

Influences of Blanching and Freezing Pretreatments on Moisture Diffusivity and Quality Attributes of Pumpkin Slices During Convective Air-Drying

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1 Abstract

In this study, the relationship between moisture diffusivity in convective air-2 drying and cellular structure through blanching and freezing pretreatment and quality 3 attributes of dried pumpkin slices were evaluated to obtain necessary information for 4 5 designing appropriate drying and pretreatment conditions. The results suggest that the loosely bound structure of cell walls due to blanching, and pores in the tissue formed by 6 7 ice crystals during freezing, increased moisture diffusivity. In addition, the functional and structural damage of cell membranes by the pretreatments, shown by the electrical 8 impedance analysis, is likely involved in moisture diffusivity during drying. In 9 particular, the sample pretreated by both blanching and freezing showed significantly 10 higher values of moisture diffusivity compared to other samples. With regards to quality 11 12 attributes, a decrease in color lightness due to starch gelatinization during blanching 13 dramatically affected the color characteristics of the dried product. Starch gelatinization due to blanching and the formation of pores during freezing significantly influenced the 14 structure of the samples after drying, which affected the rehydration rates and 15 16 mechanical properties.

18	Influences of blanching and freezing pretreatments on moisture diffusivity and
19	quality attributes of pumpkin slices during convective air-drying
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1. Introduction

33	Pumpkin is one of the most important crops cultivated and consumed
34	throughout the world, it is recognized as a highly nutritious foodstuff due to its high
35	content of nutrimental and bioactive components including polysaccharides,
36	carotenoids, vitamins, dietary fiber, minerals, vitamins, and other substances beneficial
37	to human health (Yang et al. 2007; de Escalada Pla et al. 2007; Jacobo-Valenzuela et al.
38	2011; Caili et al. 2006). Pumpkins are often distributed as both raw vegetables and
39	processed products such as frozen, pureed, precooked, or dried materials to increase
40	their storage stability and usability (Gonçalves et al. 2011; Gliemmo et al. 2009; Provesi
41	et al. 2011; Sojak and Głowacki 2010; Nawirska et al. 2009). Among these forms of
42	processing, drying is the most classical method of food preservation for extending shelf-
43	life, creating a lighter weight for transportation, and taking up less space during storage
44	(Dandamrongrak et al. 2002). Despite the development of newer drying techniques,
45	most vegetables are still air-dried because this method of dehydration remains the
46	simplest and most economical (Mazza 1983). However, air-drying has the disadvantages
47	of a longer drying time during the falling rate period, low energy efficiency (Orikasa et
48	al. 2018), and subsequent quality deteriorations such as color fading, browning, and loss
49	of nutrients (Krokida et al. 1998; Liu et al. 2014; Guiné and Barroca 2012; Horuz et al.
50	2017). Therefore, a large amount of data have been previously reported which estimate
51	moisture diffusivity and the modeling of moisture content changes during the
52	convective drying process of fruits and vegetables such as pumpkins (Doymaz 2007;
53	Molina Filho et al. 2016; Guiné et al. 2012), tomatoes (Hawlader et al. 1991), kiwifruit

(Orikasa et al. 2008; Simal et al. 2005), and carrots (Liu et al. 2014; Doymaz, 2004) to
optimize drying conditions to improve efficiency.

56 In these ongoing studies, it has been shown that pretreatments such as blanching and freezing are effective in improving drying efficiency (Lewicki 1998; 57 Mazza 1983; Dandamrongrak et al. 2002; Eshtiaghi et al. 1994) and suppressing the 58 59 increase in sample temperature and preventing the structural deformation (Tatemoto et 60 al. 2016; Ando et al. 2019a; Ando et al. 2019b) of fruits and vegetables. Nieto et al. 61 (1998) investigated the drying characteristics of apples after blanching and suggested that the degradation of the middle lamella and hemicellulosic polysaccharides also 62 affects the drying rate of fruits and vegetables. The high drying rates of prefrozen 63 samples are attributed to the high moisture transfer rates in the tissues due to the 64 65 remarkable disorder of the cell wall structure caused by the formation of ice crystals during freezing (Lewicki 1998; Tatemoto et al. 2016). Furthermore, previous studies 66 claim that destruction of the cell membrane structure and the modification of membrane 67 permeability as a result of freezing pretreatment also increases the drying rate 68 (Vaccarezza et al. 1974; Ando et al. 2016). Therefore, the state of the cell wall and cell 69 70 membrane structures should be investigated to clarify the mechanism that causes 71 changes in drying rates due to pretreatments. It has been reported that the blanching or freezing-thawing pretreatments are effective in facilitating moisture transport within the 72 sample tissues of pumpkins during drying (Arévalo-Pinedo and Murr 2007). However, 73 the relationship between structural changes in cells and the moisture transport 74 75 phenomenon has not been clarified.

