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Selecting packaging material for dry food products by trade-off of sustainability and performance: a case study on cookies and milk powder

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Abstract (max 250 words):

Alternative packaging concepts for two dry, shelf-stable food products were evaluated. A trade-off between recyclability (monolayer materials) and performance (multilayered high-barrier materials) was made for the packaging materials. Caramellized cookies were packaged in flowpacks made of PP film (OTR 1307cc/m²/d, WVTR 5g/m²/d), acryl-coated PVdC/PP film (OTR 21.8cc/m²/d, WVTR 4.2g/m²/d) as a reference material and metallized PP (MPP) film (OTR 31.2cc/m²/d, WVTR 0.4g/m²/d) and stored at 22°C and 50% relative humidity. Texture was compromised after six months of storage for the former two materials, while the latter provided an extension of textural acceptability. Whole milk powder was packaged in unsealed PE bags as a reference, representing a typical paper bag with a PE liner that is stapled shut without a seal. Alternative packages were sealed PE bags (OTR 1464cc/m²/d, WVTR 3g/m²/d), PE/PA/EVOH/PA/PE (OTR 0.25cc/m²/d, WVTR 0.95g/m²/d) and PA/EVOH/PA/PE (OTR 1.24cc/m²/d, WVTR 8g/m²/d) multilayer bags, and PP/Al/PE (OTR 0.1cc/m²/d, WVTR 0.1g/m²/d) bags and stored at room temperature and relative humidity between 70 and 90%. Unsealed bags were found to be unacceptable for storage at high humidity, due to excessive caking, discolouration and mould growth. Sealed PE bags provided adequate protection against moisture, yet not against oxygen ingress, leading to oxidative off-odours. The barrier efficiency of PA/EVOH/PA/PE was compromised by the high humidity. Both PE/PA/EVOH/PA/PE and PP/Al/PE bags provided adequate protection for over a year.

32 **Keywords:** Packaging, oxygen transmission rates, water vapour transmission rates, cookies,
33 milk powder

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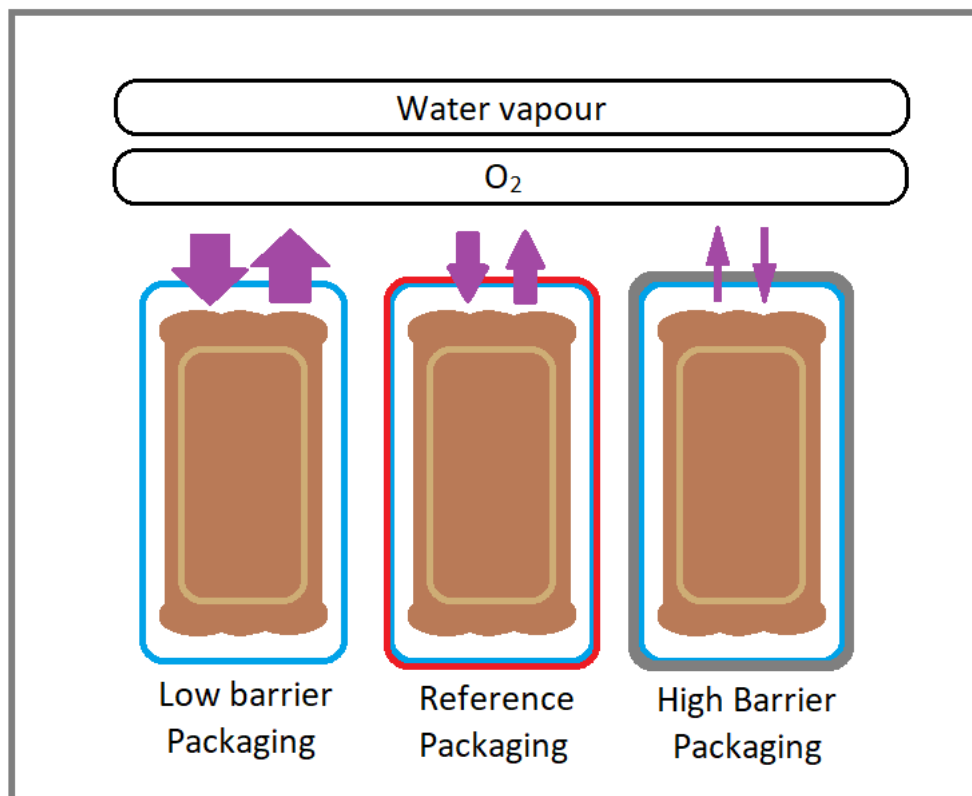
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36 **Graphical table of content**

37 **Balancing packaging sustainability and performance: a case study on**
38 **dry food products**

39 *Maarten Baele^{a,b}, An Vermeulen^{a,c}, Dimitri Adons^d, Roos Peeters^d, Frank*
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41 The effect of a typical reference and some alternative packaging materials on the shelf life of
42 two shelf-stable products, cookies and milk powder, was evaluated. Cookies were packaged in
43 PP film, acryl-coated PVdC/PP film and metallized PP film. Moisture content and texture
44 evolution in the different materials were evaluated. Milk powder was packaged in unsealed PE
45 bags, sealed PE bags, two multilayer bags and an aluminum laminate bag. Moisture content,
46 lipid oxidation and colour were monitored.



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50 Introduction

51 Dry food products are characterized by a low moisture content and low water activity and thus
52 are not susceptible to spoilage through the growth of bacteria, yeasts and moulds. Their spoilage
53 is caused by physical and chemical processes. Two such products, caramellized cookies and
54 milk powder, were considered in this study. As these products often have a shelf life of multiple
55 months up to over a year, it may often prove challenging for producers to guarantee a near-fresh
56 experience for consumers throughout the whole shelf life. As the optimal storage conditions
57 can not be guaranteed by the consumer, sufficient protection of the product through adequate
58 packaging is required [1-5]. Most importantly, protection against oxygen and water vapour is
59 needed.

60 Intense protection of these foods could rather easily be achieved by utilizing highly performant
61 packaging materials with very low permeability to oxygen and water vapour, such as metal
62 laminates [6, 7]. However, multi-layered materials are under a lot of pressure recently,
63 especially since the European Commission has recently issued the decision that by 2030, all
64 plastics placed on the European market should be either recyclable or reusable [8]. As such it
65 is opportune to evaluate whether currently established packaging concepts may be optimized
66 by, for example, switching to thinner or monolayered materials which are more easily
67 recyclable and likely lower priced. However, maintenance of a proper shelf life should be
68 paramount.

