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Storage of wafer cookies: Assessment by destructive techniques, and non-destructive spectral detection methods

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1 **Storage of wafer cookies: assessment by destructive, spectroscopic, and** 2 **hyperspectral methods**

3

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7

8 **Abstract**

9 Wafer cookies combine two or more layers of wafer sheets with intermediate layers of cream filling
10 and later coating with chocolate. During storage, wafer cookie quality, especially in terms of
11 mechanical properties, is mainly affected by moisture migration from the cream or chocolate and
12 moisture absorption from air. This study aimed to assess the storage of wafer cookies by destructive
13 (water activity, mechanical properties, and sensory acceptance) and non-destructive methods (image
14 analysis, NIR spectroscopy and hyperspectral imaging HSI). Furthermore, two packaging types were
15 considered. Samples were stored at 18°C (RH=50%) and analysed after 2, 4, 5, 6, 7 and 8 months.
16 Good linear relations (R^2 up to 0.84) were found between water activity and mechanical parameters,
17 confirming the dependence between textural aspects and water content. By adding a multi-material
18 packaging layer, the shelf life significantly increased in terms of sensory acceptance (crispness). No
19 significant differences were found between the surface colour parameter (white index) attributable to
20 fat bloom formation. PCA results of NIR and HSI spectra showed a clear separation between samples
21 acquired at time 0 and those analysed during storage that was related with the packaging type and
22 storage time. PLS models developed to estimate the storage time showed R^2 ranging from 0.926
23 (RMSECV=0.63 months) to 0.960 (RMSECV=0.52 months), while the water activity ranged from
24 0.858 to 0.928 (RMSECV=0.02). The PLS models based on HSI spectra were used to obtain
25 predictive images of water activity or storage time.

26

27 **Keywords:** wafer, hyperspectral imaging, NIR, storage, packaging.

28

29 **1. Introduction**

30 Wafer cookies are the result of combining two or more layers of low sugar wafer sheets with
31 intermediate layers of cream filling and later coating with chocolate (Dogan, 2006; Tiefenbacher,
32 2017). Between packaging and consumption, and thus during storage, the quality of wafer cookies is
33 mainly affected by absorption or moisture shifting. Two potential main sources of moisture absorbed
34 by the wafer can be identified: moisture migration from cream filling or enrobing and moisture
35 sorption from humid air. In the first case, the wafer sheet conditioning (maturing) until reaching the
36 cream and enrobing water activities can significantly reduce the migration. Instead, moisture
37 migration from air during storage is observed when the moisture barrier of the packaging film is not
38 sufficient or sealing of packages is defective (Tiefenbacher, 2017). Wafer moisture modification can
39 directly compromise the product's textural parameters. The texture at first bite and how it breaks up
40 and dissolves in the mouth are the main characteristics, and the crispness is considered a fundamental
41 textural attribute to consider during storage. Water activities of 0.45 and 0.59 are suggested as a
42 significant threshold to lose the wafer's characteristic texture properties (Martinez-Navarrete et al.,
43 2004; Tiefenbacher, 2017). Other food quality parameters, such as oxidation and rancidity, are less
44 significant due to the progress in quality of ingredients and in packaging (Tiefenbacher, 2017). For
45 wafer cookies coated with chocolate, it is important to consider surface colour changes due to the
46 presence of fat bloom. Fat bloom is considered a chocolate defect and is generally characterized by
47 the formation of a dull-white film on the chocolate's surface, negatively impacting quality (Gatti et
48 al., 2021). The exact causes of blooming are still unknown, but improper tempering conditions and
49 temperature fluctuations during storage can promote fat migration through the chocolate's particle
50 matrix with subsequent recrystallization on the surface (Aguilera and Briones, 2005) .

51

52 Normally, water activity and textural parameters are determined by destructive methods.
53 (Mohammed et al., 2014, 2013) using uni-axial compression and three-point bending experiments on
54 confectionery wafers to characterise their material behaviour. Concerning wafer sheets, relations
55 between mechanical parameters obtained by a three-point bend test (Force and the distance at the
56 fracture point) and water activity were investigated by (Martinez-Navarrete et al., 2004). Recently,
57 (Nasabi et al., 2021) evaluated the structural properties of wafer sheets made with various sources of
58 grains using a three-point bending probe. Based on our knowledge, no studies on the mechanical
59 parameters of wafers during storage have been carried out.

60 Considering non-destructive methods, several studies have reported successful application of near
61 infrared (NIR) spectroscopy for moisture and textural determination of intact bakery products as well
62 as their evolution during storage (Cevoli et al., 2015; Chakravartula et al., 2019; Sørensen, 2009; Xie
63 et al., 2003). Furthermore, NIR spectroscopy was applied to quick identification of fat bloom in
64 chocolates after storage at inadequate temperatures of transportation and storage (Gatti et al., 2021).

65 Hyperspectral imaging (HSI) is a technique that integrates spectroscopy and digital imaging to
66 simultaneously obtain a spectral and spatial three-dimensional data set (hypercube). Recently, HSI
67 has been applied to study the quality and safety attributes of food products as affected by processing
68 and storage (Andresen et al., 2013; Liu et al., 2015). Concerning storage of bakery products,
69 (Sricharoonratana et al., 2021) used the NIR-HSI (935-1720 nm) as a non-destructive method to
70 determine the storage time of freshly baked cakes packed in a polystyrene plastic box. The predictive
71 images showed different colours based on their storage time (0-9 days) that was predicted by a partial
72 least square (PLS) model ($R=0.835$, $RMSECV=1.242$ day). (Lancelot et al., 2020) investigated the
73 possibility of using NIR-HSI (950-2500 nm) to quantify the moisture content in two different
74 commercial biscuits stored at different levels of water content (from 0.114 to 0.907). Multi-linear
75 regression (MLR) was used to estimate the moisture content (2.54-23.18 %), reporting an R^2 higher
76 than 0.92. Using the MLR model, false colour images were proposed according to the water content.
77 (Whitworth et al., 2010) presented a spatial evaluation of the moisture distribution for baguette slices

78 (internal crumb) stored for 96 hours, obtained by applying a calibration modified PLS model to
79 hyperspectral images (950-2495 nm).

80 Research on the application of HSI on chocolate or cocoa-based products is scarce and no studies
81 have assessed storage. Some studies have examined the authentication and the prediction of
82 fermentation quality parameters of cocoa beans (Caporaso et al., 2018; Cruz-tirado et al., 2020). Only
83 one investigation on chocolate has been carried out and used HSI (4000 to 675 cm^{-1}) combined with
84 multivariate curve resolution to analysis of the constituents of commercial chocolate samples (Zhang
85 et al., 2015).

86 The aim of the present analysis was to study the storage of wafer cookies coated with chocolate using
87 both destructive (wafer sheet water activity, mechanical properties, and sensory crispness acceptance)
88 and non-destructive methods (image analysis, NIR spectroscopy and Vis/NIR-HSI). Furthermore,
89 two different types of packaging were considered.

90

91 **2. Materials and Methods**

92 **2.1 Samples**

93 The samples examined were vanilla cream-filled wafers (three layers) covered by a thin layer of dark
94 chocolate of about 0.6 mm. The products were supplied by Babbi GROUP SpA. Italy immediately
95 after production. The ingredients of all components (wafer sheet, cream, and chocolate) are reported
96 in Table 1. Half of the samples (n=144) were packaged using the standard packaging (S) adopted by
97 the company, while the other half (n=144) were vacuum-packed (V) adding a multi-material
98 packaging layer (PET and aluminium). The list of the packaging materials is reported in Table 2.
99 Subsequently, samples were stored at 18°C and relative humidity of 50 % for 8 months. Twenty-four
100 samples for each packaging type were collected and analysed after 2, 4, 5, 6, 7 and 8 months of
101 storage. Twelve of these for each storage time were subjected to destructive analysis, while the
102 remaining 12 to non-destructive analysis.

103

104 **2.2 Destructive analysis**

105 *2.2.1 Mechanical properties*

106 A three-point bending test was used to evaluate the mechanical properties of samples according to
107 (Tiefenbacher, 2017) that suggested this test for cream-filled sugar wafer cookies. Measurements
108 were made on 12 samples for each storage time and packaging using a texture analyser (Zwick Roell
109 500N) equipped with a load cell of 100 N. The sample was placed on two holders fixed at 35 mm
110 distance, while a parallelepiped (6x6x50 mm) was used as probe. The probe moved down for 20 mm
111 at a speed of 1.5 mm/s.

112

113 *2.2.2 Water activity*

114 The water activity (Aqualab Pawkit, water activity meter, Decagon Devices) was measured on the
115 internal wafer sheets collected from each sample (12 samples for each storage time and packaging
116 type). Before proceeding with the analysis, the cream and chocolate were removed.

117

118 *2.2.3 Sensory test*

119 The trained panel was composed of six people employed by the collaborating company. Each panellist
120 evaluated texture at first bite assigning a score between 1 and 10 based on the crispness (a
121 combination of the type of sound (i.e., short snapping and longer cracking sounds) and the force to
122 bite and chew as perceived on first bite (Duizer and Winger, 2006)). According to the criteria adopted
123 by the company, the acceptable limit of crispness was 6 (1 = not crisp - totally not acceptable, 10 =
124 very crisp -totally acceptable).