76	In this study, observations of cell wall structures and electrical impedance
77	analysis to characterize cell membrane states were applied to evaluate the changes in
78	cellular structures by blanching and freeze-thaw pretreatments in pumpkin slices. These
79	outcomes were then compared with estimated moisture diffusivity during convective
80	air-drying. The dried products are sometimes used as an additive for instant soups,
81	breads and cakes after powdering, but are often used as a cooking ingredient after
82	rehydration. Therefore, evaluation of rehydration characteristics and quality attributes
83	after rehydration can be useful for the quality design of the last products. In our study,
84	internal structures, rehydration characteristics, colors, and mechanical properties of the
85	samples were evaluated to investigate the influence of the pretreatments on the quality
86	attributes of the dried products. The results obtained enable a greater understanding of
87	the drying processes which will be beneficial for designing appropriate drying and
88	pretreatment conditions.
89	
90	2. Materials and methods
91	
92	2.1 Sample preparation
93	Pumpkins (Cucurbita maxima) of the cultivar Kofuki were obtained from a
94	local market and used for experiments within seven days of purchase. Kofuki is a mealy
95	type of pumpkin with relatively high starch and sucrose contents (Cumarasamy et al.
96	2002). The initial moisture contents of the pumpkins were gravimetrically determined to
97	be 5.377 ± 0.017 on a dry basis (g/g) from an average of eight samples. The flesh of the

98	pumpkin was shaped into a discoid shape with a diameter of 20.5 mm and a thickness of
99	3.5 mm. Four types of samples, fresh (non-treated), blanched, fresh-frozen, and
100	blanched frozen, were used for drying. For the blanching procedure, the cylindrical
101	sample was heated in boiling water for 40 s then immediately cooled in iced water. The
102	sample's temperature was maintained at 25 °C in an incubator (CN-25C; Mitsubishi
103	Electric Engineering Ltd., Tokyo, Japan) for 1 h before drying. For the freezing
104	procedure, the sample was wrapped in plastic film and stored in a freezer (HRF-90XT;
105	Hoshizaki Corp., Aichi, Japan) at -20 °C for more than 4 h, then thawed in the
106	incubator at 25 °C for 3 h.
107	
108	2.2 Drying procedure and calculation of effective moisture diffusivity
109	The measured room temperature and relative humidity were approximately
110	20 °C and 49 % respectively. During convective air-drying, samples were placed in a
111	drying chamber (DN-42; Yamato Scientific Co., Ltd., Tokyo, Japan) at controlled
112	temperatures of 40 °C, 60 °C, and 80 °C. The relative humidity in the chamber had been
113	kept below 20 % through the drying. The air velocity in the chamber was 1.5 ± 0.1 m/s
114	on average throughout continuous measurements over 3 min. After specified drying
115	times, the sample was taken out of the chamber and weighed. The moisture content was
116	calculated from both the initial moisture content and the mass.
117	The moisture transport phenomenon during drying is often described by using
118	Fick's diffusion equation. An analytical solution in the case of drying a plane sheet of
119	thin layer assuming one-dimensional moisture transport can be developed as follows

(1)

120 (Crank 1975):

121
$$\frac{M-M_{\rm e}}{M_0-M_{\rm e}} = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left\{-\frac{(2n+1)^2 D\pi^2 t}{4l^2}\right\} \,,$$

122 where M, M_{e} , and M_{0} denote the moisture content, the equilibrium moisture content 123 (equilibrium value of the moisture content determined by air temperature and relative 124 humidity), and the initial moisture content on a dry basis, respectively. D denotes the effective diffusion coefficient ($m^2 \cdot s^{-1}$), *l* denotes the half thickness of the sample slice 125 126 (m), and t denotes the time (s). Constants D and M_e were determined by fitting Eq. (1) to the averaged values of six samples using the least squares method using the software 127 (MATLAB R2018a, The MathWorks, Inc., Natick, USA). Thirty terms of the series 128 were used in the calculation which was sufficient for the convergence. The root mean 129 squared error was calculated as an index of the goodness of fit. 130

131

132 2.3 Electrical impedance analysis

The electrical impedance analysis, widely used to estimate the physiological 133 134 status of various biological tissues (Zhang and Willison 1992; Zhang et al. 1993; Ando et al. 2014; Watanabe et al. 2018), was applied to evaluate cell membrane damage in the 135 136 samples before and after each pretreatment. The impedance magnitudes $|Z|(\Omega)$ and phase differences θ (rad) of the samples were measured at 81 points (logarithmic 137 138 frequency intervals) over a frequency range from 50 Hz to 5 MHz using an impedance analyzer (IM3570, HIOKI E.E. Corp., Nagano, Japan). The electrodes were penetrated 139 from a side of the sample disk with a distance of 10 mm between the electrodes. The 140 electrodes were connected to the impedance analyzer via coaxial cables. The sample 141