69 Caramellized cookies are a common confectionary product to be served as a snack alongside
70 coffee, and thus are often packaged as single servings. Two quality parameters are important
71 for this type of product: aroma and texture [9]. First, the typical caramellized aroma should be
72 preserved. Packaging materials with good gas barrier properties commonly imply good aroma
73 barrier properties as well: most relevant aroma compounds are volatile organic substances
74 typically with a higher molecular weight than oxygen and water vapour. Consequently they

75 would permeate through the packaging material at a slower rate. Texture deterioration is due to
76 uptake of moisture, which causes the starch to shift from a glassy to a rubbery state. To the
77 consumer, this is translated as crisp and crumbly cookies turning soggy, tough, chewy and/or
78 stale [10, 11].

79 The effect of storage conditions on texture of cookies is extensively studied in literature.
80 However, the research is highly heterogenic, due to the immense amount of specific types of
81 products to be considered. Some cookies are meant to bend and be soft, so moisture loss should
82 be prevented to avoid hardening [10, 12]. Others are meant to be crispy and crunchy, so
83 moisture ingress should be avoided [13-16]. Some products are sensitive to oxidation [1, 17-
84 19]. Others are not in particular, as they may possess an intermediate water activity level at
85 which oxidation rates are minimal [5, 19, 20]. This shows that packaging materials should
86 provide a highly tailored protection.

87 As such, Romeo et al. [21] compared plastic trays open to ambient air, polyvinylchloride (PVC)
88 film, aluminum foil and N₂-flushed trays in (polyamide/polyethylene) PA/PE film for storage
89 of Italian almond pastries. The flushed overwrapped trays performed best, followed by
90 aluminum foil packaging. Similarly, Piga et al. [12] compared PVC film and aluminum (Al)
91 foil for soft italian almond cookies. Once more, PVC was outperformed by the aluminum foil
92 in terms of preserving the cookies' texture (in this case, through prevention of moisture loss).
93 Romani et al. [5] compared metallized polypropylene (MPP)/paper films, metallized polylactic
94 acid (M-PLA)/paper and an oxo-degradable MPP/paper with a pro-oxidant additive for biscuits.
95 Despite the PLA-based material being more permeable, the authors found no significant
96 differences in quality parameters dependent on moisture and oxygen ingress between the
97 different packaging types during an accelerated storage test. It should be mentioned though that
98 oxodegradable films has become very controversial due to the formation of microplastics during

99 degradation [8]. Nagi et al. [22] showed that both high density polyethylene (HDPE) and
100 aluminum laminate were appropriate for storage of cereal bran cookies.

101 Milk powder, commonly produced by spray-drying of whole milk, is a free-flowing powder
102 consisting of milk proteins and milk fat globules embedded in an amorphous lactose matrix
103 [23-25]. Three phenomena dictate the loss of milk powder quality during its storage [24-26].

104 The most crucial one is the crystallisation of the otherwise amorphous or glassy lactose.
105 Crystallisation only occurs once the glass transition temperature of lactose is exceeded. This
106 temperature depends, amongst other factors, upon the moisture content: an increase lowers the
107 glass temperature [26-28]. Thus, if powder is kept at a constant temperature, initially below the
108 glass transition temperature, crystallisation may occur due to moisture sorption. As amorphous
109 lactose is very hygroscopic, sorption of moisture will occur very rapidly provided the
110 availability. Crystallisation of lactose greatly alters the functionality of the powder, leading to
111 decreased flowability, decreased solubility and caking. A second phenomenon which can occur
112 is discolouration due to nonenzymatic browning [25]. This defect is due to the formation of
113 brown pigments as a result of the Maillard reaction, in this case the result of a condensation of
114 lysine and lactose [24]. The consequence is twofold: sensory rejection increases due to visual
115 browning, and the nutritional value of the powder decreases as lysine is an essential amino acid
116 and thus a relevant nutrient in dairy products [3]. The third relevant phenomenon is lipid
117 oxidation. This is the reaction of mostly unsaturated lipids with oxygen. Advanced lipid
118 oxidation leads to decreased nutritional value, as vitamin A and essential fatty acids are
119 degraded. In addition, rancidity develops due to the generation of off-odours [29-33].

120 Rehman et al. [4] found similar performance for kraft paper bags and LDPE/PP bags for skim
121 milk powder storage. Lim et al. [34] found paper/PE packaging material to be inferior to Al-PE
122 laminate and paper/PA/PE during storage at 40°C, but not at 20°C. Camargo Novaes et al. [35]
123 compared metallized polyethylene terephthalate (M-PET)/PE and PP/PE bags, and composite

124 and metal cans for the storage of whole milk powders at elevated temperatures and high
125 humidity. The water vapour barrier of PP/PE proved insufficient, leading to caking of the
126 powder. An et al. [36] determined water vapour and oxygen barrier requirements, based on the
127 onset of lactose crystallization, for single-serving milk powder for infants to achieve a shelf life
128 over six months. Requirements towards oxygen transmission rates (OTR) and water vapour
129 transmission rates (WVTR) were very strict due to high surface area/product ratio. As such,
130 metal or composite cans or aluminum-laminated flexible films were proposed.

131 The aim of the current study was to evaluate a range of well-defined alternative packaging
132 materials spanning a range of OTR and WVTR values as compared to a reference material. This
133 is done for caramellized cookies and whole milk powder stored in controlled temperature and
134 humidity conditions. Complex multilayered packaging materials have the benefit of providing
135 superior water vapour and gas barrier properties. For some products, the use of these packaging
136 materials may provide an extended shelf life as compared to using less complex, low-barrier
137 packaging concepts. Less complex materials, and in particular monomaterial packages, trade
138 off barrier properties for improved recyclability.

