Quality by design for packaging of granola breakfast product

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ABSTRACT
Quality by Design (QbD) considers both the critical product characteristics and the environmental variables to design an optimum packaging system. This study applied the QbD approach for packaging and shelf-life determination of Granola by i) determining the water vapour transmission rate (WVTR) of packaging films at different environmental conditions, ii) develop and validate a shelf-life model of packed granola breakfast product and iii) predict shelf-life of packed Granola. The WVTR of packaging films (BOPP and biodegradable films, i.e., NK, NM, N913) was measured according to a full factorial experimental design (3^3), i.e., 10, 30, 40 °C; 32.5 ± 0.5, 75.5 ± 0.5, 92.5 ± 3.5% RH, and a mathematical model was developed. Granola breakfast product was packed (using the mentioned materials and also a commercial packaging film-control), stored under accelerated conditions (38 °C and 90% RH) and assessed for moisture content (critical quality parameter). A shelf-life model was developed and validated for Granola describing the relationship of the food, packaging and environmental conditions, and shelf-life was inferred for normal storage conditions. The developed WVTR global model considering the dependency of temperature and relative humidity was found to fit the experimental data well (R^2 > 0.914). Granola moisture gain was the lowest in BOPP package followed by biodegradable N913 package. The shelf-life for Granola under accelerated conditions ranged from 2 to 13 days depending on the packaging film, and under normal storage conditions (20 °C and 75% RH) was 271, 269, 90, and 33 days for BOPP, N913, NK and NM packages, respectively.

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1. Introduction

Granola is a dry granulated cereal product which has a low water activity. Granola’s shelf-life is limited by chemical and physical changes and the rate of deteriorative reactions depends on its composition as well as environmental factors. Moisture content was identified as the critical quality parameter and relative humidity as the most influential environmental factor (Macedo, Sousa-Gallagher, & Byrne, 2009). Quantitative determination of quality decay of Granola is important for estimating the shelf-life and designing new processes and packaging (Galic, Scetar, & Kurek, 2011; Macedo, Sousa-Gallagher, Oliveira, Mahajan, & Byrne, 2011).

Strategies often employed to prevent deterioration include control of temperature and water activity, addition of antioxidants, removal of oxygen or modification of headspace gas composition and its retention during distribution and storage, or a combination of these with effective packaging (Kong & Singh, 2011, chap. 2). Packaging represents an essential step of the process, protecting, extending food shelf-life and minimising food waste, while adding convenience and providing information to consumers. During the distribution chain, Granola can be exposed to a range of quite different environmental conditions, and if there is a gradient between water activity inside and outside the package, this driving force causes a transfer of water molecules through the package leading to an increase of internal water activity, therefore causing an increase of moisture content and consequently a loss of Granola quality.

Environmental concerns have triggered the use of biobased packaging materials as an alternative to materials produced from non-renewable resources. Cereal products are usually packaged in paper/polyethylene packaging (Galic, Curic, & Gabric, 2009), but there is an interest in replacing its packaging and potentially use different and more environmentally friendly packaging materials. NatureFlex manufactures a transparent cellulose base material made from sustainable wood pulp, plus specially formulated biodegradable and compostable surface layers, which control the moisture permeability (http://www.innoviafilms.com/).

Moisture transfer in the packaged food depends on the water activity of the food, environmental temperature and humidity conditions of storage, and the permeability of the package to water vapour. Therefore, shelf-life determination is highly dependent on the permeability characteristics of the packaging materials, which emphasizes the importance of packaging design. An optimum
package design should balance the packaging material properties, product protection requirements, environmental and transport conditions, and cost. Two phenomena should be considered to describe the moisture uptake by a packaged, sensitive food product, (1) the transfer of water vapour through the package, and (2) the kinetic uptake of water by the food product. The shelf-life of a moisture sensitive food product can be estimated using mathematical models that describe and connect the equilibrium sorption isotherm of the product, the initial and the permissible final moisture content, the permeance properties of the package and also the environmental relative humidity and temperature. An experimental evaluation of the optimal packaging design is often avoided due to the restricted time, and predictions using mathematical modelling of quality deterioration (i.e., critical attribute) as a function of factors in the food chain are commonly conducted (Macedo, Sousa-Gallagher, Mahajan, & Byrne, 2011).