142	temperature was maintained in an incubator at 25 °C, and the test was carried out at a
143	room temperature of 25 °C. The measured impedance data were analyzed using the
144	equivalent circuit model for cellular tissues, as previously described (Ando et al. 2014;
145	Ando et al. 2017). The resistance of the extracellular fluid, R_e , the resistance of the
146	intracellular fluid, R_i , and the capacitance of the cell membrane, C_m , were all
147	individually calculated through this model. Detailed procedures for these analyses are
148	described in a previous study (Ando et al. 2017).
149	
150	2.4 Scanning electron microscopy
151	Fresh and pretreated samples were studied via scanning electron microscopy
152	(SEM) to evaluate the cell wall adhesion of tissue samples. The centers of the samples
153	were cut with a sharp knife into small blocks approximately 3.5 mm wide and 1 mm
154	thick, before being rapidly frozen in liquid nitrogen and freeze-dried. The cross-
155	sectional surfaces of the freeze-dried blocks were sputter-coated with gold in a sputter
156	coater (JFC-1500; JEOL Ltd., Tokyo, Japan). These cross-sections were then observed
157	using an SEM (JSM-5600LV; JEOL Ltd.) at an accelerating voltage of 5 kV under high
158	vacuum conditions. The internal structures of the samples after drying were also
159	observed. Small blocks with approximately 2 mm sides were cut with a sharp knife
160	from the center of the dried samples. The cross-sectional surface was then observed in
161	the same manner as previously described.

163 2.5 Color measurements

A color-difference meter (CR-300, Minolta Co., Ltd., Tokyo, Japan) was used to measure the colors of the sample surfaces during drying. After specified drying times, samples were taken out of the chamber and values of a color lightness (*L**),

redness/greenness (a^*), and yellowness/blueness (b^*) of both sides of the samples were measured and averaged. As indices of color characteristics, the chroma, C^* , and the hue angle, h, were calculated via the following equations, respectively:

170
$$C^* = \sqrt{(a^*)^2 + (b^*)^2}$$
, (2)

(3)

171
$$h = 180 \tan^{-1}\left(\frac{b^*}{a^*}\right)/\pi$$
.

172

173 2.6 Rehydration characteristics

174 Each dried sample was immersed in 200 mL of distilled water in a beaker 175 placed in a thermostatically controlled water bath at 30 °C. After the specified 176 rehydration times, the samples were removed from the water and wiped with absorbent 177 paper to remove residual water from the surface. The samples were then weighed with an electric scale. The moisture content on a dry basis (g/g) was calculated from both the 178 179 initial moisture content and the mass. The exponential equation, including the single 180 rate constant as shown below, was used to characterize the rehydration behavior of the 181 samples (Krokida and Marinos-Kouris 2003):

182
$$\frac{M - M_{\rm d}}{M_{\rm s} - M_{\rm d}} = 1 - \exp(-k_{\rm r}t), \qquad (4)$$

where M_d and M_s denote the moisture content of the dried sample and the saturated moisture content, respectively. k_r denotes the rehydration rate constant (h⁻¹), and *t* denotes the time (h). Constants k_r and M_s were determined by fitting Eq. (4) to the 186 averaged values of experimental data using the least squares method.

187

188 2.7 Mechanical properties of the rehydrated samples

Puncture tests of the rehydrated samples were carried out using a universal 189 190 testing machine (5542; Instron, Norwood, MA, USA) equipped with a 500 N load cell. The dried samples were placed on a metal base with a 10 mm diameter hole in the 191 192 center. A cylindrical plunger of 3.2 mm in diameter was then inserted at a speed of 1 mm/s into the center of the flat surface of the sample until it passed through the center 193 of the hole and completely penetrated the sample. The trigger load was set at 0.05 N. 194 195 The thickness of the samples were measured using a caliper. The value of stress was calculated by dividing the force by the cross-sectional area of the plunger. The strain 196 197 was calculated by dividing the displacement by the sample thickness. Fracture stress, $\sigma_{\rm f}$ 198 (Pa), and initial elastic modulus, E (Pa), were calculated as indices of the mechanical 199 properties. The value of E was defined as the slope of the first linear section of the stress-strain curve. The experiments were replicated 12-14 times for each sample. The 200 test was carried out at a room temperature of 25 °C. 201

202

203 2.8 Statistical analysis

Statistical analyses were performed using R software version 3.5.1 (R Core Team). Differences among the means were compared using a Tukey multiple range test with the analysis of variance at a significance level of p < 0.05.

