3.25%) and 15 cm (0.65%), which was possibly due to the excessive heating rate (7.07 °C/min) accumulating too much moisture vapor within the shell and eventually resulting in cracks in the most vulnerable part of hazelnut shell. Considering the significant shorter drying time (3.5 h) at electrode gap of 14 cm than that when using electrode gap of 15 cm (6.0 h), 14 cm was thus chosen as the ideal electrode gap for HARF drying. This result was consistent with our previous study investigating the effect of electrode gap in HARF drying on Barcelona inshell nuts (Wang et al., 2020b). A similar trend was also observed in RF drying of inshell almonds and macadamia nuts (Gao et al., 2011; Wang et al., 2014). However, all three drying conditions resulted in CRb over 33%, which was then improved by the two-step HA-HARF drying discussed in Section 3.1.3.

3.1.3. Effect of intermediate MCwb in the two-step HA-HARF drying

The two-step HA-HARF drying significantly reduced drying time (5.2 – 12.3 h) compared to HA drying (22.0 h) due to a significantly lower drying rate (<5 g water · kg dry nut⁻¹ · h⁻¹) at the later stage of HA drying, and lower intermediate MCwb in the two-step HA-HARF drying resulted in longer total drying time (Fig. 1(e) and (f)). Moreover, two-step HA-HARF drying led to dried nuts with much lower PV in comparison with HA dried nuts after drying (Table 4). Within the two-step HA-HARF drying, a higher intermediate MCwb (19%) reduced level 3 cracked nuts ratio (CR3) to 27.31%, significantly lower than that using 16% and 13% MCwb as intermediate MCwb (37.86% and 34.03%, respectively). This result was contradictory to our previous study showing that two-step HA-HARF drying using 12% MCwb as intermediate MCwb had the lowest cracking ratio for Jefferson nuts among three studied intermediate MCwb (16, 14 and 12%) (Wang et al., 2020a). Two reasons might explain this different result. On the one hand, there was varied MCwb distribution in fresh shell and kernel between samples in the two studies. The kernels in our previous study had lower MCwb (16%) than that of shells (22%), while in this study, not only both kernel and shell had higher MCwb than the samples from previous study, but also kernels showed significantly higher MCwb (33%) than that of shell (23%). Fresh Jefferson nuts with higher shell MCwb than kernel could lead to higher power absorption and dissipation in shells from RF energy, which potentially resulted in more shell cracking. Therefore, sufficient pre-HA drying is necessary to remove moisture in the shells for nuts with higher shell MCwb. On the other hand, HA
2.53a 48.33a
3.26a 27.31b
14.72a 39.44a
HA-HARF still drying of intense HA-HARF was significant in shells dried. Hence, vent different
PV 1 PV 1 PV 1 PV
1 PV (peroxide value) was tested for nut samples right after drying.
2 nd meant non-detected.

The mean value of each measurement in the same column with same lower letter in the superscript indicated non-significant difference among treatments (P < 0.05).

Table 3 – Effect of electrode gap on the HARF drying characteristics of inshell hazelnuts.

<table>
<thead>
<tr>
<th>Electrode gap (cm)</th>
<th>Drying time (h)</th>
<th>Average heating rate (°C/min) for the first 5 min of HARF</th>
<th>Kernel average temperature (°C) during RF heating between 15 min and end of HARF</th>
<th>Heating uniformity (a)</th>
<th>Heatinguniformity (a)</th>
<th>CR4 (%)</th>
<th>CR5 (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13</td>
<td>2.0a</td>
<td>7.07a</td>
<td>84.63a</td>
<td>0.134b</td>
<td>nd&lt;sup&gt;1&lt;/sup&gt;</td>
<td>14.72a</td>
<td>39.44a</td>
</tr>
<tr>
<td>14</td>
<td>3.5b</td>
<td>4.36b</td>
<td>70.76b</td>
<td>0.156&lt;sup&gt;a&lt;/sup&gt;</td>
<td>nd&lt;sup&gt;1&lt;/sup&gt;</td>
<td>3.25b</td>
<td>36.35b</td>
</tr>
<tr>
<td>15</td>
<td>6.0</td>
<td>3.41&lt;sup&gt;c&lt;/sup&gt;</td>
<td>58.69&lt;sup&gt;c&lt;/sup&gt;</td>
<td>0.165&lt;sup&gt;a&lt;/sup&gt;</td>
<td>nd&lt;sup&gt;1&lt;/sup&gt;</td>
<td>0.65&lt;sup&gt;c&lt;/sup&gt;</td>
<td>33.89&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

The mean value of each measurement in the same column with same lower letter in the superscript indicated non-significant difference among treatments (P < 0.05).
1 PV (peroxide value) was tested for nut samples right after drying.
2 nd meant non-detected.