139 For cookies packaged per unit in flowpack wrappers, PP film and metallized PP film (further
140 referred to as MPP film) were compared to an acryl-coated PP/PVdC film (further referred to
141 as reference film), specifically focusing on texture and moisture content. For whole milk
142 powder, an unsealed PE bag (representing a paper bag with plastic liner that is glued or stapled
143 shut) was compared to sealed PE bags (representing sealed common plastic liners), two
144 multilayer bags (PE/PA/EVOH/PA/PE and PA/EVOH/PA/PE) and a PP/Al/PE bag. In this
145 case, moisture content, colour, caking and lipid oxidation were the considered parameters. The
146 set-up, product types and packaging types of this test were selected in close collaboration with
147 packaging and food producers as part of a broader research project aiming to optimize
148 packaging and storage concepts for different food products.

149 Materials & methods

150 Food products and packaging

151 Caramellized cookies contained 4.9% protein, 19% fat and 72.6% carbohydrate, of which
152 38.1% sugars (data provided by the producer). The cookies were packaged individually in
153 flowpack wrappers on an industrial form-fill-seal line by the producer. Packaging was
154 performed in ambient air, no gas flushing of the packages was performed. Three different films
155 were used: the first was a PP film with an acryl/PVdC coating, which is a very common material
156 for packaging of cookies and will henceforth be called the 'reference film' [5, 37]. The other
157 two were a standard PP film (having lower water vapour barrier properties than the reference),
158 and metallized PP (MPP) (having higher water vapour barrier properties, but lower oxygen
159 barrier properties than the reference). Water vapour transmission rate (WVTR) of the materials
160 were gathered from the material's technical sheets. For the O₂ transmission rate (OTR) of the
161 films, the technical sheets did not provide consistent measurement conditions. Thus, the OTR
162 was experimentally determined according to ASTM standard F1927 using a Mocon Ox-Tran
163 2/21 (Mocon, Minneapolis, USA). 99.9% pure O₂ was used as a test gas. Measurements were
164 performed at 23°C, at 0% relative humidity at the inside of the test material and 50% at the
165 outside of the test material. These conditions were chosen to reflect a real storage situation. The
166 transmission properties of the materials are provided in Table 1.

167 Milk powder was delivered in 25kg paper bags. Based on the specification sheet, the powder
168 contained 2.67% moisture, 26.4% fat, 24.6% protein and 39.9% lactose. After delivery, the milk
169 powder was weighed in 1 kg portions in paper bags lacking any water vapour or gas barrier.
170 These were used to contain the portions and to prevent dusting of the powder during further
171 handling. The portions, as contained in the paper bags, were stored in three types of plastic bags
172 (as shown in Table 2) and sealed using a Multivac C300 vacuum packaging machine (Multivac,
173 Mechelen, Belgium). Before sealing, the machine's chamber was subjected to a vacuum step at

174 5 mbar, flushed with a modified atmosphere packaging (MAP) gas mixture (30% CO₂, 70%
175 N₂) until a pressure of 350 mbar, after which the bags were sealed and the vacuum was
176 compensated in the chamber. The gas mixture was created using a Witt MG18-3MSO gas mixer
177 (Gasetechnik, Germany). CO₂ was included in the MAP gas mixture as suggested by the milk
178 powder supplier, who empirically found improvements in keeping quality of the milk powder
179 (lipid oxidation) due to CO₂ inclusion. As a reference, PE bags that were kept unsealed were
180 used to represent a package lacking any barrier against oxygen and water vapour (Table 2),
181 such as the paper bags in which the powder was supplied [4]. The WVTR of the materials were
182 determined according to ASTM standard F1249 using a Mocon Perma-tran 700 (Mocon,
183 Minneapolis, USA). Measurements were performed at 38°C, at 0% relative humidity at the
184 inside of the test material and 90% at the outside of the test material. The OTR of the materials
185 was determined as described above. As a final fifth alternative, PP/Al/PE bags with superior
186 oxygen and water vapour barrier properties (data collected from the supplier's technical data
187 sheets) were used.

188 **Storage and sampling**

189 The cookies were stored in a dry climate chamber at 22°C for the first two months of storage.
190 For the remainder of the test, the cookies were moved to tightly sealed cabinets containing jars
191 with an aqueous saturated NaBr (Sigma-Aldrich, Overijse, Belgium) solution, allowing for a
192 controlled relative humidity (RH) equilibrium set at 50% [38]. Measurements showed that the
193 actual RH fluctuated between 40 and 60%. The cabinets were kept in 22°C incubators (KB8400,
194 Termaks, Bergen, Norway). These conditions were chosen to simulate a typical storage period
195 in a European retail environment.

196 The milk powder bags were stored in a tightly sealed container, located within a climate room.
197 The amount of sample bags had to be limited due to size restraints of this container. The
198 temperature of this room was set at 30°C for the first two months of storage, then at 22°C for

199 the remainder of the test. The elevated temperature was included to simulate a period of
200 temperature abuse during storage or transport [4]. Below a perforated grid supporting the bags,
201 the bottom of the cabinet was filled with an aqueous saturated KNO_3 (VWR international,
202 Leuven, Belgium) solution, allowing a relative humidity equilibrium within the cabinet in the
203 order of 90% RH [38]. Measurements revealed that the relative humidity started at
204 approximately 70% and only slowly rose to 90% during the test. These conditions, including
205 temperature abuse and high humidity, were chosen to simulate overseas container transport.

206 Temperature and relative humidity within the storage cabinets were monitored using a Testo
207 623 temperature-humidity logger (Testo, Ternat, Belgium).

208 Once per month over the span of one year, samples were removed from storage for analysis.
209 For the cookies, multiplicates were used and one analysis was performed per sample, for
210 multiple samples. For the milk powder, one bag was opened per analysis day per treatment
211 (packaging type). On these samples, analyses were performed at least in triplicate. In this case,
212 the amount of samples was smaller due to space restrictions of the climate cabinet and the large
213 size of the individual bags. It should be mentioned that this results in reduced robustness of the
214 statistical analysis of these results.

215 Analyses

216 Fatty acid composition

217 Lipids were extracted from the cookies using the Bligh and Dyer method, in which chloroform
218 was substituted by dichloromethane [39]. Fat from the milk powder was extracted using the
219 IDF method 265 [40]. Fatty acid composition was determined using AOCS method Ce 1b-89
220 [41] using capillary gaschromatography as described by Mestdagh et al. [42]. The fatty acid
221 composition of the cookies is shown in Table 3. The fatty acid composition of the milk powder
222 is shown in Table 4.