208 **3. Results and discussion**

209

210 Figure 1 shows changes in moisture content versus drying time for the fresh samples. As found in previous studies, the moisture content decreased faster at higher 211 212 drying temperatures, a trend that was also observed in each pretreated sample. The solid lines in Fig. 1 represent the least squares regression analysis of the model, shown as Eq. 213 214 (1), showing the good agreement with the experimental data. The effective diffusion 215 coefficient D values determined from the analysis are shown in Table 1. For each 216 condition, the root mean squared error between the experimental and approximate data were in the range of 0.026 to 0.051. The D value of the pretreated samples tended to 217 218 increase under any temperature during drying, as compared to the fresh samples. In 219 particular, the values of the blanched-frozen samples showed the highest D values at 220 1.10–1.11 times higher than those of the fresh samples. The D values of the fresh-frozen 221 samples were slightly higher than those of blanched samples at 60 °C and 80 °C, whereas those of the blanched and fresh-frozen samples at 40 °C showed almost the 222 same values. These results confirm that blanching and freezing pretreatments are 223 224 effective for facilitating moisture transfer in pumpkin tissues during convective air-225 drying, which is in line with a previous study by Arévalo-Pinedo and Murr (2007) which showed the same effect during the vacuum drying of pumpkins. In addition, the 226 results show that the blanching-freezing pretreatments are the most effective in 227 increasing the drying rate. 228



Figure 2 shows the impedance characteristics on the complex plane (Cole-Cole

230 plot) of the fresh and pretreated samples. The impedance characteristics of the fresh sample displayed a relatively large semicircle with a diameter of 20 k Ω while those of 231 232 the pretreated samples appeared markedly shrunk. It has been reported that the shrinkage of the impedance characteristics of plant tissues occurs during freezing (Wu 233 234 et al. 2008; Zhang and Willison 1992) and heating (Zhang et al. 1993; Halder et al. 2011). The phenomenon is thought to be a result of structural damage to the cell 235 236 membranes. Therefore, the impedance characteristic results suggest that the cell 237 membranes in the pretreated pumpkin tissues were damaged during the blanching and 238 freezing-thawing processes.

The measured impedance data were then analyzed with the modified Hayden 239 model (Ando et al. 2017). The solid lines in Fig. 2 represent approximations given by 240 241 the model fitted by the complex nonlinear least squares method. Note that the straight-242 line sections of the low-frequency areas were removed because they occurred due to the polarization phenomenon at the electrode surface (Pliquett 2010; Kalvøy et al. 2011) 243 and are not related to the cellular structure. The measured impedance and approximate 244 values show a good agreement for all samples, which confirms that the present model is 245 246 acceptable for the application. The estimated values of the parameters in the model are shown in Table 2. The values of cell membrane capacitance, $C_{\rm m}$, was highest in the 247 fresh samples, while the values in other samples decreased. The high capacitance of 248 biological tissues is thought to depend on the lipid bilayer structure of the cell 249 membrane (Ashrafuzzaman and Tuszynski 2012). Therefore, the high $C_{\rm m}$ value of the 250 251 fresh sample potentially occurred as a result of the maintenance of the membrane

252	structures. However, the $C_{\rm m}$ values of the blanched and fresh-frozen samples decreased
253	to 43 % and 27 %, respectively. A decrease in $C_{\rm m}$ has been reported in previous studies
254	on the heating of spinach (Watanabe et al. 2017) and Japanese radish (Ando et al. 2017).
255	This phenomenon was attributed to the thermal denaturation of phospholipids which
256	constitute the cell membrane. It has been previously reported that C_m values decreased
257	to 25 % in apples (Ando et al. 2019b) and 53 % in carrots (Ando et al. 2016) during
258	freezing treatment at -20 °C. These results are thought to stem from the formation of ice
259	crystals during the freezing process. The lower $C_{\rm m}$ values of the fresh-frozen samples
260	suggest that freezing treatment is destructive to the cell membrane structure. The
261	blanched-frozen samples that were stressed by both heating and freezing treatments
262	showed the lowest C_m value (7.5 % of that of the fresh sample), suggesting considerable
263	damage to the cell membrane structure.
264	In healthy cells, the low electrolyte concentration of the extra-cellular fluid and
265	high electrolyte concentration of the intracellular fluid are separated by the
266	permselectivity of the cell membranes. Therefore, the high values of extra-cellular fluid
267	resistance, R_e , and low values of intra-cellular resistance, R_i , of the fresh samples
268	indicates that the cell membranes are functioning normally. In the pretreated samples,
269	the R_e values decreased, and the R_i values increased indicating a difference in electrolyte
270	concentration between the intra- and extra-cellular fluids. This difference occurred as a
271	result of the cell membranes being unable to function correctly. In a study by Halder et
272	al. (2011) the impedance of potato tissues during heating sharply declined in the