Table 4 – Effect of intermediate MC on the two-step HA-HARF drying characteristics of inshell hazelnuts.

<table>
<thead>
<tr>
<th>Intermediate MC of inshell hazelnuts (%), wet basis</th>
<th>HA pre-drying time (h)</th>
<th>CR3 caused by pre-drying (%)</th>
<th>HARF heating time (min)</th>
<th>Total heating time (h)</th>
<th>PV (meq O2/kg oil)&lt;sup&gt;1&lt;/sup&gt;</th>
<th>CR4 (%)</th>
<th>CR5 (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>19</td>
<td>4.0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>17.4&lt;sup&gt;a&lt;/sup&gt;</td>
<td>70&lt;sup&gt;a&lt;/sup&gt;</td>
<td>5.2&lt;sup&gt;d&lt;/sup&gt;</td>
<td>nd&lt;sup&gt;1&lt;/sup&gt;</td>
<td>3.26&lt;sup&gt;a&lt;/sup&gt;</td>
<td>27.31&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>16</td>
<td>7.0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>20.0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>25&lt;sup&gt;b&lt;/sup&gt;</td>
<td>7.4&lt;sup&gt;c&lt;/sup&gt;</td>
<td>nd&lt;sup&gt;1&lt;/sup&gt;</td>
<td>2.74&lt;sup&gt;b&lt;/sup&gt;</td>
<td>37.86&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>13</td>
<td>12.0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>18.5&lt;sup&gt;a&lt;/sup&gt;</td>
<td>20&lt;sup&gt;a&lt;/sup&gt;</td>
<td>12.3&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.01&lt;sup&gt;b&lt;/sup&gt;</td>
<td>1.74&lt;sup&gt;b&lt;/sup&gt;</td>
<td>34.03&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>HA drying</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>22.0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>0.22&lt;sup&gt;a&lt;/sup&gt;</td>
<td>0.50&lt;sup&gt;c&lt;/sup&gt;</td>
<td>17.52&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

1 PV (peroxide value) was tested for nut samples right after drying.
2 nd meant non-detected. The mean value of each measurement in the same column with same lower letter in the superscript indicated non-significant difference among treatments (P < 0.05).

3.2. Quality and storability of dried hazelnuts from different drying methods

Fig. 2 reports MC<sub>aw</sub> of dried inshells and kernels and a<sub>w</sub> of dried kernels from different drying methods. MC<sub>aw</sub> of dried inshells and a<sub>w</sub> of dried kernels from all three drying methods were less than 10% and 0.65, respectively and at the same significant level (P > 0.05), while MC<sub>aw</sub> of the dried kernels using two-step HA-HARF was significantly lower than that from other two drying methods (P < 0.05), indicating two-step drying condition (43 °C/40% RH) used in this study was more intense than that used in the previous study (38 °C/60% RH). Hence, less HA drying time was chosen in this study to prevent from potentially more structure change and cracking of the shells (Galedar et al., 2008). Overall, although the two-step HA-HARF drying using 19% MC<sub>aw</sub> as an intermediate MC<sub>aw</sub> still showed higher CR<sub>3</sub> than that of HA drying, it saved 76.3% of drying time and resulted in nuts with much lower PV, hence was chosen to be the ideal intermediate MC<sub>aw</sub> in the two-step HA-HARF drying.

Fig. 2 – Moisture content of dried inshell hazelnuts and kernels and water activity of dried kernels using different drying methods. Note: Drying time for HARF drying, two-step HA-HARF drying, and HA drying was 3.5 h, 5.2 h, 22 h, respectively.
HA-HARF drying was beneficial for thermal transfer and moisture diffusion between shell and kernel. Similar results were found in two-step HA-HARF using 16, 14, and 12% MC_{wb} as intermediate MC_{wb} for drying fresh inshells with moderate MC_{wb} (~20%) (Wang et al., 2020a).