223 Moisture content, water activity and moisture sorption

224 Before analysis of the cookie samples, the cookies were ground into powder using a pestle and
225 mortar. Moisture content was determined by mixing approximately 5g of sample with sand in
226 a metal cup. The sand had been dried at 105°C for 24h in advance and cooled to room
227 temperature in a dessicator prior to use. The mixture of sand and sample was dried for 2h at
228 105°C, cooled down in a dessicator and weighed. The sample was then dried for another 30
229 minutes, cooled and reweighed. This drying and weighing was repeated until the weight
230 stabilised [43]. The cookies were ground into powder before this analysis.

231 Water activity at 25°C was determined using an Aqualab Vapor Sorption analyzer (Decagon
232 Devices Inc., Meter Group Europe, München, Germany).

233 The same apparatus was used to determine the adsorption-type vapour sorption isotherms using
234 the dynamic dewpoint isotherm (DDI) method. The adsorption was started at the experimentally
235 determined initial a_w and moisture content values. The sorption data were analysed using the
236 VSA downloader software (Decagon Devices Inc., Meter Group Europe, München, Germany).

237 Texture measurements

238 Cookies were subjected to a three-point bend test using an Instron 5942 Single Column system
239 (Instron Benelux, Boechout, Belgium) [5, 14, 15, 21]. Using a 500N load cell, a plastic blade
240 (CNS Farnell, UK) was lowered onto the sample at a constant rate of 1mm/s. The cookie was
241 supported on the two opposite ends at exactly 5 cm apart. Care was taken so that the probe was
242 lowered onto the middle of the cookie. The measurement initiated as soon as the measured force
243 reached 0.1N. The required force (N) to maintain the probe at a constant speed was returned as
244 a function of the displacement (mm). The test ended as soon as the force decreased to 5% of
245 the maximum force measured during the test. The data was gathered in the Instron Bluehill 3
246 software. ‘Hardness’ was defined as the value of the peak force (N) applied during the test [5,
247 44]. This peak force was registered right before the cookie breaks. ‘Fracturability’ was defined

248 as the inverse of the displacement of the probe (or deformation of the cookie) right before the
249 cookie breaks, when peak force is registered. An arbitrary threshold was set at 0.6 mm
250 deformation, as none of the fresh cookies extended past this point before breaking during the
251 initial measurements. The product of hardness and fracturability can be defined as the ‘rigidity’
252 (stress/strain or peak force in N/displacement at peak force in mm) [45]. The ‘end of test’
253 extension was used as an extra measure for the brittleness of the cookies: for a brittle cookie,
254 the test will end very quickly after the peak force is registered, as both halves of the now broken
255 cookie will fall off the support and no longer exert a pressure on the probe. When the cookie
256 has become less brittle, it keeps exerting force on the probe as it bends, and thus the apparatus
257 will keep registering force at higher deformation. As such, extreme values found in these texture
258 measurements cannot be regarded as statistical outliers, but are rather real cases of failure.

259 [Headspace oxygen concentration](#)

260 Oxygen concentrations within the milk powder bags were monitored non-destructively with the
261 aid of OxyDot sensors on the inside of the bags, read out optically using the Oxysense 210T
262 system (Oxysense Inc., Dallas, US) [46]. These measurements were not performed for the
263 opaque PP/Al/PE bags, as transparency of the bags was a requirement to allow the sensors to
264 be read out.

265 [Hexanal](#)

266 Hexanal, a reaction product from the oxidation of linoleic acid, was monitored as a marker for
267 the extent of lipid oxidation in milk powder [47]. Lipid oxidation was also identified by the
268 supplier of the milk powder as a highly common index of failure. Hexanal concentration was
269 determined using headspace solid-phase microextraction (HS-SPME) coupled with a gas
270 chromatograph with mass spectrometer (GC-MS).

271 Phosphate buffer was prepared by dissolving 8.95g/L disodium hydrogen phosphate
272 dodecahydrate (Chem-lab Analytical, Zedelgem, Belgium) and 3.04g/L potassium dihydrogen

273 phosphate (Chem-lab Analytical, Zedelgem, Belgium) in water and adjusting to pH 2 with
274 orthophosphoric acid (Fischer Scientific, Merelbeke, Belgium). Antioxidant solution consisted
275 of 0.25g butylated hydroxyanisole (Sigma-Aldrich, Overijse, Belgium) in 0.5ml HPLC grade
276 methanol (Fischer Scientific, Merelbeke, Belgium). Internal standard solution consisted of
277 25mg/ml hexanal-d12 (CDN Isotopes, Quebec, Canada) in HPLC grade methanol further
278 diluted to 37.5 μ g/ml in water. A calibration standard solution was prepared from a 0.815mg/ml
279 solution of hexanal (Sigma-Aldrich, Overijse, Belgium) in HPLC grade methanol, further
280 diluted to 0.815 μ g/ml in water.

281 Samples were prepared by adding 1g of milk powder to a 20 ml glass headspace vial, also
282 containing 4ml of phosphate buffer, 5 μ l of antioxidant solution and 15 μ l of internal standard
283 solution. External calibration curves were created by standard addition, through adding 0-75-
284 150-300-450-600 μ l of this standard solution to headspace vials containing 1g of milk powder
285 (that had been stored in vacuum prior to use, to avoid excessive oxidation of these samples),
286 5 μ l of antioxidant solution, 15 μ l of internal standard solution and 4ml of buffer solution. The
287 vials were sealed with PTFE/silicone caps (Agilent Technologies, Palo Alto, CA).

288 The HS-SPME process was performed automatically by a CTC-CombiPAL autosampler
289 (Agilent Technologies, Palo Alto, CA). The samples were pre-incubated at 75 $^{\circ}$ C for 5 minutes,
290 whilst being agitated at 500 rpm. Extraction was done for 10 minutes with a DVB/CAR/PDMS
291 gray fiber (Supelco, Bellefonte PA, US). The fiber was desorbed for 2 minutes at 250 $^{\circ}$ C in the
292 inlet. For chromatographic analysis, an Agilent 7890A GC with a 5975C mass spectrometer
293 (Agilent Technologies, Palo Alto, CA) and equipped with an Agilent J&W DB-624 60m,
294 0.25mm, 1.40 μ m capillary column was used. Helium was used as carrier gas at a constant flow
295 rate of 1.3ml/min. The oven temperature was programmed as follows: initial temperature 50 $^{\circ}$ C,
296 hold time 5 min; ramp up to 140 $^{\circ}$ C at 4.5 $^{\circ}$ C/min. The mass spectrometer detector (MSD)
297 conditions were the following: transfer line to MSD, 280 $^{\circ}$ C; ionization energy, 70 eV;

298 operating in selective ion mode (SIM). The selected ions monitored were m/z 44, m/z 46, m/z
299 56 and m/z 64, quantifiers were m/z 56 for hexanal and m/z 64 for hexanal-d₁₂.