temperature range of 52–60 °C. This outcome was found to be due to cell membrane

damage followed by the release of intra-cellular water into the extracellular region. Therefore, changes in the R_e and R_i values of the blanched samples observed in this study were attributed to this same phenomenon due to heating stress to the cell membranes. During the freezing process, the interior of the cells is rapidly dehydrated with ice crystal growth in the extracellular region. This stress causes the alteration of membrane transport properties (Palta 1990) resulting in fatal disruption of the cell membrane (Ando et al. 2012).

281 Changes in the R_e and R_i of the fresh-frozen samples can be explained by this phenomenon. The change ratio of the R_e and R_i values of blanched-frozen samples 282 tended to increase, as with the changes in the $C_{\rm m}$ values, compared to the blanched or 283 fresh-frozen samples. These results suggest that the cell membranes were markedly 284 285 damaged in the blanched-frozen sample. Figure 3 shows the SEM images of crosssections of the fresh and pretreated samples. Cells with an approximate 50 µm diameter 286 in the tissues of fresh samples (Fig. 3-A, a) were densely arranged. The cell walls were 287 split, and the interiors of the cells containing starch particles of approximately 10 µm in 288 diameter were exposed. In the fresh samples, the cell walls strongly adhered to each 289 290 other, whereas the blanched samples showed a loosely bound structure of the cell walls 291 (Fig. 3-B, b), likely due to a β -elimination reaction splitting the homogalacturonan chains that primarily comprise the pectin structure (Sila et al. 2009). In the blanched 292 sample, the cell walls were divided at the middle lamella when the tissue was cut due to 293 this reaction. Therefore, the saclike structures of the cell walls were exposed, and the 294 295 interiors of the cells were not observed. The tissues of the fresh-frozen samples showed

sparse structures presumed to have occurred due to ice crystal formation during freezing
(Fig. 3-C, c). Although the structures are largely disrupted in the fresh-frozen samples,
the separations between the cell walls as seen in the blanched samples were not
observed. In the blanched-frozen samples, a sparse structure was observed as with the
fresh-frozen sample, and the cell walls were largely separated compared to the
blanching samples (Fig. 3-D, d).

302 In terms of the relation of the damages to the cell membrane caused by the 303 pretreatments and the moisture diffusivity, the samples with a higher change ratio of the parameters, i.e., with more significant damage to the cell membranes, had higher D 304 values. This result is consistent with a previous study by Ando et al. (2012) which 305 306 claims that structural and functional damage to cell membranes leads to an increase in 307 water permeability and accelerates moisture transfer in plant tissues. In addition, it was assumed that damage to the cell walls, i.e., the separation of cell walls attributed to 308 309 changes in pectin structures by heating, and physical damage due to the growth of ice crystals during freezing also contributed to an increase in moisture diffusivity. In 310 particular, the highest D values of the blanched-frozen samples were attributed to the 311 312 marked damage to both cell membranes and cell wall structures, suggesting that 313 blanching-freezing pretreatment is effective in increasing the drying rate and reducing the drying time required. 314

Figure 4 shows the changes in the color parameters, lightness L^* , chroma C^* , and Hue angle *h* during drying at 60 °C. The lower values of L^* and C^* of the blanched and blanched-frozen samples before drying compared to other samples is likely due to

leakage of the gas spaces present in the tissue, as reported in studies on the vacuum 318 impregnation of pears (Perez-Cabrera et al. 2011) and papayas (Yang et al. 2017). It is 319 theorized that the internal gas expands and therefore forces its way out of the tissue 320 during blanching. This occurrence results in the replacement of the gas phase by the 321 322 liquid phase, inducing more homogenous refractive indices in the tissues. This event promotes light absorption against scattering resulting in the tissue samples becoming 323 324 transparent with decreasing in lightness and chroma (Chiralt and Talens 2005). 325 Furthermore, swollen starch particles due to gelatinization during heating may have also contributed to the optical properties. The values of L^* and C^* of the fresh-326 frozen samples were slightly lower than those of the fresh samples due to the 327 328 destruction of cellular structures and the inflow of cellular water into the intercellular spaces during freezing. The L^* values of the blanched and blanched-frozen samples 329 tend to decrease even further from the initial low values. This result may be attributed to 330 the high amorphous starch fractions of the gelatinized starch maintained during drying 331 (Xiang et al. 2018) which indicates restrained light scattering and low lightness. The L^* 332 and C^* values of the fresh-frozen and blanched-frozen samples tend to decrease more 333 334 substantially than those of the fresh and blanched samples during the drying process. This result may be explained by the oxidization of carotenoids (Song et al. 2017) which 335 is prone to occur in the frozen-thawed tissues where the cell walls and membranes are 336 significantly destroyed (Park 1987). The fact that the decrease in the C^* value was 337 almost depended on the decrease in the b^* value (decrease in yellowness) supports this 338 339 view. The values of the hue angles h of the fresh samples were nearly constant during