There are many references to modelling chemical and physical deterioration and food shelf-life (e.g., Sousa-Gallagher, Mahajan, & Yan, 2011, chap. 2). The most widely studied critical quality attribute is vitamin C in various systems (Feng, Zhana, Qiaob, Wub, & Xiaob, 2012). Shelf-life prediction based on lipid oxidation was good agreements between predicted and experimental evaluation of the optimal packaging design is often avoided due to the restricted time, and predictions using mathematical models that describe and connect the equilibrium sorption isotherm of the product, the initial and the permissible final moisture content, the permeance properties of the package and also the environmental relative humidity and temperature. An experimental evaluation of the optimal packaging design is often avoided due to the restricted time, and predictions using mathematical modelling of quality deterioration (i.e., critical attribute) as a function of factors in the food chain are commonly conducted (Macedo, Sousa-Gallagher, Mahajan, & Byrne, 2011).

The kinetics of food decay are necessary to define a measurable index of deterioration, and analysis is performed by following the variation of each quality index over time during storage, and then comparing the measured value to a threshold. Macedo et al. (2009) found that moisture content MC and firmness could be taken as the indicator of shelf-life and that there is a relative high negative correlation between MC and firmness (−0.76), therefore MC was the only factor to be assessed. The Granola critical moisture quality threshold was found to be 8.9% (d.b.), for Granola initial MC of 6.5% (d.b.), defined from the correlation between moisture content and the sensory acceptability in terms of texture, for the environmental conditions studied.

The sorption isotherms of dried food products are crucial to model moisture gain during storage and distribution. Macedo, Sousa-Gallagher, and Byrne (2011) determined the Granola isotherms in a range of temperatures found during storage, and an overall model which accounts for the temperature effect on moisture content was developed.

**2.2. Critical quality parameters**

The critical quality parameters were identified by Macedo et al. (2009) by exposing the granola breakfast product to different environmental conditions (temperature and relative humidity). Quality parameters such as moisture content, firmness, colour and particle size distribution were measured throughout storage time and sensory characteristics were also evaluated. Moisture content of granola was measured by the AOAC (1980) oven method at 104 °C for at least 8 h. The texture properties of granola were measured using a TA-XT2i texture analyser, and the firmness (N) was expressed as the maximum compression force, the highest peak of the plot force (N) versus distance (mm). The colour changes of granola were monitored by the colorimeter Chroma Meter CR-300, using CIELAB $L_{a}$, $a_{b}$, $b_{c}$ parameters. Sensory characteristics, such as appearance/colour, firmness/texture and overall evaluation/acceptability, were evaluated at different sampling times (0, 3, 8 and 15 day) by a consumer panel using a hedonic scale (1 dislike extremely and 9 like extremely).

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**2.3. Sorption isotherms**

The sorption isotherms of dried food products are crucial to model moisture gain during storage and distribution. Macedo, Sousa-Gallagher, and Byrne (2011) determined the Granola isotherms in a range of temperatures found during storage, and an overall model which accounts for the temperature effect on moisture content was developed.