125

126 *2.2.4 Data analysis*

127 Significant differences between means of the destructive parameters at different storage times, within
128 the same packaging type, were explored using ANOVA (post doc: Tukey's HSD, p-level < 0.05).
129 Subsequently, possible statistical relations between parameters were investigated (Matlab2018a[®]).

130 **2.3 Non-destructive analysis**

131 *2.3.1 Image analysis*

132 Colour surface evaluation was carried out using an electronic eye (visual analyser VA400 IRIS Alpha
133 M.O.S., France) composed of a chamber (420 x 560 mm), LED light system [98 CRI (colour
134 rendering index), D65 (light of a cloudy day at 12:00), 6700 °K (colour temperature)] and a CCD
135 camera (16 million colours). Light and camera were in the upper part of the system. The built-in zoom
136 was automatically calibrated by the software (E-Eye software Alpha-Soft, version 14.0). The analysis
137 was set up with a resolution of 1214×911 pixels. Images were acquired by placing samples at 80 mm
138 from the camera. Raw images were processed in RGB scale and subsequently converted in Ciel* a*b*
139 scale. For each wafer image, the white background was automatically removed and the mean L*, a*
140 and b* values were calculated considering the wafer region. Subsequently, the white index (WI) was
141 calculated, as reported by (Kumara et al., 2003; Popov-Raljić and Laličić-Petronijević, 2009):

$$142 \quad WI = 100 - [(100 - L^*)^2 + a^{*2} + b^{*2}]^{1/2}$$

143

144 *2.3.2 NIR spectroscopy*

145 Samples were evaluated with an FT-NIR spectrophotometer (MATRIX™ –F, Bruker Optics) in
146 diffuse reflectance in the range from 833 to 2500 nm (8 cm⁻¹ resolution). Three scans were obtained
147 by placing the optical fibre probe (IN 261, Bruker Optics, Mass., USA) in direct contact with the
148 product surface at three different points. The background was acquired by placing the probe in a
149 support covered made of spectralone and subsequently subtracted from each spectrum. The mean
150 spectrum of the three acquisitions was calculated and used for chemometric elaboration.

151 The first part of the spectra until 1000 nm was deleted as it contains no useful chemical information,
152 but only instrumental noise. Subsequently, absorbance data were normalised using the Standard
153 Normal Variate (SNV) technique. To remove both additive and multiplicative effects, spectra were
154 treated by applying the first Savitzky–Golay derivative (D1) and then mean centered (MC). PCA

155 analysis was used as an explorative technique to group samples as a function of packaging type and
156 storage time. Subsequently, PLS models were developed to estimate the storage time and water
157 activity considering the samples together and split by type of packaging. The models were validated
158 by the venetian blind cross-validation method (segments: 10), while the permutation test was used to
159 evaluate the model robustness and significance. To avoid the model over-fitting, the optimal number
160 of latent variables (LV) were chosen by detecting the global minimum of root mean square error in
161 cross validation (RMSECV).

162 The data elaboration was carried out by using PLS Toolbox for Matlab2018a®.

163

164 2.3.3 Hyperspectral imaging

165 A push-broom linear array hyperspectral camera working from 400 to 1000 nm for a total of 272
166 bands (Nano-Hyperspec VNIR, Headwall Photonics, Inc., Fitchburg, MA, USA) and characterised
167 by a spatial resolution of 640 points was adopted to acquire HS images. The camera was equipped
168 with a focal lens of 17 mm, and was installed with the optical axis perpendicular to the underlying
169 conveyor belt (speed of 8 mm/s) at a height of 540 mm. Two halogen lamps (120 W), inclined by
170 15 °, were installed at about 320 mm from the conveyor belt plane. Ambient light was isolated using
171 a specific box. The sensor exposure time and frame period were set at 30 ms. The white and dark
172 reference reflectance spectra were obtained by acquiring a high-reflectance matte white panel (R_w)
173 and covering the camera lens with its cup (R_d), respectively. Each sample was scanned longitudinally
174 to obtain the raw reflectance spectrum (R_r). The calibrated reflectance (R_c) was subsequently
175 calculated:

$$176 \quad R_c = \frac{R_r - R_d}{R_w - R_d}$$

177 The region of interest (ROI) was selected by the k-means clustering method (Euclidean distance),
178 performed using 3 clusters (figure 1) [cluster 1: background to be discarded (light grey); cluster 2:

179 sample shadow (dark grey); cluster 3: ROI (black)]. For each sample, the ROI mean spectrum was
180 calculated and converted from reflectance to absorbance.

181 Spectral bands between 400 and 450 nm were removed due to a low signal-to-noise ratio produced
182 by the sensor.

183 The spectra were smoothed (Savitzky-Golay method; polynomial order: 2; smoothing points: 15) to
184 reduce noise from the spectra (Amigo et al., 2013), and subsequently pre-processed by SNV, D1 and
185 MC. AS for NIR spectroscopy, PCA analysis was used as an explorative technique to group samples
186 as a function of packaging type and storage times, and the PLS models to estimate the storage time
187 and water activity considering together the samples and split by type of packaging. Predictive models
188 were validated by the venetian blind cross-validation method (segments: 10), and to avoid the model
189 over-fitting, the optimal number of latent variables (LV) were chosen by detecting the global
190 minimum of root mean square error in cross validation (RMSECV). Permutation test was used to
191 evaluate the model robustness and significance. Finally, PLS models were used to obtain predictive
192 spectral maps of water activity or storage time distribution. Results of the estimated value from the
193 models were interpolated in each pixel of the hyperspectral image.

194 The data elaboration was carried out by using PLS Toolbox and MIA Toolbox for Matlab2018a®.

195

196 **3. Results**

197 The mean and standard deviation of the water activity of the wafer sheet measured during the storage
198 are reported in Table 3, for both the packaging type (standard: S, vacuum: V). A significant increase
199 was observed from 0 to 2 months of storage (passing from 0.27 to 0.38) after which the mean values
200 remained constant until 5 (S) and 6 (V) months. Finally, the water activity increased reaching values
201 of 0.47 (S) and 0.42 (V) after 8 months of storage. For the same storage time, significant differences
202 between the mean values of the two types of packaging were achieved starting from 6 months. To
203 avoid moisture migration from the cream during storage, the wafer sheets were conditioned until
204 reaching a water activity value near to that of the cream (0.26). Consequently, the increase in water

205 activity is probably due to ambient humidity. The primary packaging was not sealed, and
206 consequently the first A_w increases (from 0.27 to 0.38), found for both packaging types, was likely
207 due to the humidity present in the headspace of the second packaging.

208 From force-distance curves, the force (F_{max}) and distance (D) at the first yield points were identified
209 and reported in Table 3 as a function of storage time and packaging type. An initial increase in both
210 mechanical parameters was observed passing from 0 to 2 months. These trends can be attributed to
211 the significant A_w increase (from 0.28 to 0.38), and reflect in an increase in sample deformability,
212 which in turn increases its resistance to fracture. For both packaging types, from 2 to 6 months, no
213 significant differences were observed between the means, with limited variations in water activity
214 (from 0.38 to 0.40). After 6 months, the D parameter increased reflecting the progressive plasticising
215 effect of water in the product, becoming more deformable in line with the increase in water mobility
216 associated with water interaction (Martinez-Navarrete et al., 2004). Good linear relations ($S: R^2=0.84$;
217 $V: R^2=0.78$) were found between water activity and mean values of D , confirming the dependence of
218 mechanical behaviour on the effect of the water on the water sheet (Tiefenbacher, 2017).

219 evaluated texture at first bite assigning a score between 1 and 10 based on the crispness (a
220 combination of the type of sound (i.e., short snapping and longer cracking sounds) and the force to
221 bite and chew as perceived on first bite (Duizer and Winger, 2006)). According to the criteria adopted
222 by the company, the

223 Sensory acceptance related to product crispness, was evaluated by six trained panellists. The scale
224 ranged from 1 (not crisp - totally not acceptable) to 10 (very crisp -totally acceptable) and the
225 acceptable limit of crispness was 6. At lower values, the product is commercially unacceptable. The
226 mean values, as a function of storage time and packaging type, are shown in Figure 2a. Sensory
227 acceptance related to crispness linearly decreased with storage time, which was more pronounced for
228 samples stored in standard packaging ($S=-0.6347$; $V=-0.2186$). Furthermore, the acceptance of
229 samples packaged under vacuum did not reach the unacceptable level (6), while the samples in
230 standard packaging reached this level after 6/7 months. Accordingly, the shelf life of the products

231 packaged in standard conditions defined by the company is 6 months. Relations between sensory
232 acceptances and water activities (Figure 2b) or mechanical parameters D (Figure 2c) were
233 investigated. In both cases (A_w and D), stronger linear relations were found considering the samples
234 packaged under vacuum ($R^2=0.82$ and 0.91), probably due to a more accentuated linearity between
235 acceptance and storage time. The acceptance limit (6) corresponds to a wafer sheet water activity of
236 about 0.44. This A_w value is in agreement with those (0.40-0.45) reported by (Tiefenbacher, 2017)
237 regarding the limit to define “good” and “compromised” wafers in terms of sensory (textural)
238 properties.