300 Oxidation was not evaluated for the cookies. The extent of lipid oxidation in a food product is
301 based on many factors such as fatty acid composition, the level and kind of pro- and
302 antioxidants, presence of free fatty acids. Based on internal experience, the producer specified
303 that oxidation is not a specific point of failure for their cookies, while texture was the key
304 concern. Sakač et al. [16] stored cookies in 40µm PP film and only detected significant
305 oxidation after 14 months of storage. Romani et al. [5] found little difference in hexanal
306 formation when comparing cookies stored in packaging materials with OTR ranging from 1.6
307 to 15 cc/m²/d.

308 Colour

309 Colour evolution of the milk powder was monitored using a Konica Minolta CM2500-D
310 (Konica Minolta Sensing Europe, Bremen, Germany) handheld colour analyser set at D65
311 illuminant and 10° standard observer. Measurements were performed on a sample of powder
312 that had been placed in transparent bags, in a layer of at least 2cm thick. The results were
313 rendered as tristimulus colour parameters in the CIELAB colour space, and resulted from
314 averaging 10 measurements per sample. The cookies' colour was not analyzed instrumentally
315 in this test.

316 Statistics

317 Data were analysed using R Statistics 3.5 [48]. Significance of the main effects 'packaging
318 type', and 'time of storage' and the interaction effect were evaluated using analysis of variance
319 (ANOVA) on univariate general linear models. The appropriateness of these models was
320 checked using a histogram of the residuals. A main effect was interpreted as a global difference
321 over all levels of the other factor. An interaction effect was interpreted as an evolving effect of
322 packaging type over time. Levene's test was used to check homogeneity of variances. Estimated

323 marginal means were used to compare pairs of levels of one factor for all levels of the other
324 two factors. Comparisons were based on Fischer's least significant difference test (LSD).

325 Results

326 Cookies

327 Moisture content and water activity

328 The cookies' moisture sorption isotherm is shown in Figure 1. The isotherm showed a type III
329 shape, as classified by Brunauer et al. [49]. While type II isotherms are also reported for biscuits
330 and cookies, this is a common finding for sugary confectionery products [13, 50-53]. As
331 discussed by Lewicki et al. [54], these curves possess inflection points that indicate crucial
332 moisture levels. Exceeding these levels imparts critical physical changes to the product. These
333 were automatically calculated by the data gathering software, showing two inflection points in
334 the curve: one at a_w 0.45 and 3.5% moisture content, and one at a_w 0.80 and 16% moisture
335 content. The latter seems rather irrelevant, as the cookies would be far from acceptable from a
336 consumer's point of view at this level of moisture content. Thus, a water activity of 0.45 and
337 moisture content of 3.5% was regarded as a preliminary index of failure for the cookies. As an
338 extra control, a GAB model was fitted to the curve [53-55]. A good overall fit was achieved
339 and an inflection point at 4.5% moisture and a_w 0.50. This would indicate that for the cookies,
340 a critical range exists between 3.5 and 4.5g moisture per 100g dry matter, or between a water
341 activity of 0.45-0.50. The lower value of this range will be used as an indicative critical point.

342 **Figure: FIGURE_1**

343 *Figure 1: Moisture sorption isotherm of crumbled cookies. The dotted lines indicate the curve's two inflection points.*

344 The initial moisture content of the cookies was between 1.3% and 1.9%, corresponding to a
345 water activity of 0.14 ± 0.03 . As there was little difference in WVTR between the PP film and
346 the reference film, moisture content of the cookies packaged in these two films progressed
347 similarly. Cookies packaged in PP film surpassed a moisture content of 3.5% after 3 months,

348 cookies packaged in the reference film after 4 months (Figure 2). Analysis of variance showed
349 no significant difference in change over time when comparing only these two packaging types
350 ($p=0.069$). Cookies packaged in the MPP film had an overall lower moisture content and
351 reached 3.5% moisture after about 8 months of storage.

352 **Figure: FIGURE_2**

353 *Figure 2: Moisture content (dry matter base) of cookies packaged in (left) PP film, (middle) reference film and (right) MPP*
354 *film as a function of storage time. The dotted line indicates a critical moisture content, determined from the moisture sorption*
355 *isotherm.*

356 **Figure: FIGURE_3**

357 *Figure 3: Water activity of cookies packaged in (left) PP film, (middle) reference film and (right) MPP film as a function of*
358 *storage time. The dotted line indicates a critical water activity, determined from the moisture sorption isotherm.*

359 Considering water activity (Figure 3), analysis of variance showed no significant difference in
360 change over time between the PP film and reference film ($p=0.111$). The a_w 0.45 threshold was
361 passed between 7-9 months of storage. Cookies packaged in the MPP film did not exceed an a_w
362 of 0.45 within the duration of the test. The results for water activity showed discrepancies from
363 the expectations based on the moisture adsorption isotherm and the moisture content during
364 storage (Figure 4). This indicates that in reality, the cookies' water activity after certain storage
365 periods corresponded with somewhat higher moisture contents than found in the moisture
366 sorption isotherm. This is illustrated in Figure 4. It should be noted however that in order to
367 determine the sorption characteristics of the biscuit, the sample was ground in a mortar into a
368 powder, thus increasing considerably the specific surface of the matrix and also removing
369 possible 'capillary' effects within the cookie. It is therefore expected that the number of sorption
370 places increased in parallel and thus that at a particular moisture content, a lower water activity
371 is observed for the powdered cookie on which the sorption isotherm was determined compared
372 to the stored biscuit. The opposite was observed however, especially at water contents lower
373 than 6 % (on db). This deviation can be explained based on the recent report of Karrila and
374 Karrila (2020) [56]. In their study using the same instrument and DDI method to determine the

375 sorption isotherm it was observed the sorption characteristics of a powdered starch based snack
376 was dependent upon the particle size. At the same moisture content powders with a particle size
377 was $< 180 \mu\text{m}$ had a lower water activity compared to powder of the same snack of which the
378 particle size was $3000 \mu\text{m}$. These authors explained this phenomenon by the absence of rate
379 effects in the absorption process of water to its binding places if the particle size of the powder
380 is small enough, while these are relevant if the particle size of the powder is too big. Most likely
381 the powder size of the ground biscuits was too high to neutralise such rate effects in our
382 conditions.