drying. However, the values of other samples decreased, especially for the blanched and 340 blanched-frozen samples which showed lower values compared to the fresh-frozen 341 samples. Decreases in hue angles during the air-drying of blanched pumpkins have been 342 previously reported (Song et al. 2017). This phenomenon is thought to be a result of the 343 344 degradation of carotenoid pigments and the formation of brown compounds due to Maillard reactions during drying. However, in this study subequal decreases in h values 345 346 were observed even when drying at low temperatures. This result suggests that the 347 reaction that occurs in the blanching process forms brown compounds, then they are concentrated with drying and strongly reflected in the h values of gelatinized dried 348 349 tissues with lower scattering and higher transparency. These trends of color change are similar at other drying temperatures, and they are dependent on moisture content, not 350 351 drying time (data not shown).

Figure 5 shows the internal structure of the pumpkin slice samples after drying 352 353 at 60 °C. In the fresh samples, structures with densely packed starch particles approximately 10 µm in diameter are observed (Fig. 5a). Structures of the blanched 354 samples show a smooth surface (Fig. 5b) as observed in dried starch noodles (Xiang et 355 356 al. 2018). This result demonstrates the state in which gelatinized starch particles are accumulated and densely compressed by the drying shrinkage. In the fresh-frozen 357 samples, although starch particles were observed as with the fresh samples, pores 358 formed due to ice crystal formation during freezing were distributed throughout the 359 inside (Fig. 5c). The blanched-frozen samples also showed a porous structure with many 360 361 airspaces (Fig. 5d), though the starch particles were gelatinized entirely. Figure 6 shows

changes in the moisture content during rehydration at 30 °C for each dried sample. The 362 moisture content of the blanched-frozen samples reached its saturation at the lowest 363 364 time of 1 h, while other samples required more than 3 h. The estimated values of the rehydration rate constant $k_{\rm r}$ are 0.96, 2.10, 2.25, and 7.38 h⁻¹ for the fresh, blanched, 365 366 fresh-frozen, and blanched-frozen samples, respectively. Here, the determination coefficients for all samples are greater than 0.99, which indicates the model was 367 368 appropriate for explaining the rehydration phenomenon. Water can generally be 369 absorbed more efficiently by amorphous food materials than by crystalline materials during hydration (Xiang et al. 2018). Therefore, the higher k_r values of the blanched and 370 blanched-frozen samples could be explained by their higher amorphous starch fractions 371 372 due to gelatinization during blanching. In addition, the porous internal structures of the 373 fresh-frozen and blanched-frozen samples means that there is a larger surface area to absorb water which may also contribute to the high values of k_r . These results show that 374 the blanching and freezing treatments before drying are effective in increasing the 375 rehydration rate of the dried samples. 376

Figure 7 shows the representative stress-strain curves by the puncture test of the fresh and pretreated pumpkin slices before drying and after drying-rehydration. Before drying, the stress of the fresh sample was significantly higher than that of pretreated samples, which is considered to be due to the integrity of the cellular structure. Although the value was decreased in the samples after drying-rehydration, the stress of the fresh sample was higher than that of other samples as before drying. Table 3 shows the fracture stress, $\sigma_{\rm f}$, and initial modulus, *E*, of the samples before drying and

after drying-rehydration. Before drying, the highest values of $\sigma_{\rm f}$ and E are observed in 384 fresh samples attributable to the maintained integrity of the cell walls and cell 385 386 membrane structures. However, in the blanched samples, the $\sigma_{\rm f}$ value markedly decreases (94 % decrease) compared to the fresh samples, which is potentially 387 388 attributable to the loosely bound structure of the cell walls, as shown in Fig. 3b, due to the degradation of pectin structures caused by heating (Sila et al. 2009). The $\sigma_{\rm f}$ value of 389 390 the fresh-frozen samples also decreased purportedly due to the destruction of the cell 391 wall structures caused by ice crystal formation during freezing and cell membrane damage demonstrated by the impedance analysis. However, the $\sigma_{\rm f}$ value of the fresh-392 frozen samples was slightly retained compared to the blanched samples because the 393 dissociation of the cell walls by heating did not occur. 394 395 Conversely, the fresh-frozen sample showed a marked decrease in the *E* value