239 Chocolate colour changes, mainly related to fat bloom, were evaluated considering the WI as reported
240 by other studies (Kumara et al., 2003; Popov-Raljić and Laličić-Petronijević, 2009). Bloom can
241 appear in either a uniform white film or as spots randomly placed on the chocolate surface.
242 Consequently, mean WI values calculated on the entire top surface of the wafers were considered.
243 Means and standard deviations obtained considering 12 samples for each storage time and packaging
244 type are reported in Table 3. These values are in the same range as those reported in several studies
245 on dark chocolate (Popov-Raljić and Laličić-Petronijević, 2009). No significant differences were
246 found between the mean values with the same packaging time, and between the two different
247 packages for the same storage times. Very restricted variations of mean values were observed, passing
248 from 31.9 to 32.8. This confirms that there was no evident fat bloom formation during storage due to
249 improper temperature or temperature fluctuations during storage.

250 Figure 4 shows the NIR spectra of all samples from 833 to 2200 nm. The range from 2200 to 2500 nm
251 was removed due to instrumental noise. Two spectral pre-treatments were reported, SNV (Figure 3a)
252 and first derivative (Figure 3b). Considering that the penetration depth of NIR radiation in food is
253 approximately 1 mm (Almeida et al., 2006; Huang et al., 2016), the spectra are mainly affected by
254 the dark chocolate (thickness of 0.6 mm). However, there may also be an influence on the wafer sheet
255 composition. Cocoa butter is the main ingredient in chocolate, although it also contains sugar and
256 cocoa mass. These ingredients contribute to the spectral signals because of the presence of peptide

257 and covalent bonds, proteins, carbohydrates and lipids (Gatti et al., 2021; Giovenzana et al., 2015;
258 Moros et al., 2007). For all samples, it is possible to identify peaks to around 1730, and 1760 nm
259 which match the C-H bonds in CH₂ groups (cocoa butter). Fatty acids and esters can be associated
260 with absorption bands at 1180, 1215, 2145 and 2190 nm, and amide II and amide III at 2010 and 2050
261 nm, respectively (small amount of protein). Moreover, spectra show peaks at 1440 nm and 2080 nm
262 corresponding to O-H absorbance in sugar. The small amount of water, mainly present in the wafer
263 sheet, could be identified at around 1930 nm. Consequently, according to the increases in water
264 activity, especially from 0 to 2 months of storage, notable differences were found in the spectral range
265 from 1900 to 2000 nm between the samples analysed at time 0 (black lines) and during storage (grey
266 lines).

267 PCA analysis was used as an explorative technique to group samples as a function of packaging type
268 and storage time. The score plot obtained considering all samples in the spectral range from 1000 to
269 2200 nm is shown in Figure 4a. A clear separation between samples acquired at time 0 (black squares)
270 and those analysed during storage (grey symbols) was mainly observed along PC2 (12.25 %).
271 Samples stored in standard (S) and vacuum (V) packaging can be grouped considering the PC1 and
272 PC2. Evaluating the X-loading plot (Figure 4b), it was possible to assert that the discrimination along
273 the PC2 is due to the NIR region from 1850 to 2000 due to the significant increase in water content
274 from 0 to 2 months of storage.

275 Considering only the samples analysed during storage (from 2 to 8 months), three additional PCA
276 were developed for both (Figure 5a) and single (Figure 5b and 5c) packaging types. The score plots
277 showed notable sample distributions along PC1 according to storage time, passing from 2 to 8 months.
278 This behaviour was more pronounced for samples stored in standard packaging, which is probably
279 due to the greater increase in water activity. Also, in this case the highest X variable variances
280 associated with the highest X-loadings were observed in the range from 1800 to 2000 nm (water
281 band). Changes in the spectral ranges characteristic of fats were not observed, thus confirming that
282 there was no fat bloom formation in chocolates (Gatti et al., 2021) as deduced by the colour analysis.

283 PLS models were developed to estimate the storage time and water activity from NIR data considering
284 the spectral range from 1000 to 2200 nm. The results, in terms of determination coefficient (R^2) and
285 root mean square error (RMSE), are reported in Table 4. The model's robustness, according to the
286 number of selected latent variables (LVs), was evaluated with a permutation test. *P*-values lower than
287 0.05 were obtained for cross-validation by employing a randomization t-test, thus confirming the
288 significance of the original models at a 95% confidence level.

289 . R^2 (cross-validation) from 0.926 (RMSECV=0.63 months) to 0.960 (RMSECV=0.52 months) and
290 from 0.858 (RMSECV=0.02) to 0.924 (RMSECV=0.02) were achieved for storage time and water
291 activity, respectively. In general, the best performances were obtained for storage time and
292 considering separately the samples stored in standard and vacuum packaging.

293 Figure 6 shows the mean Vis/NIR spectra (calculate considering the ROI of the hyperspectral images)
294 of all samples in the wavelength range from 450 to 1000 nm. Two spectral pre-treatments were
295 reported, namely SNV (Figure 7a) and D1 (Figure 7b). The absorbance spectra are similar to those
296 reported by (Fernandes et al., 2018; Millar and Hall, 2005) for dark chocolate and cocoa samples,
297 respectively. As expected, the highest absorbances were observed in the VIS range, until 700 nm, due
298 to the sample's dark colour. The first derivative allowed to emphasise the presence of peaks and
299 highlight differences between the spectra.

300 The PCA score plot obtained considering all samples in the spectral range from 450 to 1000 nm is
301 shown in Figure 7a. A clear separation between samples acquired at time 0 (black squares) and those
302 analysed during storage (grey symbols) was observed along PC1 (56.59 %) and PC2 (23.92 %). The
303 X-loading plot (Figure 7b) suggests that the discrimination might be attributed to the Vis absorption
304 region, especially around 500 and 650 nm which is associated with blue and red colours, respectively.
305 These two bands were also proposed by (Fernandes et al., 2018) as absorption maxima of cocoa beans
306 in the Vis/NIR range. Sample discrimination was not observed according to the packaging type and
307 storage time. Subsequently, only the NIR range (700-1000 nm) and samples analysed during the
308 storage (from 2 to 8 months) were considered. The PCA score plot showed sample distributions along

309 PC2 according to the storage time, passing from 2 to 8 months, for both packaging types (Figure 8).
310 As for the NIR spectroscopy results, the discrimination was more pronounced for samples stored in
311 standard packaging.
312 As for NIR data, PLS models were developed to estimate the storage time and water activity.
313 Considering the PCA results, only the spectral range from 7000 to 1000 nm was taken into account.
314 The results, in terms of R^2 , RMSE and number of LV (model robustness tested by permutation test
315 with $p < 0.05$) are reported in Table 4. R^2 (cross-validation) from 0.931 (RMSECV=0.62 months) to
316 0.956 (RMSECV=0.55 months) and from 0.869 (RMSECV=0.02) to 0.928 (RMSECV=0.02) were
317 obtained for storage time and water activity, respectively. The best performances were obtained for
318 the storage time considering separately the samples stored in standard and vacuum packaging. Very
319 similar PLS results were achieved for both spectroscopic techniques.
320 The calibration PLS models were used to obtain predictive spectral images based on their water
321 activity or storage time. In particular, the water activity or storage time spatial distribution was
322 achieved by interpolating the results of the estimated value from the models in each pixel of the
323 hyperspectral image. The prediction maps of three representative samples (samples stored in standard
324 packaging) are shown in Figure 9. The colour bar (from blue to yellow) indicates the scale of the
325 reference values (water activity from 0.2 to 0.5; storage time from -1 to 10 months). For both
326 references values, the spatial distributions are in alignment with the measured values. The mean
327 predicted water activity was 0.27 (A), 0.40 (B) and 0.46 (C), while for the storage time was 0.2 (A),
328 3.9 (B) and 7.6 (C) months. The non-homogeneous colour distribution suggests that there was a
329 spectral variation as a function of the pixel position. This is probably due to the product surface
330 heterogeneity related to the wafer texture, and a possible non-uniform distribution of the chocolate
331 layer, affecting the physical and chemical properties of each pixel (Chen et al., 2021).

332

333 **Conclusions**

334 The storage of wafer cookies packaged with two different packaging was assessed using destructive
335 (water activity, mechanical properties, and sensory acceptance) and non-destructive methods (image
336 analysis, NIR spectroscopy and hyperspectral imaging). Good linear relations were found between
337 water activity and mechanical parameters, confirming that changes in wafer moisture can directly
338 compromise the product's textural parameters. A correspondence between sensory acceptance limit
339 (6) and the water activity value of 0.44 was found. Other studies defined this water activity value as
340 the limit to describe "good" and "compromised" wafers, in terms of sensory (textural) properties. The
341 packaging characterised by the addition of a multi-material layer significantly affected the product's
342 shelf-life in terms of sensory acceptance. The surface colour (white index) was not changed,
343 confirming there was no evident fat bloom formation during storage due to improper temperature or
344 temperature fluctuations. NIR spectroscopy and hyperspectral imaging allowed to determine the
345 storage time and water activity in real-time and in a non-destructive manner, and to detect differences
346 according to the type of packaging. Furthermore, hyperspectral imaging permitted visualization of
347 the spatial distribution of the water activity and storage time.