Table 5 summarizes the effect of drying methods on color, lipid oxidation, bioactive compounds and enzyme activity of dried kernels during 6 months of inshell storage at 35 °C. There were no significant difference (P > 0.05) in L* and ΔE among the nuts from three drying methods at each sample point (month 0, 3, and 6), indicating that drying methods had no significant effect (P > 0.05) on the color of kernels. Regarding lipid oxidation, PV of HARF and two-step HA-HARF dried kernels were significantly (P < 0.05) lower than that of HA dried kernels throughout the storage. It was noteworthy that PV of HA dried kernels at month 6 was significantly (P < 0.05) lower than that at month 3, implying that the highest lipid hydroperoxide occurred at about month 3. This result indicated that lipid autoxidation might have peaked around the 3 months of storage for HA dried kernels while lipid autoxidation in HARF and two-step HA-HARF dried kernels did not reach the peak even after 6 months of storage. There was no significant difference in FFA for dried kernels among the three drying methods at month 0 and month 6. However, FFA of HA dried kernels at month 3 was significantly (P < 0.05) higher than that of dried kernels using other two drying methods, implicating that the peak of lipid oxidation by enzymatic hydrolysis for HA dried nuts might also happen around 3 months of storage. Although the US hazelnut industry does not have a standard about acceptable range for PV and FFA, PV and FFA of HARF and two-step HA-HARF dried kernels during 6-month storage were within the acceptable range (PV < 1.0 mg O_{2}/ kg oil and FFA < 0.6%) for good quality of almond and walnut (Gao et al., 2011; Wang et al., 2007) while HA dried ones exceed the threshold after 3 months of storage. During the lipid oxidation, the free radicals generated during initiation and propagation are stabilized by radical resonance, which caused shift of the double bonds and cis-trans isomerization. Conjugated dienes (K_{232}) and trienes (K_{270}) are produced due to the rearrangement of the methylene-interrupted double bonds in polyunsaturated fatty acids (PUFA), and are used as an indicator of oxidation (Shahidi and Zhong, 2010). K_{232} of dried kernels from three drying methods were at the same significant level (P > 0.05) at month 0, while K_{232} of HA dried kernels showed significantly (P < 0.05) lower values than that of HARF and two-step HA-HARF dried kernels at month 3 and month 6. It was noted that PUFA took up around 10% to 15% of oil composition in hazelnut oil (Parcerisa et al., 1998) and either dienes or trienes was a temporary form that could further become lipid hydroperoxide contributing to the propagation of lipid autoxidation (Shahidi and Zhong, 2010). According to the results of PV and FFA, a possible reason was the rearrangement of the methylene-interrupted double bonds reaching the peak before month 3 for HA dried nuts. K_{270} of all dried nuts remained relatively low values, indicating that trienes could hardly be formed for hazelnut during the lipid oxidation. In general, RF waves have the unique advantages of volumetric heating within the materials due to friction of rotation of dipole molecules (i.e. water) and through ionic conduction under a high-voltage alternating electric field (Jiao et al., 2018). Since hazelnuts contain a high amount (~60%) of non-polar oil with heat-sensitive bioactive compounds, use of RF drying could help retain nut quality with a differential heating effect as well as much less drying time. Zhang et al. (2016a) found that inshell walnuts dried by HARF for 100 min resulted in less PV (0.34 meq/kg) and FFA (0.22%) than those dried by HA for 240 min (0.58 meq/kg for PV, 0.35% for FFA). PV and FFA also showed lower values for HARF dried walnuts during the storage compared to HA dried walnuts. Ling et al. (2015) found RF treated pistachios had lower PV during storage at 35 °C compared to the untreated nuts. These results demonstrated that both HARF and two-step HA-HARF could bring lower lipid oxidation of hazelnuts, promising for retaining nut quality and storability.

In respect to TPC and DPPH antioxidant capacity, HARF dried kernels showed the highest TPC and DPPH at month 0, followed by two-step HA-HARF as a result of short drying time since most bioactive compounds are thermally sensitive. However, both TPC and DPPH of two-step HA-HARF dried kernels were higher than that of HARF and HA dried kernels at month 3 and month 6, probably due to the lowest kernel MC_{wb} in two-step HA-HARF dried kernels leading to a more stable environment for antioxidants. Besides, HARF and two-step HA-HARF dried kernels showed lower PPO activity than that of HA dried ones throughout the storage, which might explain a slightly higher L* value (lightness) for HARF and two-step HA-HARF dried nuts than that of HA dried ones at month 0 and month 3 as shown in Table 5. POD activity of two-step HA-HARF and HARF dried kernels were significantly lower than that of HA dried kernels throughout the storage. Similar results were found in a study investigating HARF heating for stabilization of rice bran, in which PPO and POD activity of high-temperature HARF treated rice bran were significantly lower than that of conventional extruded rice bran (Liao et al., 2020a). Liao et al. (2020b) also found HARF treated wheat germ had significantly lower POD and PPO activity than that treated by fluidized bed heating. The inactivation effect on enzymes could attribute to a higher temperature during HARF heating, which would be beneficial for lowering metabolism of hazelnuts for delaying lipid oxidation during the storage. In short, two-step HA-HARF drying not only retained higher antioxidants in dried kernels than that of HARF and HA drying during the storage, but also lowered enzyme activity of dried kernels compared to HA drying.