396 (96 % decrease). The reduction of turgor pressure due to changes in cell membrane 397 states is a major factor for the mechanical parameters, especially the elasticity of 398 vegetable tissues (Chassagne-Berces et al. 2009; Ando et al. 2012). Therefore, the low *E* 399 value of the fresh-frozen samples can be explained through structural and functional 400 damage to the cell membranes by freezing treatment, as suggested by the impedance 401 analysis. The blanched-frozen samples which were subjected to heating and freezing 402 stresses show markedly decreased values of $\sigma_{\rm f}$ and *E*.

403 As for the samples after drying-rehydration, although the fresh sample had the 404 highest values of $\sigma_{\rm f}$ and *E* compared to other samples, they were significantly decreased 405 compared to the fresh sample before drying, likely due to structural destruction during

406	drying (Ando et al. 2014), indicating that it is difficult to restore the values by
407	rehydration. In the blanched samples, the values of $\sigma_{\rm f}$ and <i>E</i> decreased to 33 % and
408	13 %, respectively, through drying-rehydration treatment for the same reason. The lower
409	value of E of the fresh-frozen sample than that of the blanched sample indicates that the
410	formation of pores in tissues due to freezing treatment results in a further reduction in
411	elasticity of the rehydrated sample. The values of $\sigma_{\rm f}$ and E of the blanched-frozen
412	sample did not change before or after drying-rehydration, indicating that structural
413	destruction occurs mostly before drying and no further mechanical change occurs
414	during the drying-rehydration process. The results reveal that the blanching and freezing
415	pretreatments were effective in increasing the rehydration rate of the dried materials but
416	lead to a reduction in the parameters of mechanical properties.

417

418 4. Conclusions

419

This study aimed to clarify the relationship between the moisture diffusivity of 420 421 pumpkin slices during convective air-drying and changes in the cellular structure due to blanching and freezing pretreatments, as well as the quality attributes of the dried 422 products. The loosely bound structure of the cell walls likely due to a β -elimination 423 reaction splitting the homogalacturonan chains caused by thermal blanching was 424 observed in blanched sample, whereas the formation of pores presumed to have 425 426 occurred due to ice crystal development during freezing was observed in frozen samples, respectively. In particular, the samples treated with both blanching and 427

freezing showed significantly destroyed structures of the cell walls. The electrical 428 impedance analysis shows a decrease in the cell membrane capacitance and the changes 429 430 in the intra- and extra-cellular fluid resistances those reflect the structural and functional damages to the membranes for the pretreated samples. This trend was remarkable in the 431 432 blanched-frozen sample, and that of blanched and fresh-frozen samples was almost the same level. The estimated value of moisture diffusivity was lowest in fresh samples at 433 434 each drying temperature, potentially due to the cell wall and cell membrane structures 435 maintaining their integrity and restraining water transfer. Among pretreated samples, the 436 blanched-frozen samples show the highest value of moisture diffusivity at 1.10–1.11 times higher than those of the fresh samples at each drying temperature. This result 437 suggests the structural and functional damages to the cell walls and cell membranes by 438 439 the pretreatments facilitates moisture transfer increasing the drying rate. Changes in color were found to appear predominantly in the blanched samples, 440 and the influence of freezing was limited. This change is potentially attributed to an 441 increase in transparency due to starch gelatinization, and the formation of brown 442 compounds concentrated with drying. Moreover, starch gelatinization by blanching and 443 444 the formation of pores during freezing greatly influenced the structures of the dried 445 samples, resulting in increases of the rehydration rate. In particular, the rehydration rate of the blanched-frozen samples showed the highest value, 7.7 times higher compared to 446 the fresh sample. However, significant reductions in the parameters of the mechanical 447 properties by the pretreatments were observed in the mechanical test of the sample after 448 drying-rehydration. These findings may be valuable in predicting drying times and 449

450	quality attributes and designing appropriate drying and pretreatment conditions. The
451	calculation of total energy spent throughout the process of pretreatment and drying and
452	the optimization of the process conditions taking into account for the consumer
453	acceptability of the texture and other quality attributes should be addressed in future
454	work.
455	
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457	
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459	
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Figure 1 Changes in the moisture ratio during drying of the non-treated pumpkin slices. The data are mean values of 6 replicates. The solid lines represent approximations given by Eq. (1).