348

349 **References**

- 350 Aguilera, M., Briones, V., 2005. Image analysis of changes in surface color of chocolate. *Food Res.*
351 *Int.* 38, 87–94. <https://doi.org/10.1016/j.foodres.2004.09.002>
- 352 Almeida, M., Torrance, K.E., Datta, A.K., Almeida, M., 2006. Measurement of Optical Properties of
353 Foods in Near- and Mid-Infrared Radiation. *Int. J. Food Prop.* 29, 651–664.
354 <https://doi.org/10.1080/10942910600853667>
- 355 Amigo, J.M., Marti, I., Gowen, A., 2013. Hyperspectral Imaging and Chemometrics : A Perfect
356 Combination for the Analysis of Food Structure , Composition and Quality, in: Marini, F. (Ed.),
357 Data Handling in Science and Technology. Elsevier B.V., Amsterdam: The Netherlands, pp.
358 343–370. <https://doi.org/10.1016/B978-0-444-59528-7.00009-0>
- 359 Andresen, M.S., Dissing, B.S., Hanne, L., 2013. Quality assessment of butter cookies applying

360 multispectral imaging. *Food Sci. Nutr.* 1, 315–323. <https://doi.org/10.1002/fsn3.46>

361 Caporaso, N., Whitworth, M.B., Fowler, M.S., Fisk, I.D., 2018. Hyperspectral imaging for non-
362 destructive prediction of fermentation index , polyphenol content and antioxidant activity in
363 single cocoa beans. *Food Chem.* 258, 343–351. <https://doi.org/10.1016/j.foodchem.2018.03.039>

364 Cevoli, C., Gianotti, A., Troncoso, R., Fabbri, A., 2015. Quality evaluation by physical tests of a
365 traditional Italian flat bread. *Eur. Food Res. Technol.* 240, 1081–1089.
366 <https://doi.org/10.1007/s00217-015-2429-7>

367 Chakravartula, S.S., Cevoli, C., Balestra, F., Fabbri, A., Dalla, M., 2019. Evaluation of the effect of
368 edible coating on mini-buns during storage by using NIR spectroscopy. *J. Food Eng.* 263, 46–
369 52. <https://doi.org/10.1016/j.jfoodeng.2019.05.035>

370 Chen, Z., Wang, Q., Zhang, H., Nie, P., 2021. Hyperspectral Imaging (HSI) Technology for the
371 Non-Destructive Freshness Assessment of Pearl Gentian Grouper under Different Storage
372 Conditions. *Sensors* 21, 1–13. <https://doi.org/https://doi.org/10.3390/s21020583>

373 Cruz-tirado, J.P., Antonio, J., Barbin, F., Baeten, V., 2020. Authentication of cocoa (*Theobroma*
374 *cacao*) bean hybrids by NIR-hyperspectral imaging and chemometrics. *Food Control J.* 118,
375 107445. <https://doi.org/10.1016/j.foodcont.2020.107445>

376 Dogan, I.S., 2006. Original article Factors affecting wafer sheet quality. *Int. J. Food Sci. Technol.* 41,
377 569–576. <https://doi.org/10.1111/j.1365-2621.2005.01117.x>

378 Duizer, L.M., Winger, R.J., 2006. Instrumental measure of bite forces associated with crisp products.
379 *J. Texture Stud.* 37, 1–15.

380 Fernandes, D., Leonardo, B., Maciel, F., Henrique, C., Bazoni, V., Ribeiro, S., Duarte, R., Carvalho,
381 S., Bispo, S., Yoko, E., Spi, P., 2018. Classification and compositional characterization of
382 different varieties of cocoa beans by near infrared spectroscopy and multivariate statistical
383 analyses 55, 2457–2466. <https://doi.org/10.1007/s13197-018-3163-5>

384 Gatti, R.F., Santana, F.B. De, Poppi, R.J., Ferreira, D.S., 2021. Portable NIR spectrometer for quick
385 identification of fat bloom in chocolates. *Food Chem.* 342, 128267.

386 <https://doi.org/10.1016/j.foodchem.2020.128267>

387 Giovenzana, V., Beghi, R., Hassinger, A.P., Guidetti, R., 2015. Assessment of Tempering Degree
388 during the Chocolate Pre-Crystallisation Phase Using near Infrared Spectroscopy. *NIR news* 26,
389 8–10. <https://doi.org/https://doi.org/10.1255/nirn.1501>

390 Huang, M., Kim, M.S., Chao, K., Qin, J., Mo, C., Esquerre, C., Delwiche, S., Zhu, Q., 2016.
391 Penetration Depth Measurement of Near-Infrared Hyperspectral Imaging Light for Milk
392 Powder. *Sensors* 16, 1–11. <https://doi.org/10.3390/s16040441>

393 Kumara, B., Jinap, S., Man, Y.B.C., Yusoff, M.S.A., 2003. Note : Comparison of Colour Techniques
394 to Measure Chocolate Fat Bloom. *Food Sci. technology Int.* 9, 295–299.
395 <https://doi.org/10.1177/108201303036045>

396 Lancelot, E., Courcoux, P., Chevallier, S., Le-Bail, A., Jaillais, B., 2020. Prediction of water content
397 in biscuits using near infrared hyperspectral imaging spectroscopy and chemometrics. *J. Near*
398 *Infrared Spectrosc.* 28, 140–147. <https://doi.org/10.1177/0967033520902538>

399 Liu, D., Zeng, X.A., Sun, D.W., 2015. Recent Developments and Applications of Hyperspectral
400 Imaging for Quality Evaluation of Agricultural Products: A Review. *Crit. Rev. Food Sci. Nutr.*
401 55, 1744–1757. <https://doi.org/10.1080/10408398.2013.777020>

402 Martinez-Navarrete, N., Moraga, G., Talens, P., Chiralt, A., 2004. Water sorption and the
403 plasticization effect in wafers. *Int. J. Food Sci. Technol.* 39, 555–562.
404 <https://doi.org/10.1111/j.1365-2621.2004.00815.x>

405 Millar, S., Hall, A., 2005. Evaluating chocolate blends [WWW Document]. *NEW Food*. URL
406 <https://www.newfoodmagazine.com/article/2320/evaluating-chocolate-blends/>

407 Mohammed, I.K., Charalambides, M.N., Williams, J.G., Rasburn, J., 2014. Modelling the
408 microstructural evolution and fracture of a brittle confectionery wafer in compression. *Innov.*
409 *Food Sci. Emerg. Technol.* 24, 48–60. <https://doi.org/10.1016/j.ifset.2013.11.015>

410 Mohammed, I.K., Charalambides, M.N., Williams, J.G., Rasburn, J., 2013. Modelling the
411 deformation of a confectionery wafer as a non-uniform sandwich structure 2462–2478.

412 <https://doi.org/10.1007/s10853-012-7034-6>

413 Moros, J., Inos, F., Garrigues, S., Guardia, M. De, 2007. Near-infrared diffuse reflectance
414 spectroscopy and neural networks for measuring nutritional parameters in chocolate samples.
415 *Anal. Chim. Acta* 584, 215–222. <https://doi.org/10.1016/j.aca.2006.11.020>

416 Nasabi, M., Naderi, B., Akbari, M., Aktar, T., Kieliszek, M., 2021. Physical , structural and sensory
417 properties of wafer batter and wafer sheets influenced by various sources of grains. *LWT - Food*
418 *Sci. Technol.* 149, 111826. <https://doi.org/10.1016/j.lwt.2021.111826>

419 Popov-Raljić, J. V., Laličić-Petronijević, J.G., 2009. Sensory Properties and Color Measurements of
420 Dietary Chocolates with Different Compositions During Storage for Up to 360 Days. *Sensors* 9,
421 1996–2016. <https://doi.org/10.3390/s90301996>

422 Sørensen, L.K., 2009. Application of reflectance near infrared spectroscopy for bread analyses. *Food*
423 *Chem.* 113, 1318–1322. <https://doi.org/10.1016/j.foodchem.2008.08.065>

424 Sricharoonratana, M., Keith, A., Teerachaichayut, S., 2021. Use of near infrared hyperspectral
425 imaging as a nondestructive method of determining and classifying shelf life of cakes. *LWT -*
426 *Food Sci. Technol.* 136, 110369. <https://doi.org/10.1016/j.lwt.2020.110369>

427 Tiefenbacher, K.F., 2017. *The Technology of Wafers and Waffles I Operational Aspects*, 1st Editio.
428 ed. Academic Press Elsevier.

429 Whitworth, M.B., Millar, S., Chau, A., 2010. Food quality assessment by NIR hyperspectral imaging,
430 in: *SPIE Defense, Security, and Sensing*, Orlando, Florida (USA). pp. 1–12.

431 Xie, F., Floyd, E., Xiuzhi, S., 2003. Comparison of near-infrared reflectance spectroscopy and
432 texture ... *Cereal Chem.* 80, 25–29.

433 Zhang, X., de Juan, A., Tauler, R., 2015. Multivariate Curve Resolution Applied to Hyperspectral
434 Imaging Analysis of Chocolate Samples. *Appl. Spectrosc.* 69, 993–1003.
435 <https://doi.org/10.1366/14-07819>

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438 **Figure captions**

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440 **Figure 1.** Selection of the region of interest (ROI) by k-means clustering (cluster 1: background;
441 cluster 2: sample shadow; cluster 3: ROI)

442 **Figure 2.** Sensory acceptance as a function of storage time (a), water activity (b) and distance (D)
443 mechanical parameter (c). S: standard packaging, V: vacuum packaging.

444 **Figure 3.** NIR spectra: a) Standard Normal Variate (SNV) pre-processing; b) first derivative (D1)
445 pre-processing (S: standard packaging, V: vacuum packaging; t0: initial storage time).

446 **Figure 4.** Results of the PCA model developed considering all NIR spectra (from 1000 to 2200 nm,
447 SNV+D1+MC pre-processing): a) score plot (PC1 vs PC2), b) X-loadings of the first two component.
448 (S: standard packaging, V: vacuum packaging; t0: initial storage time).