**2.4. Assessment of water vapour transmission rate (WVTR) of packaging films**

The WVTR of the biodegradable packaging films, NatureFlex 30 NK (NK), NatureFlex 23 NM (NM), NatureFlex 55 N913 (N913) and Propafilm RGP 30 (BOP) was calculated for each film using a full factorial experimental design (3²) at different RH (32.5 ± 0.5, 75.5 ± 0.5, 92.5 ± 3.5%) and T (10, 30, 40 °C). The WVTR of each film was determined by the gravimetric “cup” method described in ASTM E-96-95 (1995). The test cups consisted of a cylindrical cup (6.4 cm bottom diameter, 8.4 cm depth and 7.4 cm top inner diameter) and a lid. In the lid, an opening of 6.6 cm in diameter was made in the centre. The exposed film surface was 43 cm². The cups were filled with a saturated salt solution to provide the equilibrium relative humidity. The different films were placed to cover the opening and secured beneath by the lids, and to guarantee airtight sealing silicon grease was applied around the edge of the cups and under the lids. The cups were kept in dry conditions at different

**2.1. Production of granola breakfast product**

Granola breakfast product was produced through wet granulation of cereals (e.g., Oat and Corn flakes, Puffed Rice, Malted buckwheat, Malted barley) and prebiotic ingredients (i.e., oat beta glucan and inulin) using honey as a binder, in high shear mixers, which involves a size enlargement process with regular shaped granules with a high degree of compaction (Hanley, Pathare, Bas, & Byrne, 2008) and further optimised according to Pathare (2010).

The aim of this work was to i) determine the water vapour transmission rate of different packaging materials (BOP, NK, NM and N913) at different environmental conditions, ii) develop and validate an integrative shelf-life model of packed granola breakfast product under accelerated storage conditions, and iii) predict the shelf-life of packed granola under normal storage conditions. An oral communication was presented at the proceedings of the Seventh International Conference on Predictive Modelling in Foods (Macedo, Sousa-Gallagher, Oliveira et al., 2011).
temperatures. To reach the dry conditions all the cups were placed above the desiccant anhydrous calcium chloride and enclosed in an airtight plastic container. Weight of each container was measured at regular intervals. The WVTR of the BOPP, NK, NM and N913 films was obtained for a set of 9 different conditions, measuring the water transferred through the film at 9 different times in duplicated experiments, totalling 162 individual data points for each film. The BOPP was reported by the manufacturer (Innovia Films Ltd) to have a water vapour transmission rate, at 38°C and 90% RH, of 5.0 g/m²·day; the N913 was reported to be 14 g/m²·day; for the NK the water vapour transmission rate reported was 14 g/m²·day and for the and NM film was 10 g/m²·day.

2.5. Packaging

Pouches (8.5 cm × 10 cm) were made from the different biodegradable packaging films (BOPP, NK, N913, NM). In addition, a film used in the packaging of a commercial brand (control) was also tested similarly to the other film pouches. Granola samples (30 g) were packed in each of the pouches and packages were closed by heat sealing. Care was taken to minimize the pouches headspace and to ensure that the pouches were leak proof.

2.6. Shelf-life assessment

Seven sample pouches of each film (control, BOPP, NK, N913, NM) were suspended from the top of a large airtight container, ensuring that pouches were not in contact with each other and that all were exposed to the same accelerated environmental conditions (38°C and 90% RH). One pouch was taken from each container at 7 or 14 days interval up to 82 storage days, and moisture content (MC) was assessed in triplicate.

2.7. Shelf-life model development

A shelf-life model was developed describing the relationship of the food, packaging and environmental conditions. The shelf-life of the Granola was assumed to be controlled by the WVTR. The package headspace was neglected once WVTR was assumed to be accounted totally by the gain in moisture content of Granola. Lee, Yam, and Piergianni (2008, pp. 1–41, 79–108, 141–176 and 479–585) reported that the measurement of WVTR is a very important parameter for shelf-life determination of moisture sensitive food products, where shelf-life depends largely on the rates of permeation, which may occur quickly or slowly depending on the barrier property of the package.

The different packaging materials selected were good moisture barriers, and the mass balance and permeation of the package system is described by Eq. (1), which states that the mass gain is equal to the mass transfer flow rate across the package that is proportional to the driving force.

\[
W_s \frac{dM}{dt} = PA(p_{\text{wout}} - p_{\text{win}})
\]  
(1)

where \(W_s\) is the product dry weight (g); \(M\) is the moisture content of Granola (gH₂O/g dry solids); \(t\) is the time (days); \(P\) is the permeance (g/m²·day Pa); \(A\) is the packaging surface area (m²); and \(p_{\text{wout}}\) and \(p_{\text{win}}\) (Pa) are water vapour pressures outside and inside the package.