383 **Figure: FIGURE_4**

384 *Figure 4: Moisture sorption isotherm (line) of crumbled cookies and measured a_w - moisture content (dry basis) couples of*
385 *intact cookies stored in PP film (●), reference film (○) and MPP film (▼) during the storage test. The lines indicate the*
386 *critical a_w and moisture content as derived from the isotherm.*

387 **Texture measurements**

388 Textural attributes of bakery products are highly product-specific and often hard to interpret
389 and relate to consumer experience [15, 45, 57, 58]. Fresh caramellized cookies are characterised
390 by a ‘snap’ when force is applied by the consumer’s teeth. In texture measurements, this was
391 reflected by three characteristics: 1) a high peak force, 2) a low deformation at peak force and
392 3) a sharp and immediate drop in force after the cookie breaks (Figure 5). A loss of ‘snap’ was
393 expected after absorption of a certain amount of moisture. This was reflected by a lower peak
394 force, higher deformation (due to the cookie bending rather than breaking) and a tail on the
395 texture curve, due to the force required to further bend the cookie as illustrated in Figure 5. This
396 figure shows results for a cookie that was kept without packaging material at room temperature
397 for 2 weeks.

398 **Figure: FIGURE_5**

399 *Figure 5: Comparison of typical texture profiles of fresh or ‘breaking’ (-●-) and failed or ‘bending’ (-×-) cookies. The*
400 *‘failed’ were kept without packaging at room temperature for two weeks before analysis.*

401 Despite obvious sensory changes on the texture of the cookies throughout storage, little
402 conclusion could be made from the evolution of the cookies' hardness (Figure 6). Due to the
403 high variability of the data, detecting statistically significant trends was hindered. Analysis of
404 variance showed no overall effect of packaging type on hardness ($p=0.257$), but it did affect the
405 changes throughout time ($p=0.016$) (see Supplemental Table 1). When the hardness was divided
406 by the deformation at peak force, negative trends appeared for all packaging types (Figure 7).
407 The decrease in rigidity was exhibited a less pronounced trend for the MPP film, which may be
408 linked to its better moisture barrier. Between 7 and 8 months a pronounced drop in the rigidity
409 was observed, most notably for the cookies packaged in PP film. From a sensory point of view,
410 loss of rigidity would mean that the cookies became tougher and less crispy. This corresponds
411 better with the observed water activity evolution (Figure 3) than with the moisture content
412 (Figure 2). Most notably for PP films, water activity levels consistently exceed a_w 0.45 after 8
413 months of storage, corresponding to the rigidity drop mentioned above. For the PP film and
414 reference films, the moisture level had indeed exceeded 3.5% (Figure 2), but this had happened
415 some months before, indicating that critical moisture could be at a higher level than initially
416 proposed. The deformation at peak force (Figure 8) for the PP film and reference film packaged
417 cookies consistently surpassed the arbitrary threshold mentioned earlier, indicating that from
418 this point the cookies were no longer as crispy as if fresh. Apart from some outliers, cookies
419 packaged in the high-moisture barrier MPP film did not exceed this threshold as consistently.
420 The 'end of test' extension (Figure 9) followed a course highly similar to the extension at peak
421 force. After 5 months in the case of the reference packaging and 6 months in the case of PP film
422 packaging, no measurements were below the 0.6mm threshold and multiple high deformation
423 values (over 1mm and up to 15mm) were measured. It is remarkable that this occurred earlier
424 for the reference film, since this film has better moisture barrier than the PP film (Table 1). This
425 observation may be influenced by the sampling times being 1 month apart from each other and

426 test variability. More frequent analysis may have provided smoother curves showing clearer
427 results. The values for cookies packaged in MPP film started showing outliers and increased
428 towards the end of the storage period, but not as consistent and pronounced as for the other two
429 packaging types. It is likely that some outliers were due to microleaks, allowing more water
430 vapour to enter the packages. As the metallization imparted some more rigidity to the films, it
431 is possible that seals were not as complete as they were on the PP and reference films, as they
432 were all packaged on the same packaging lines regardless of the material used.

433 **Figure: FIGURE_6**

434 *Figure 6: Hardness of cookies stored in PP film (left), reference film (middle) and MPP film (right) as a function of storage*
435 *time.*

436 **Figure: FIGURE_7**

437 *Figure 7: Rigidity of the cookies stored in PP film (left), reference film (middle) and MPP film (right) as a function of storage*
438 *time.*

439 **Figure: FIGURE_8**

440 *Figure 8: Deformation at peak force of cookies stored in PP film (left), reference film (middle) and MPP film (right) as a*
441 *function of storage time. The dotted line represents an arbitrary threshold based on measurements on the fresh cookies.*

442 **Figure: FIGURE_9**

443 *Figure 9: Deformation at end of test for cookies stored in PP film (top left), reference film (top middle) and MPP film (top*
444 *right) as a function of storage time, and detail of the same plots at low extension (bottom row). The dotted line represents an*
445 *arbitrary threshold based on measurements on the fresh cookies.*

446 For the sake of completeness, it is important to notice that from a sensory point of view, the
447 cookies did not simply progress towards lower hardness and higher deformation at peak force.
448 For example, the cookies packaged in PP film evolved from hard and crispy to soft and bendable
449 (mosts clearly illustrated in Figure 8 by the dip in rigidity for PP film packaged cookies during
450 months 6-8) further towards tough and chewy, illustrated by the return to higher rigidity during
451 months 9-12. Most likely, this is due to redistribution of moisture and subsequent
452 recrystallisation of sugar [12].