449 **Figure 5:** Score plots (sample vs PC1) of the PCA models developed considering only the NIR spectra
450 (from 1000 to 2200 nm, SNV+D1+MC pre-processing) of samples analysed during storage (from 2
451 to 8 months): a) samples stored in standard and vacuum packaging; b) samples stored in standard
452 packaging; c) samples stored in vacuum packaging

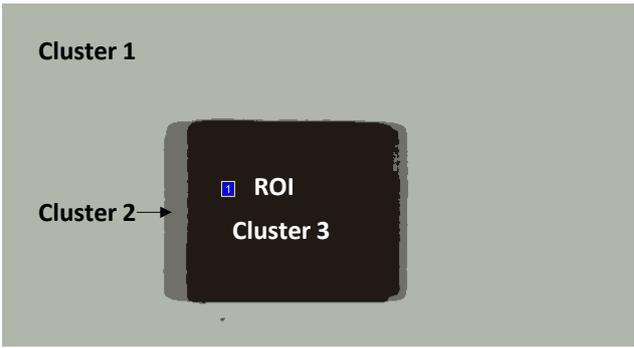
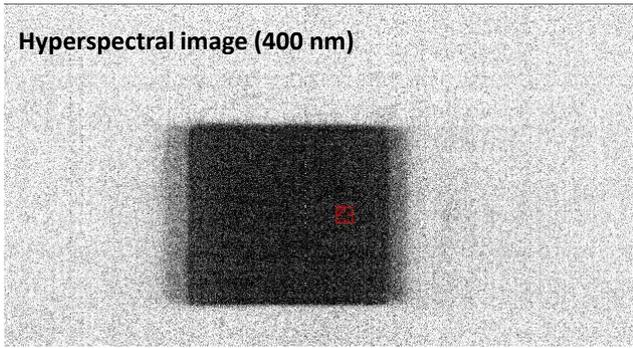
453 **Figure 6:** Vis/NIR HSI spectra: a) Standard Normal Variate (SNV) pre-processing; b) first derivative
454 (D1) pre-processing (S: standard packaging, V: vacuum packaging.).

455 **Figure 7.** Results of the PCA models developed considering all Vis/NIR HSI spectra (from 500 to
456 1000 nm; SNV+D1+MC pre-processing): a) score plot (PC1 vs PC2), b) X-loadings of the first two
457 component. (S: standard packaging, V: vacuum packaging; t0: initial storage time).

458 **Figure 8.** Score plot (PC2) of the PCA model developed considering only the vis/NIR HSI spectra
459 (from 700 to 1000 nm, SNV+D1+MC pre-processing) of samples analysed during storage (from 2 to
460 8 months).

461 **Figure 9.** Prediction maps of water activity and storage time of three representative wafer cookies
462 stored in standard packaging (A=0 months, B=4 months; C=8 months).

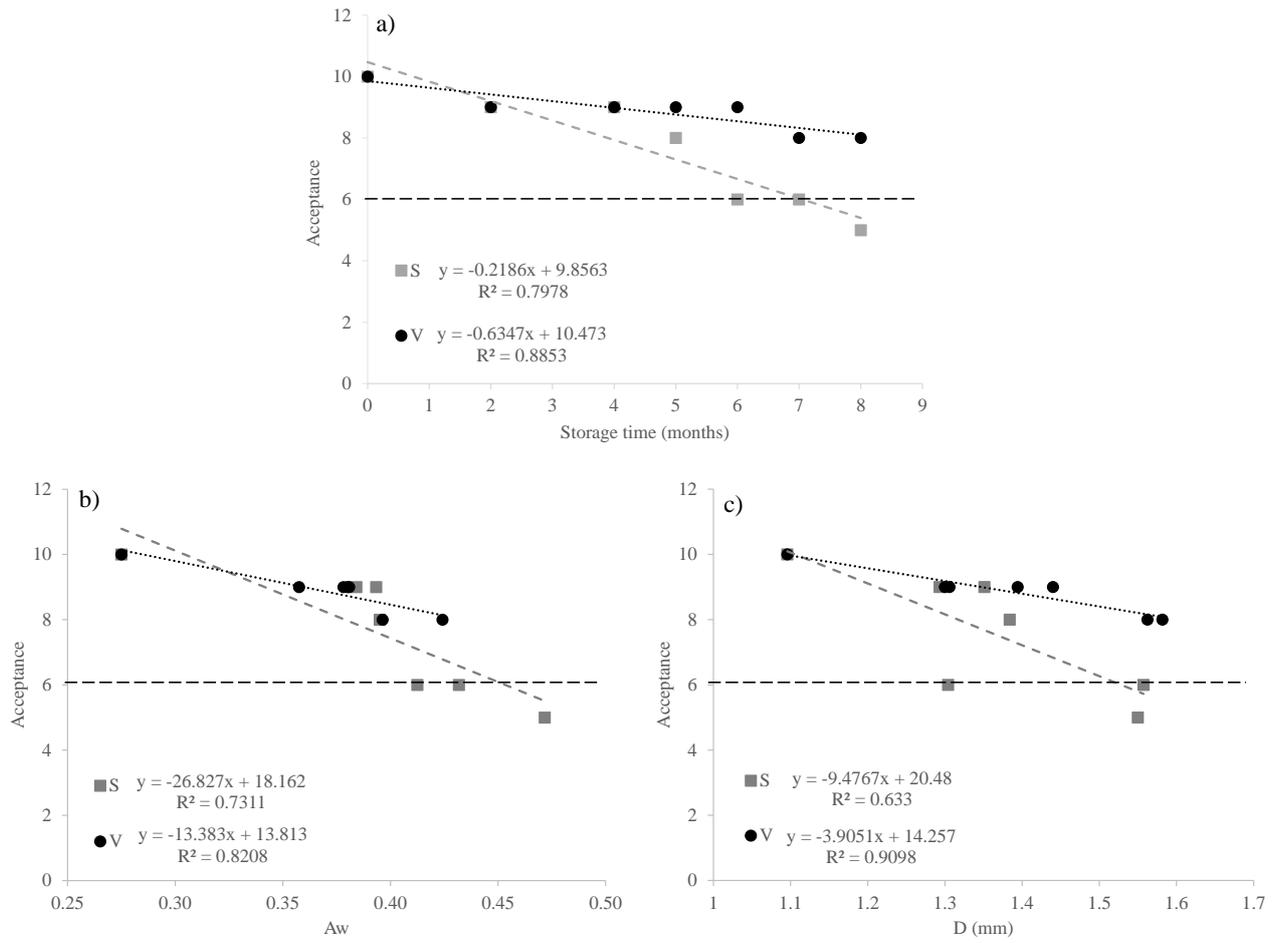
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465 Fig.1

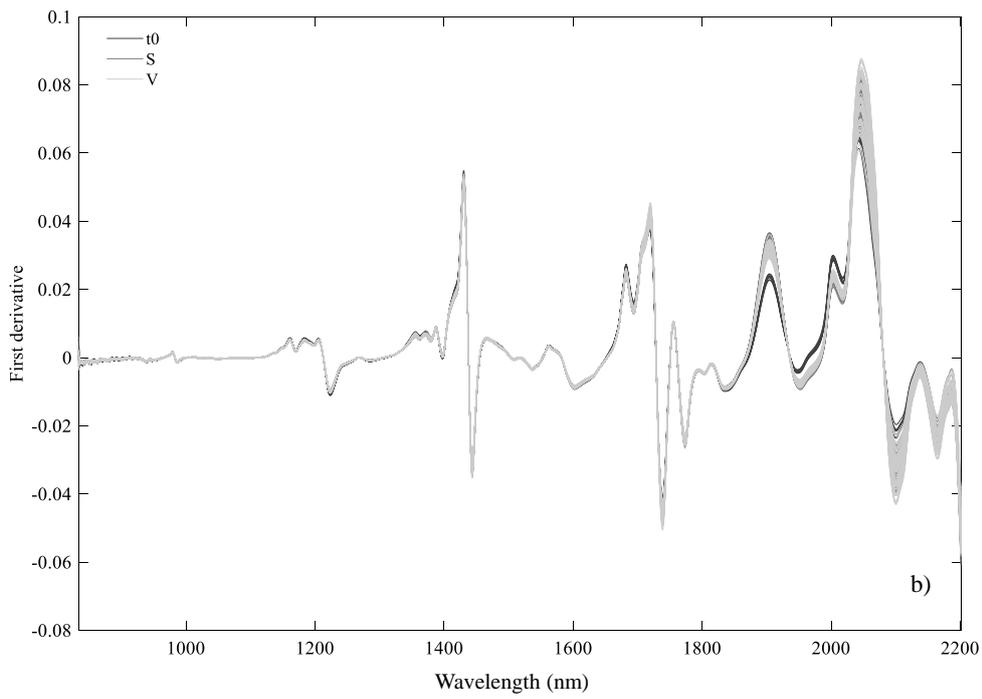
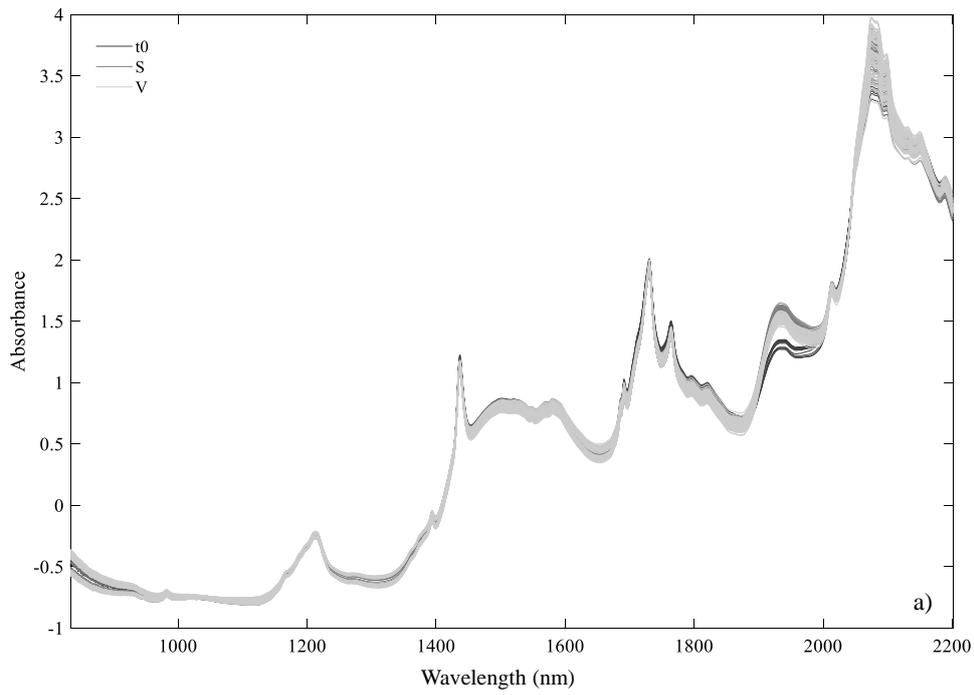
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468 Fig.2