From the definition of relative humidity (RH) and water activity (\(a_w\))

\[
p_{\text{wout}} = p_s \times RH
\]  
(2)

\[
p_{\text{win}} = p_s \times a_w
\]  
(3)

where, \(p_s\) is the water vapour pressure of saturation (Pa) at the temperature of the system \((T)\) (K), RH is the relative humidity of the environment and \(a_w\) the food water activity, assuming pseudo-steady state is the same as relative humidity of the headspace.

The \(p_s\) may be estimated by the following equation (Lee et al., 2008):

\[
p_s = 611.2\exp\left(\frac{53.479 - 6808}{T} - 5.09\ln T\right)
\]  
(4)

Combining the Granola moisture sorption model shown in Eq. (5) (Macedo, Sousa-Gallagher, & Byrne, 2011) with the mass balance and permeation of the packaging system (Eq. (1)) to establish the interaction between the moisture gain by the Granola and the internal environment, led to Eq. (6), and the shelf-life of Granola was then calculated by using finite differences to solve the differential equation discretely.

\[
M = a + b a_w
\]  
(5)

\[
W_s \frac{dM}{dt} = PA \left[ RH - \left( M - a \right) \right]^{1/c}
\]  
(6)

where \(W_s\) is product dry weight (28.17 g); \(A\) is pock area (0.017 m²); \(p_s\) (38°C) is 6556 Pa; permeance \((P)\) was determined based on the WVTR (38°C and 90% RH), sorption isotherms parameters \(a\), \(b\) and \(c\) were 4.055, 43.72 and 3.718 respectively (Macedo, Sousa-Gallagher, & Byrne, 2011); \(MC\) is the initial moisture content (0.0650 gH₂O/g dry solids) and \(MC_c\) is the critical moisture content (0.0890 gH₂O/g dry solids).

To estimate the shelf-life of Granola, the Eq. (6) was integrated numerically. The shelf-life of Granola was predicted at accelerated conditions (38°C and 90% RH) and normal storage conditions (20°C and 75% RH).

The validation of the shelf-life of granola breakfast product was performed using the different packaging materials (BOPP, NK, N913, NM) at 38°C and 90% RH, and the quality decay (MC) of packaged Granola was studied throughout time.

3. Results and discussion

3.1. Water vapour transmission rate

The water vapour transmission rate (WVTR) data was very well fitted by a global model which considers an Arrhenius-type dependency of WVTR on temperature, and an activation energy independent of RH, with a pre-exponential factor varying exponentially with RH (Eq. (7)):

\[
\text{WVTR} = \left[ \exp(e RH) \right] \exp\left[ - \frac{E_a}{R} \frac{1}{T} - \frac{1}{T_{\text{ref}}} \right]
\]  
(7)

As WVTR is the constant rate of transfer of water vapour across the film, the total water vapour transferred varied linearly with time. The full set of experimental data points were thus fitted jointly, providing the results shown in Table 1. The goodness of fit

<table>
<thead>
<tr>
<th>Film</th>
<th>(d)</th>
<th>(e)</th>
<th>(E_a) [kJ/mol]</th>
<th>(R^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOPP</td>
<td>0.29 ± 0.08</td>
<td>0.40 ± 0.13</td>
<td>73.5 ± 6.8</td>
<td>0.928</td>
</tr>
<tr>
<td>N913</td>
<td>0.088 ± 0.022</td>
<td>2.35 ± 0.18</td>
<td>76.0 ± 5.0</td>
<td>0.972</td>
</tr>
<tr>
<td>NK</td>
<td>0.17 ± 0.03</td>
<td>3.89 ± 0.16</td>
<td>41.1 ± 1.8</td>
<td>0.986</td>
</tr>
<tr>
<td>NM</td>
<td>1.03 ± 0.28</td>
<td>2.47 ± 0.26</td>
<td>41.1 ± 4.3</td>
<td>0.914</td>
</tr>
</tbody>
</table>
can be seen in Fig. 1. The visual impression of the impact of T and RH on the water vapour transmission rate is shown in Fig. 2.