453 Milk powder

454 Oxygen concentration

455 Oxygen concentrations measured in three of the used bags are shown in Figure 10. For the
456 unsealed bags, no concentrations were monitored as these bags were constantly subjected to
457 atmospheric oxygen due to the lack of a seal. The PP/Al/PE bags were not monitored as the
458 bags needed to be transparent to be able to read out the oxygen sensors. Despite the PE,
459 PE/PA/EVOH/PA/PE and PA/EVOH/PA/PE bags having the same thickness, differences in
460 measured oxygen concentration were massive. Due to the very high oxygen permeability (Table
461 2), milk powder packaged in PE bags was exposed to near-atmospheric oxygen concentrations.
462 However, towards the end of the test the concentrations in some of the bags dropped. The cause
463 of this is unclear, as oxygen consumed by the powder would be expected to be replenished through
464 permeation through the PE bag. Concentrations in the PE/PA/EVOH/PA/PE multilayer bags
465 remained below 1% for the majority of the test period, with a minor downward trend. The other
466 type of multilayer bags was highly similar in composition, only missing a PE layer on the
467 outside of the material. As PA and EVOH are polar polymers, and a high humidity was
468 maintained throughout the test, moisture affected the EVOH and PA barrier layers, where it
469 increased its permeability to oxygen, as seen in Figure 10 [59].

470 **Figure: FIGURE_10**

471 *Figure 10: Oxygen content measured within the headspace of (left) PE bags, (middle) PE/PA/EVOH/PA/PE bags, (right)*
472 *PA/EVOH/PA/PE bags during storage of milk powders.*

473 Moisture content and water activity

474 The milk powder manifested a combined type I – type II moisture sorption isotherm behaviour
475 (Figure 11) [49, 60]. Literature often finds type II isotherms for whole milk powder, and critical
476 points are found at a water content approaching 8% and a water activity of 0.50 [3, 25, 26, 61].
477 At this point amorphous lactose, which is crucial for physical properties such as flowability,
478 will start to crystallise.

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Figure: FIGURE_11

Figure 11: Moisture sorption isotherm of milk powder.

Both moisture content and water activity were clearly affected by packaging type ($p=0.000$ in both cases, see Supplementary Table 2). Initial moisture content was 2.7%. As expected, moisture was absorbed rapidly by the powder in the unsealed bags during the first 4 months of storage (Figure 12). A maximum in moisture content between 8 and 9% was reached after the fourth month of storage. During the following two months, a decrease in measured moisture content was observed. This was due to the crystallisation of lactose in the powder, after a critical amount of moisture had been reached. This corresponded with critical moisture contents found in literature [25, 61] and with the observed isotherm (Figure 11). After this drop, the moisture content increased again slowly during the remainder of the test. The observed evolution of the moisture content in the unsealed bags showed that a sealed packaging system providing a moisture barrier is really required for keeping milk powder in the high relative humidity conditions used in this test. None of the powder samples stored in sealed plastic bags showed a similarly pronounced phenomenon during the test. It took less than two months for a difference to appear between the powder stored in PE bags as compared to the PE/PA/EVOH/PA/PE multilayer bags. At the end of the storage test the difference in moisture content was not greater than 1%. After 12 months of storage, the PE/PA/EVOH/PA/PE bags reached approximately 4% of moisture. For the PA/EVOH/PA/PE bags, this level was already surpassed after 2 months of storage. Just over 6% of moisture was measured in the powder stored in the PA/EVOH/PA/PE bags at the end of the test. For the aluminum laminate bags, the initial moisture content of the milk powder was well retained (Figure 12).

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Figure: FIGURE_12

Figure 12: Moisture content of milk powder stored in (top left) unsealed PE bags, (top middle) PE/PA/EVOH/PA/PE multilayer bags, (top right) PA/EVOH/PA/PE multilayer bags, (bottom left) PE bags, (bottom right) aluminum laminate bags.

505 Initial water activity (Figure 13) of the milk powder was just below 0.3. Powder stored in the
506 unsealed bags showed a sharp rise in a_w after the first month of storage. Coincident with the
507 drop in moisture content, the water activity of these powders dropped and rose back to the
508 former level after 7 months of storage. The high relative humidity of the storage atmosphere
509 and high water activity of the powder in these bags allowed moulds to start growing on the
510 surface of the powder, visually noticeable from the 10th month of storage. For the powder in
511 PA/EVOH/PA/PE bags, a notable increase in a_w was observed towards the end of the test. For
512 the powder in aluminum laminate bags, the a_w changed very little. Despite the 3-fold difference
513 in water vapour barrier of the PE/PA/EVOH/PA/PE multilayer bags and the PE bags (Table 2),
514 the a_w evolution was highly similar between these two packaging types. Only at month 4 and
515 month 11 significant differences were found. Yet, no moisture-related cases of failure occurred
516 for these packaging types, implying that transmission rates of 0.95 and 3 g/m²/d are acceptable
517 transmission rates for milk powder stored in these conditions. Between the PE bags and the
518 PA/EVOH/PA/PE bags (WVTR 3 and 8 g/m²/d, respectively) there is also an almost 3-fold
519 difference in transmission rate, while in this case the water activity results do follow a different
520 trend. The absolute difference in WVTR between these two packaging types seems more
521 crucial, implying that the minimal WVTR for this case is somewhere between 8 and 3 g/m²/d.

522 **Figure: FIGURE_13**

523 *Figure 13: Water activity of milk powder stored in (top left) unsealed PE bags, (top middle) PE/PA/EVOH/PA/PE multilayer*
524 *bags, (top right) PA/EVOH/PA/PE multilayer bags, (bottom left) PE bags, (bottom right) aluminum laminate bags.*

525 The importance of moisture and crystallisation of lactose was reflected in the fact that powder
526 stored in unsealed PE exhibited strong caking. Apart from being an obvious visual index of
527 failure, this caking also greatly imparted the solubility of the powder. This was observed after
528 6 months of storage. The sorption isotherm shown in Figure 11 indicates an inflection point at
529 an a_w value of 0.40. Treatments showing caking indeed had water activities exceeding a_w 0.4,
530 indicating a critical a_w value existing around that point.