### 3.3. Correlation of shell cracking level with physical properties of hazelnut

Since cracking ratio of inshell hazelnuts was affected by drying methods, Table 6 shows the correlation between cracking level and physical properties of hazelnuts dried by different drying methods. Level 2 cracked nuts accounted the most for two-step HA-HARF and HA dried nuts, followed by level 3 cracked ones, while level 3 cracked nuts took up the largest portion of HARF dried nuts. The different results between HARF and two-step HA-HARF was because HARF heating on fresh hazelnuts with higher MC_{wb} induced more RF power input than that of half-dried nuts, leading to more water vapor oppressing the shell (Wang et al., 2014). It was noted that HA dried samples only had 1.47% of level 1 cracked nuts, dramatically lower than that from other two drying methods, which might be explained by the moderate drying temperature making the shell less expanding so that cracks generated during HA drying could hardly recover after cooling of nuts. The weight and diameter of both inshell and kernel were in a significant positive correlation (P < 0.01) with cracking level for all three drying methods as shown in Table 6. Larger mean size inshells (2.35–2.41 cm) and kernels (1.77–1.87 cm) were found at level 3 cracked nuts rather than level 0, level 1, and level 2 nuts.
Similarly, level 3 cracked nuts had the largest mean weight of inshells (4.46–4.78 g) and kernels (1.90–2.05 g) among all nuts. Larger size and weight of nuts usually contained a larger amount of water, thus receiving higher RF power input, which in turn resulted in stronger moisture diffusion and higher moisture vapor pressure between kernel and shell. Besides, kernel weight percentage (M/kernel) was in a significant negative correlation (P < 0.01) with cracking level for two-step HA-HARF and HA drying, i.e., lower M/kernel led to a higher cracking level. Larger air-gap of nuts (Δd) also resulted in a higher cracking level, showing a significant positive correlation (P < 0.01) with cracking level for HARF and two-step HA-HARF drying. The stereomicroscopic images of the longitudinal section of inshells were taken to examine the air-gap volume between shell and kernel for nuts dried by the different methods (Fig. 3). It was observed that air-gap became larger in higher level cracked nuts using all three drying methods. Inshell hazelnuts with larger air-gap between the shell and kernel could also be a potential reason triggering shell cracks as air has low thermal conductivity. Therefore, moisture vapor generated from the kernel accumulated within the shell, slowing down the release of the vapor through the shell (Wang et al., 2018b). Although lower shell density was found as a possible reason for Jefferson nuts more vulnerable to shell cracks than other hazelnut cultivars (Wang et al., 2018b), there was no significant difference in shell density among cracking levels of Jefferson nuts.

### 3.4. Comparison of quality and storability of dried hazelnuts with different cracking levels

Table 7 represents quality and storability parameters of dried hazelnuts with different cracking levels from different drying methods. Since HA dried level 1 cracked nuts only constituted...
Table 7 - Comparison of quality and storability of dried hazelnut with different cracking levels using different drying methods.

<table>
<thead>
<tr>
<th>Drying method</th>
<th>Month</th>
<th>L*</th>
<th>PV¹</th>
<th>PV²</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Level 0</td>
<td>Level 1</td>
<td>Level 2</td>
<td>Level 3</td>
</tr>
<tr>
<td>HARF</td>
<td>0</td>
<td>66.05⁵</td>
<td>63.75⁶</td>
<td>62.82⁶</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>60.81⁵</td>
<td>61.40⁶</td>
<td>60.35⁶</td>
</tr>
<tr>
<td>Two-step</td>
<td>0</td>
<td>65.75⁵</td>
<td>64.15⁶</td>
<td>63.78⁶</td>
</tr>
<tr>
<td>HA-HARF</td>
<td>3</td>
<td>60.95⁵</td>
<td>61.52⁶</td>
<td>60.23⁶</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>63.19⁵</td>
<td>59.58⁶</td>
<td>60.91⁶</td>
</tr>
<tr>
<td>HA</td>
<td>3</td>
<td>63.37⁵</td>
<td>–</td>
<td>61.44⁶</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>61.3⁵</td>
<td>–</td>
<td>60.76⁶</td>
</tr>
</tbody>
</table>

The mean value of each measurement in the same row with same lower letter in the superscript indicated non-significant difference among different cracking levels (P < 0.05).

¹ PV (peroxide value) representing the primary oxidation products (hydroperoxide compounds) was reported as meq O₂/ kg oil.