At accelerated shelf-life testing conditions of 38 °C and 90% RH, the model predicts the WVTR values shown in Table 2. The error of these predictions needs to consider the joint confidence intervals and parameter co-linearity between the 3 different parameters. The limits of a joint confidence region are defined by the sum of squares of the model error being within the limit of statistical significance. These limits of uncertainty of the model predictions are also shown in Table 2.

The BOPP packaging film was the least influenced film by relative humidity (RH) (Fig. 2); the effect of RH was minor and essentially the increase of temperature was the only significant factor. The N913, NK and NM films showed that the effect of temperature was much more pronounced at higher relative humidity. At any given temperature, increasing the relative humidity had an effect of increasing the water vapour transmission rate that was higher for the NK and NM films, and was particularly relevant at higher temperatures. The water vapour transmission rate reported (Innovia Films Ltd) for BOPP and N913 was 5.0 g/m² day and 14 g/m² day respectively, which is slightly lower than found for BOPP and just right for N913. For the NK and NM films, the water vapour transmission rate reported (Innovia Films Ltd) was 14 g/m² day and 10 g/m² day, respectively, which are far lower than those determined in this study, probably because of the high influence of relative humidity.

The concept of permeability is normally associated with the quantitative evaluation of the barrier property of a packaging material (Valentas, Rotstein, & Singh, 1997). Packaging film permeability can be described as i) transmission rate that describes permeation per area basis, ii) permeance that describes permeation per area per pressure difference basis, and iii) permeability which describes permeation on per area per pressure difference per thickness basis. The WVTR values were converted into permeance \( P \), by dividing the WVTR by the driving force \( \Delta p \), expressed as \( g/(\text{day m}^2 \text{ Pa}) \) (Eq.(8)).

\[
P = \frac{\text{WVTR}}{\Delta p}
\]

where, \( \Delta p \) is the water vapour pressure difference between the two sides of the film \( \Delta p = \rho_{\text{out}} - \rho_{\text{in}} \).

3.2. Assessment of shelf-life of packed granola breakfast product

An analysis of variance (ANOVA) of the shelf-life data showed that the moisture uptake by Granola was significantly affected \( (p < 0.05) \) by storage time and type of packaging material. Therefore, the permeance of the packaging film is crucial for control of the moisture uptake by Granola and consequently its shelf-life. The BOPP package showed to be the best barrier to water in comparison to all the materials studied, and showing a similar performance to the control package. Granola moisture content packed in BOPP package was 12.6 ± 0.213% (d.b.) and in control package was 15.6 ± 3.26% (d.b.), after 82 days of accelerated storage conditions. The N913 film showed moisture content values of 22.2 ± 0.293%

Fig. 1. Goodness of fit of the overall WVTR model describing the water vapour transfer through the BOPP (a), N913 (b), NK (c) and NM (d) films in the temperature range of 10–40 °C, and relative humidity of 33–95% for up to 14 days. Data points correspond to the experimental data and solid line to the predicted model.
(d.b.) after 82 days of accelerated storage conditions, and showed to be the best material, among the biodegradable NatureFlex films, to protect the food product against water absorption.

The predicted shelf-life of packaged Granola was determined by solving the Eq.(6) numerically for each type of packaging film at accelerated (38°C and 90% RH) and normal (20°C and 75% RH) storage conditions (Table 3). Validation experiments were performed at the accelerated conditions (38°C and 90% RH), and Granola’s shelf-life was evaluated based on critical moisture quality threshold as defined from the correlation between moisture content and the sensory acceptability in terms of texture. Palazon et al. (2009) developed a shelf-life model based on the storage temperature for a homogenized fruit-based baby food and set the threshold shelf-life of the product according to the overall acceptability by a sensory panel, considering a 50% of acceptability.