Figure 2 Representative Cole-Cole plots for the fresh, blanched, fresh-frozen, and blanched-frozen pumpkin slice samples. The solid lines represent approximations given by the modified Hayden model (Ando et al. 2017).



Figure 3 Scanning electron micrographs of cross sections of the fresh (A, a), blanched (B, b), fresh-frozen (C, c) and blanched-frozen (D, d) pumpkin slices (A, B, C and D: $\times 100$ images, a, b, c and d: $\times 500$ images).



(b) Chroma C*



(c) Hue angle *h*



Figure 4 Changes in the lightness (a), chroma (b) and Hue angle (c) of pumpkin slice samples during convective air-drying at 60 °C. The data are mean values of 8 replicates. Bars denote standard error.



Figure 5 Scanning electron micrographs of cross sections of the fresh (a), blanched (b), fresh-frozen (c) and blanched-frozen (d) pumpkin slices after convective air-drying at $60 \,^{\circ}$ C.



Figure 6 Changes in the moisture content of the pumpkin slice samples dried at 60 °C during rehydration at 30 °C. The data are mean values of 6 replicates. The solid lines represent approximations given by the exponential model shown as Eq. (4).





(b) After drying and rehydration



Figure 7 Representative stress-strain curves by the puncture test of the fresh and pretreated pumpkin slices before drying (a), and after drying at 60 $^{\circ}$ C and rehydration at 30 $^{\circ}$ C (b).

Figure captions

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Figure 7 Representative stress-strain curves by the puncture test of the fresh and pretreated pumpkin slices before drying (a), and after drying at 60 °C and rehydration at 30 °C (b).

	Fre	sh	Blan	ched	Fresh-1	frozen	Blanched	l-frozen
Drying temperature	$D imes 10^{10}$ (m ² /s)	RMSE (-)	$D imes 10^{10}$ (m ² /s)	RMSE (-)	$D \times 10^{10}$ (m ² /s)	RMSE (-)	$D imes 10^{10}$ (m ² /s)	RMSE (-)
40 °C	1.06	0.039	1.13	0.045	1.11	0.051	1.16	0.049
60 °C	2.11	0.034	2.20	0.034	2.30	0.035	2.32	0.032
80 °C	3.59	0.030	3.73	0.028	3.89	0.026	4.00	0.028

 Table 1
 Effective diffusion coefficient of moisture during convective air-drying of pumpkin slices estimated from Eq. (1).

RMSE: root mean squared error for the model fitting.

	$C_{\rm m}~({\rm pF})$	$R_{\rm e}$ (k Ω)	$R_{\rm i}$ (k Ω)	
Fresh	$509^{a} \pm 8$	$22.47^{a} \pm 0.61$	$0.80^{\circ} \pm 0.03$	
Blanched	$219^{b}\pm14$	$1.39^{b} \pm 0.03$	$2.56^{b} \pm 0.17$	
Fresh-frozen	$138^{\circ} \pm 17$	$1.22^{b} \pm 0.05$	$2.03^{b} \pm 0.14$	
Blanched-frozen	$38^{d} \pm 2$	$1.02^{b} \pm 0.03$	$6.99^{a} \pm 0.43$	

Table 2 Equivalent circuit parameters obtained from the model fitting.

 C_m : capacitance of cell membrane, R_e : extracellular fluid resistance, R_i : intracellular fluid resistance. The values of C_m , R_e and R_i represent the mean values of 12 replicates (± standard error). Different superscripts indicate significant differences (p < 0.05) between the means compared by a Tukey' s multiple range test.

Condition	Fracture stress $\sigma_{\rm f}\left(Pa\right)$	Initial modulus E (Pa)				
Before drying						
Fresh	$36535^{a} \pm 1908$	$44090\ ^{a}\pm 3399$				
Blanched	$2129^{bc}\pm114$	$16649^{b} \pm 1167$				
Fresh-frozen	$4607^{b} \pm 518$	$1770^{\circ}\pm149$				
Blanched-frozen	$326^{\circ} \pm 18$	$1388\degree\pm128$				
After drying at 60 °C and rehydration at 30 °C						
Fresh	$1918^{a} \pm 143$	$7718^{a}\pm534$				
Blanched	$710^{\rm b}\pm40$	$2174^{b} \pm 361$				
Fresh-frozen	$722^{\text{ b}}\pm108$	$1018^{b} \pm 197$				
Blanched-frozen	$442^{b} \pm 39$	$1025^{b} \pm 114$				

 Table 3
 Mechanical properties of the pumpkin slice samples.

The values represent the mean values of 12–14 replicates (\pm standard error). Different superscripts indicate significant differences (p < 0.05) between the means compared by a Tukey' s multiple range test. The values of the samples before drying and after drying-rehydration were separately compared.