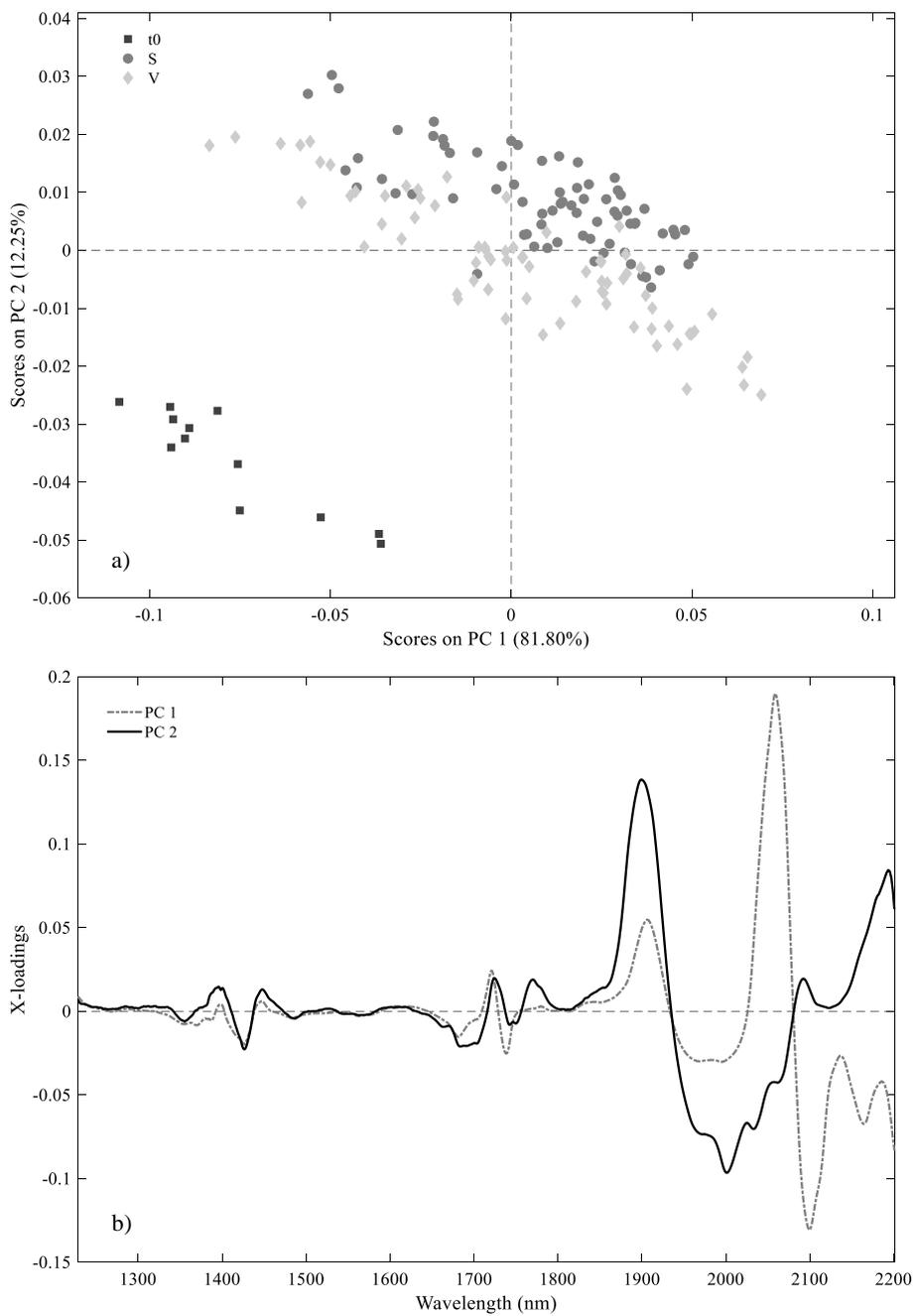
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471 Fig.3