The predicted and experimental shelf-life at 38°C and 90% RH showed a good agreement always with a conservative estimate. This is due to the laboratory storage conditions. The model assumes that the only resistance to water vapour transfer is the permeability of the film. In laboratory settings the environmental conditions surrounding the packages are not disturbed and therefore it is

### Table 2

<table>
<thead>
<tr>
<th>Film</th>
<th>WVTR model</th>
<th>Lower limit joint confidence interval</th>
<th>Higher limit joint confidence interval</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOPP</td>
<td>7.019</td>
<td>5.792</td>
<td>7.814</td>
</tr>
<tr>
<td>N913</td>
<td>13.438</td>
<td>10.39</td>
<td>16.30</td>
</tr>
<tr>
<td>NK</td>
<td>27.695</td>
<td>28.75</td>
<td>29.81</td>
</tr>
<tr>
<td>NM</td>
<td>45.87</td>
<td>35.62</td>
<td>54.66</td>
</tr>
</tbody>
</table>

### Table 3

Shelf-life of granola for the different packaging films under accelerated (38°C and 90% RH) and normal storage (20°C and 75% RH) conditions. Permeance was predicted by Eqs. (7) and Eq.(8), and shelf-life predicted using Eq.(6). Validation was performed experimentally under accelerated conditions.

<table>
<thead>
<tr>
<th>Packaging film</th>
<th>Accelerated storage conditions 38°C and 90% RH</th>
<th>Normal storage conditions 20°C and 75% RH</th>
</tr>
</thead>
<tbody>
<tr>
<td>Permeance (g/m² day Pa)</td>
<td>Shelf-life (days)</td>
<td>Experimental shelf-life (days)</td>
</tr>
<tr>
<td>Control</td>
<td>–</td>
<td>21</td>
</tr>
<tr>
<td>BOPP</td>
<td>0.0012</td>
<td>13</td>
</tr>
<tr>
<td>NK</td>
<td>0.0047</td>
<td>3</td>
</tr>
<tr>
<td>NM</td>
<td>0.0078</td>
<td>2</td>
</tr>
<tr>
<td>N913</td>
<td>0.0023</td>
<td>7</td>
</tr>
</tbody>
</table>

* Average shelf-life of similar commercial products.
likely that there is a relevant external resistance to mass transfer. In a real scenario of transportation and storage in large cold rooms with blast cold air causing turbulence this resistance will be minimised and therefore the conservative estimate of the model is safer for practical purposes. Jena and Das (2012) found that the shelf-life of coconut milk powder packed in aluminium foil laminated could be predicted satisfactorily by empirical models showing a good fit with relative deviation percent below 10%.

4. Conclusions

The developed WVTR global model considering the dependency of temperature and relative humidity was found to fit very well the experimental data ($R^2 > 0.914$). The BOPP packaging film was the least influenced film by relative humidity, and temperature was the major influential parameter. For N913, NK and NM, the effect of temperature was much more pronounced at higher relative humidity.

Moisture uptake for packaged Granola was found to be significantly affected by both storage time and packaging material. The BOPP film was found to be the best barrier to water followed by the biodegradable N913 film. Under accelerated conditions, the predicted shelf-life of Granola ranged from 13, 7, 3, and 2 days in BOPP, N913, NK, NM pouches, respectively showing a good agreement with the experimental shelf-life, always with a conservative estimate. Under normal storage conditions the predicted shelf-life of Granola ranged from 271, 269, 90, and 33 days in BOPP, N913, NK, and NM pouches, respectively.

Integrative mathematical modelling describing the effect of system variables on critical Granola characteristics and on the packaging film permeability according to a QbD approach facilitates simulation and packaging design, making change of packaging easier and less expensive due to the reduction on the amount of experimental work required for its validation.

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