² FFA (free fatty acid) representing the accumulation of natural fatty acids and hydrolytic rancidity was reported as % oleic acid.

Fig. 3 - Appearance of longitudinal section of inshell hazelnuts with different cracking levels from different drying methods. White arrows indicated the air-gap between shell and kernel.

about 1% of HA dried nuts, they were not included in storability study. In respect to L*, there was no significant difference among level 0, level 1, and level 2 cracked nuts from all three drying methods throughout the storage, while level 3 cracked nuts showed lower L* value than that of level 0 nuts at month 0 for all three drying methods. The PV values of HARF and two-step HA-HARF dried nuts at month 0 and month 3 were very low regardless of the cracking levels, and only PV of HARF dried level 3 cracked nuts at month 6 exhibited relatively higher values than nuts at other cracked levels. There was no significant difference in PV of all HA dried nuts throughout the storage. However, it was noted that PV of HARF and two-step HA-HARF dried level 3 cracked nuts at month 3 and month 6 were significantly lower than that of HA dried level 0 nuts at month 0.
and month 3, respectively. These results indicated that shell cracks did not negatively affect PV of nuts from all three drying methods. General hypothesis concerning about shell cracks was that the crack influenced the integrity of shell, resulting in kernel exposing to oxygen that strengthened the initiation and propagation of lipid autoxidation. However, hazelnut kernel is covered by pellicle, which was still intact after drying based on the observation, probably providing sufficient oxygen barrier together with the woven polypropylene bags to prevent inshell nuts from aggravated oxidation. The much lower PV of HARF and two-step HA-HARF dried nuts was possibly because of inactivation of enzymes involved in the lipid oxidation such as lipoxygenases and POD treated by high temperature of HARF, which was verified by lower POD activity of HARF and two-step HA-HARF dried nuts compared to that of HA dried ones during the storage (Table 5). Regarding other indicators, FFA of dried nuts from each drying method was almost at the same statistical level among all cracking levels. There was no significant difference in K_{322} of HA dried nuts with different cracking levels throughout the storage, and K_{322} of HARF and two-step HA-HARF dried nuts with different cracking levels at month 0 and month 3. However, HARF dried nuts at month 6 showed higher K_{322} along with increasing cracking level, while two-step HA-HARF dried nuts at month 6 had lower K_{322} with increasing cracking level. Considering the results reported in Table 5, the 6-month HARF dried nuts with cracking level from 1 to 3 might exceed the peak of lipid oxidation regarding the rearrangement of the methylene-interrupted double bonds while the two-step HA-HARF dried nuts might not, demonstrating that two-step HA-HARF dried nuts had lower lipid oxidation than that of HARF dried nuts during the storage. Overall, although shell cracks to level 3 slightly impacted kernel color of dried nuts from all three drying methods right after drying, level 3 cracked nuts dried by the two-step HA-HARF exhibited significantly less lipid oxidation than that of HA dried level 0 nuts, meaning cracking did not significantly affect lipid oxidation.

4. Conclusion

This study demonstrated that HARF heating was able to significantly shorten drying time for fresh inshell hazelnuts with high MC_{wb} from 22.0 h to 3.5 h compared to that of HA drying. The two-step HA-HARF drying using 19% MC_{wb} as an intermediate MC_{wb} reduced level 3 cracked nuts (CR_{3}) to ~27 %, lower than that of HARF alone. Both HARF and two-step HA-HARF dried nuts had less lipid oxidation and enzyme activity, higher retention of bioactive compounds, and similar color to HA dried ones during 6-month of storage at 35 °C. Shell cracking was highly correlated with the size and weight of inshell nuts and kernels, kernel weight percentage, and the air-gap volume between shell and kernel. Slightly shell cracking (the crack at width < 0.3 mm or length < 10 mm) did not affect color of dried nuts from all three drying methods during the 6 months of storage. It is noteworthy that shell cracks did not significantly increase lipid oxidation of inshell nuts during the 6-months of storage at 35 °C, probably due to the barrier of intact pellicle to oxygen. Meanwhile, HARF and the two-step HA-HARF dried nuts had lower PV values than that of HA dried ones right after drying and throughout the storage, which might be attributed to significantly reduced oxidative enzyme activity by high temperature RF process.

Studies are needed to look for other strategies and scale-up the two-step HA-HARF drying for improving heating uniformity and reducing severely shell cracked hazelnuts. It is also desirable to investigate the use of HARF heating to dry hazelnut kernels and roast kernels for improving processing efficiency and expanding the potential commercial application of this technology.

Conflict of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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