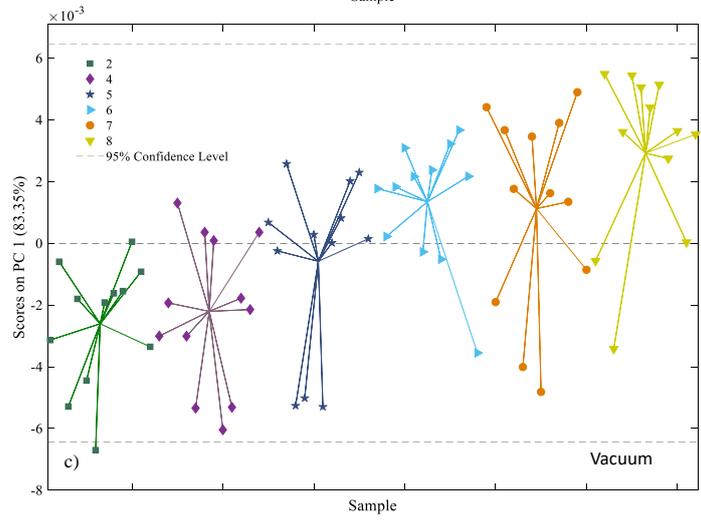
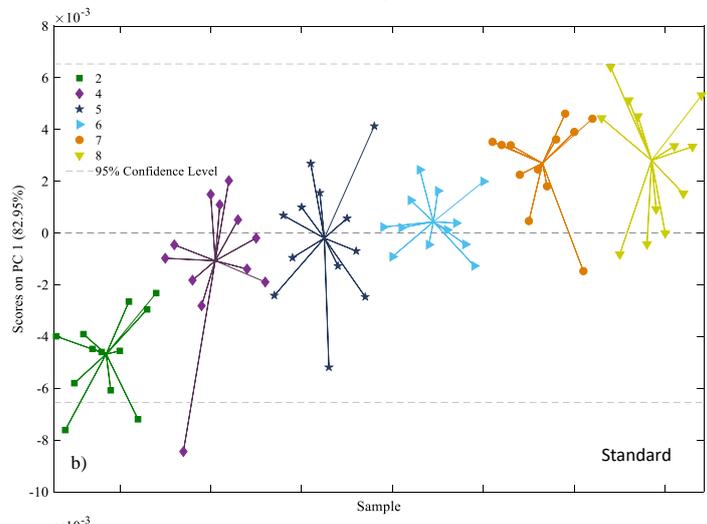
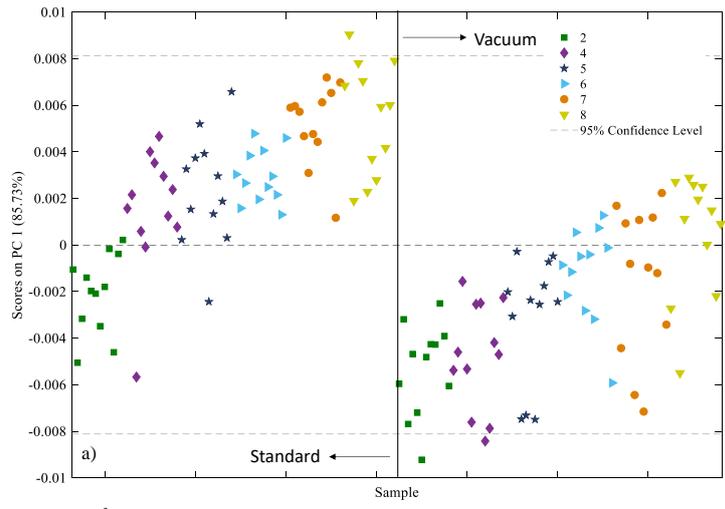
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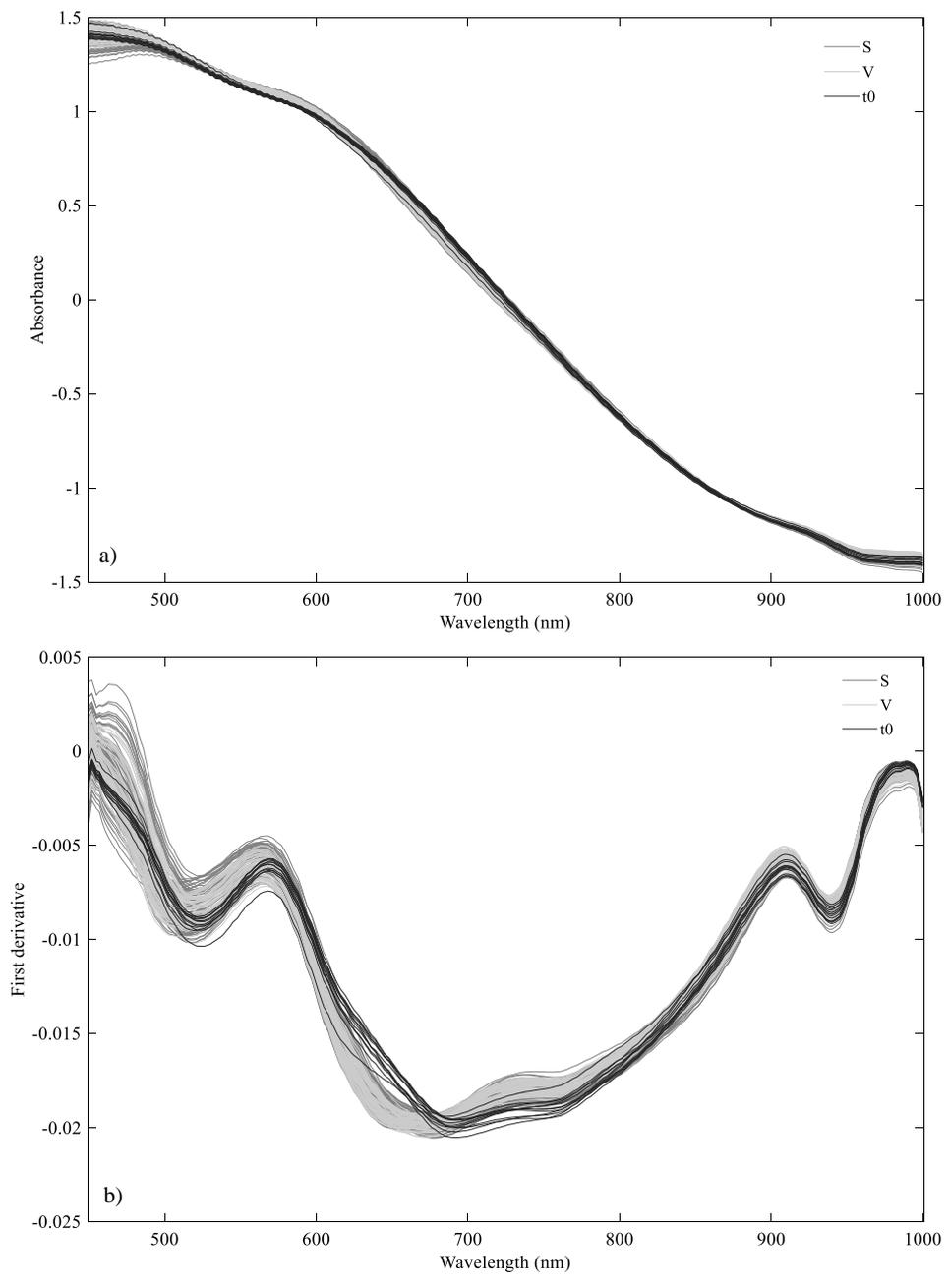
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474 Fig.4

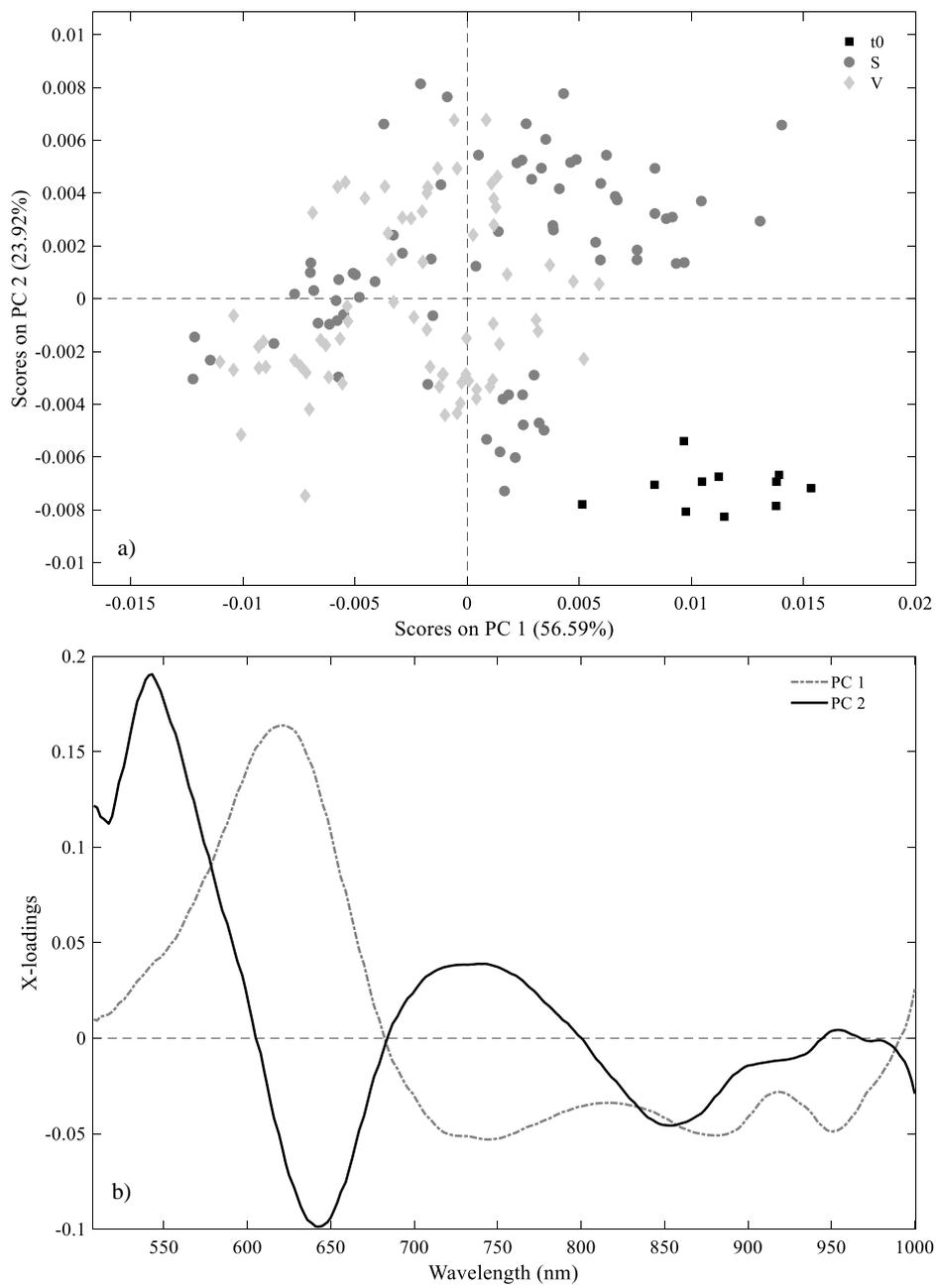
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 477 Fig.5
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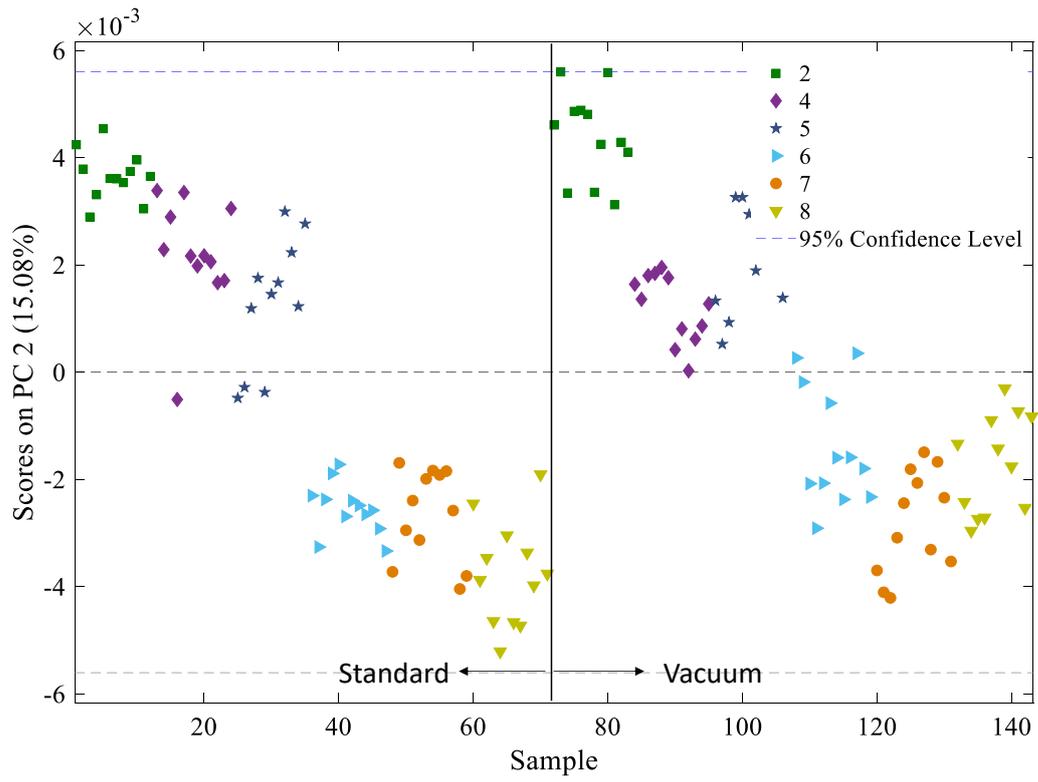
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480 Fig.6
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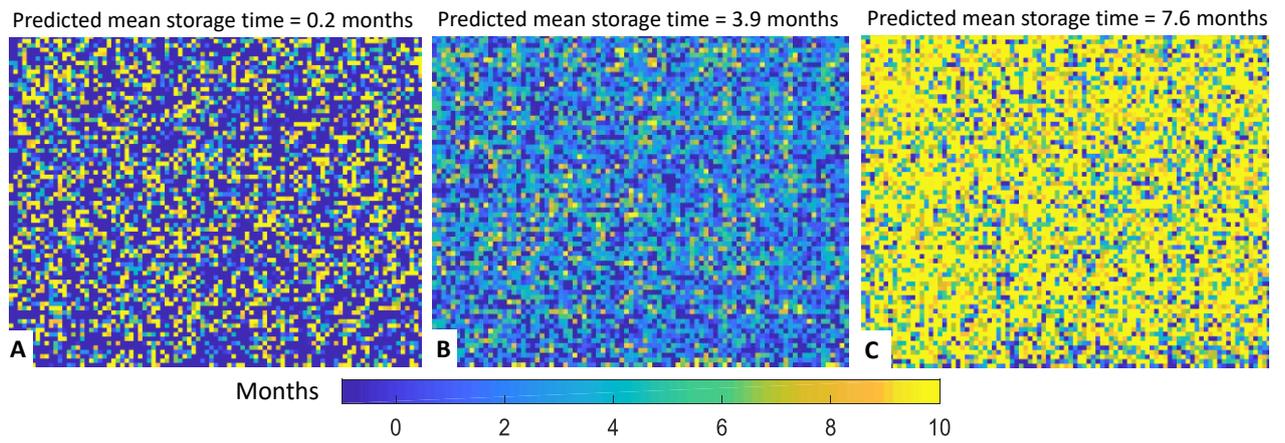
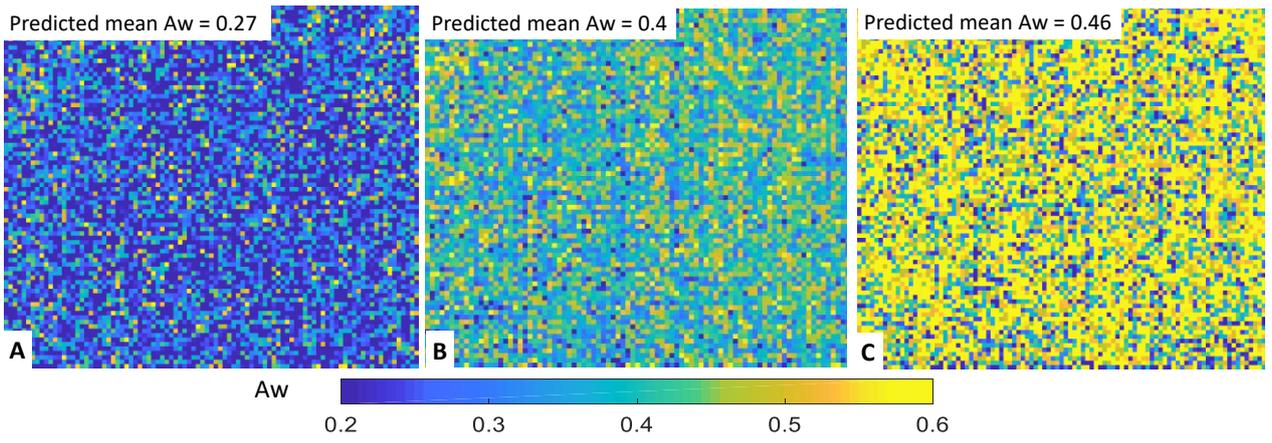
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486 Fig.8

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489 Fig.9

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491 **Table 1:** Wafer cookie ingredients

	
Wafer sheet (15.6%)	Wheat flour, potato starch, coconut oil, skimmed milk powder, sugar, soy whole flour, emulsifier: soy lecithin; egg yolk powder, raising agents: sodium carbonates, diphosphates; salt.
Vanilla cream (58.1%)	Coconut oil, sugar, dextrose, Skimmed milk powder, soy whole flour, salt, flavourings, Bourbon vanilla pod extract.
Chocolate enrobing (26.3%)	Cocoa paste, sugar, cocoa butter, emulsifier: soy lecithin; vanilla extract.

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494 **Table 2:** Packaging materials

Packaging I	Packaging II	Packaging III
		
<p>1) Multimaterial: wax paper (5 μm) and aluminum (10 μm) 2) Paper</p>	<p>3) Calendered PVC film (300 μm) 4) Paper box (weight 400 gsm) 5) POF shrink film (19 μm)</p>	<p>6) Multimaterial: PET (12 μm), aluminium (8 μm) and PE (110 μm).</p>

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Table 3: Means and standard deviations (in brackets) of water activity (A_w), mechanical parameters (Fmax and D) and white index (WI) evaluated during the storage.

Packaging	Storage time (months)	A_w	F max (N)	D (N/mm)	WI
S	0	0.27 ^a (0.01)	50.63 ^a (8.45)	1.09 ^a (0.12)	32.06 ^a (0.59)
	2	0.38 ^b (0.01)	68.86 ^b (7.89)	1.29 ^b (0.13)	32.55 ^a (0.45)
	4	0.39 ^b (0.01)	68.19 ^b (12.18)	1.35 ^b (0.15)	32.45 ^a (0.34)
	5	0.39 ^b (0.01)	59.60 ^{a,b} (8.71)	1.38 ^b (0.12)	32.82 ^a (0.44)
	6	0.41 ^c (0.01)	65.15 ^{a,b} (9.46)	1.30 ^b (0.15)	32.18 ^a (0.29)
	7	0.43 ^d (0.01)	67.26 ^b (9.27)	1.56 ^c (0.15)	31.96 ^a (0.53)
	8	0.47 ^e (0.01)	57.16 ^a (7.16)	1.55 ^c (0.14)	32.38 ^a (0.46)
	V	0	0.27 ^a (0.01)	50.63 ^a (8.45)	1.09 ^a (0.12)
2		0.36 ^b (0.01)	68.05 ^b (6.23)	1.39 ^b (0.11)	32.24 ^a (0.46)
4		0.38 ^b (0.01)	59.12 ^{a,b} (10.29)	1.42 ^b (0.15)	32.79 ^a (0.35)
5		0.38 ^b (0.01)	61.74 ^{a,b} (9.85)	1.30 ^b (0.11)	32.65 ^a (0.45)
6		0.38 ^b (0.01)	71.35 ^b (9.40)	1.31 ^b (0.12)	32.42 ^a (0.32)
7		0.40 ^c (0.01)	69.33 ^b (8.12)	1.56 ^c (0.13)	32.63 ^a (0.37)
8		0.42 ^d (0.01)	51.10 ^a (7.39)	1.58 ^c (0.12)	32.01 ^a (0.45)

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Note: S: standard packaging; V: vacuum packaging. Within the same type of packaging (S or V), means with the same letters are not significant different ($p < 0.05$)

503 **Table 4:** Results of the PLS models developed to estimate the storage time and water activity from
 504 NIR spectroscopy and Vis/NIR HIS data.

Parameter	Samples	LV	Calibration		Cross-validation		
			R ²	RMSEC	R ²	RMSECV	
NIR spectroscopy	Storage time (0-8 months)	S	10	0.989	0.26	0.958	0.53
		V	11	0.987	0.29	0.960	0.52
		S+V	12	0.956	0.49	0.926	0.63
	Water activity	S (0.27-0.47)	7	0.951	0.01	0.924	0.02
		V (0.27-0.42)	7	0.922	0.01	0.878	0.02
		S+V (0.27-0.47)	7	0.887	0.02	0.858	0.02
Vis/NIR HSI	Storage time (0-8 months)	S	10	0.957	0.54	0.955	0.56
		V	11	0.956	0.42	0.956	0.55
		S+V	12	0.931	0.57	0.931	0.62
	Water activity	S (0.27-0.47)	4	0.936	0.01	0.928	0.02
		V (0.27-0.42)	7	0.905	0.01	0.876	0.02
		S+V (0.27-0.47)	8	0.884	0.02	0.869	0.02

505 *Note: S: standard packaging; V: vacuum packaging; LV: latent variable; R²: determination coefficient; RMSE: root*
 506 *mean square error.*

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