531 Lipid oxidation

532 Hexanal was monitored as a marker for secondary lipid oxidation. It is only one of many
533 different compounds formed during lipid oxidation, but it is one of the most predominant
534 compared to other secondary oxidation products and imparts a distinct grassy or hay-like smell
535 [29-33]. The significant increase over time measured in the sealed PE bags stands out (Figure
536 14). Oxidation was likely significant for this packaging concept due to the high amount of
537 oxygen available within these bags (Figure 10), and these values correspond with findings from
538 Lloyd et al. for ambient-packaged whole milk powder in 5kg portions stored for one year in
539 bags with OTR 1.5cc/m²/d [32]. The unsealed bags were also exposed to high oxygen levels,
540 but no corresponding high hexanal measurements were observed. A possible explanation for
541 this is that hexanal was formed but already decomposed further [32]. Another possibility is that
542 the formed hexanal was lost to the atmosphere, due to the volatility of this compound and the
543 lack of a seal that could restrain the hexanal within the packaging system. There was little
544 difference between the hexanal concentrations measured for the other packaging concepts.
545 Lloyd et al. also found that when a modified atmosphere with lowered oxygen content (<4%)
546 is preserved, formation of hexanal is mitigated [32]. 50ng/g was found to be a sensory threshold
547 for hexanal in milk [31, 62]. Seen as the powder is meant to be reconstituted as one part powder
548 for 9 parts water, 500ng/g was used as threshold for the powder. This was only exceeded in the
549 case of the powder packaged in sealed PE bags, after 6 months of storage. Despite other bags
550 having oxygen concentrations exceeding 5% over the course of the storage period (Figure 10),
551 no effect on hexanal formation could be observed, which was rather unexpected.

552 Figure: FIGURE_14

553 *Figure 14: Hexanal content of milk powders stored in unsealed bags (○), PE bags (●), PE/PA/EVOH/PA/PE multilayer*
554 *bags (▼), PA/EVOH/PA/PE multilayer bags (△) and Aluminum laminate bags (×).*

555 Colour

556 As seen in Figure 15, milk powder stored in unsealed bags exhibited a clear decrease in L*
557 value (lightness) and increase in a* (redness) and b* value (yellowness), already during the first

558 few months of storage. Visually, this was clearly noticeable as the powder takes a more yellow-
559 brown hue. These findings correspond to findings by Celestino [63]. Moisture is important for
560 the colour of milk powder: the rate of Maillard reaction, responsible for browning, increases
561 with water activity [25, 64]. This is due to increased mobility of reagents due to increased water
562 availability. No visually noticeable colour defects were observed in other packaging concepts,
563 which was confirmed by the limited difference in tristimulus colour parameters.

564 **Figure: FIGURE_15**

565 *Figure 15: Colour evolution of milk powder packaged in unsealed bags (○), PE bags (●), PE/PA/EVOH/PA/PE multilayer*
566 *bags (▼), PA/EVOH/PA/PE multilayer bags (△) and Aluminum laminate bags (×).*

567 Discussion

568 The initial comparison of fresh and failed cookies texture measurements as shown in Figure 5
569 seemed clear, but during the the actual storage test results were harder to interpret, mostly due
570 to the intrinsic variability associated to these measurements [5]. Despite this, it was rather clear
571 that cookies packaged in PP film and reference film started showing consistent textural failure
572 after six months. Rigidity, deformation at peak force and deformation at end of test seemed
573 useful measurements to determine this. Hardness or peak force was not as useful, as this did not
574 change as much or as clearly as anticipated. The fact that the producer's logo was baked into
575 the cookies, providing an irregular elevation, provided extra variability. Further studies should
576 consider extensive preliminary testing to get to know the variability and textural changes to be
577 expected for a certain kind of product.

578 Some caution had to be taken with the proposed moisture content threshold at 3.5% and water
579 activity threshold of 0.45. The latter was not achieved for the MPP film packaged cookies, while
580 texture failures did appear for these samples. Figure 4 suggests that during the actual storage
581 test, the moisture content of 3.5% corresponded with a water activity of just below 0.300, so
582 this may well be a more realistic threshold. Ideally, sorption isotherms should be determined

583 on the product as it is: crumbling the cookie for the determination of the isotherm may have
584 influenced the isotherm in the current study.

585 Romeo et al. [21] found that PA/PE film and aluminum foil was to be preferred over PVC film
586 to prevent texture change during storage of soft almond cookies. Nitrogen flushing of packages
587 provided additional benefits. Likewise, Piga et al. [12] found that storage in PVC film brought
588 about rapid texture deterioration in cookies and preferred aluminum foil. Mandala et al. [15]
589 experienced problems when attempting to relate moisture content to textural properties. Small
590 changes in moisture did not translate to significant changes in texture properties, due to high
591 intrinsic variability. Careful, product-specific interpretation of the data was needed. Romani et
592 al. [5] found that a metallized PLA/paper laminate may be suitable as a replacement for
593 metallized PP/paper laminate with comparable barrier properties for storage of biscuits. In the
594 current study, the MPP film provided some shelf life extension based on moisture-related
595 spoilage phenomena. However, due to the processing properties of this film, further
596 optimization of the packaging infrastructure was needed for this film to avoid microleakage.

597 It should be remarked however that consumer appreciation and shelf life of cookies is however
598 more complex than texture alone: despite lipid oxidation not being a primary concern of the
599 cookie producer, a certain degree of gas barrier may be beneficial to maintaining the cookie's
600 lipid quality and overall aroma. Therefore it would be interesting to include evaluation of aroma
601 evolution in future testing. This would require the use of a trained sensory panel.

602 Another remark is that these single-cookie packages are usually wrapped in a bundle film. This
603 imparts an extra hurdle for transmission of vapour and gases. The internal atmosphere in the
604 bundle film will be different from the ambient air, thus a different gradient will be present,
605 slowing down transmission. However, it is entirely possible that a consumer may open and
606 discard the bundle film shortly after purchase, so as a measure of precaution the shelf life should
607 be determined for the single-packaged rather than the bundle-packaged cookies. With this in

608 mind, the use of PP film would trade in only a fraction of the shelf life for an improved
609 recyclability of the packaging material.

610 It is clear that unsealed bags were not appropriate for storage of milk powder in conditions of
611 relative humidity above 70%. Moisture content increased rapidly, and crystallisation of lactose
612 occurred after 4 months of storage. By then, browning had already set in as well. Due to severe
613 caking further in storage, moisture could no longer easily redistribute within the powder and
614 accumulated on the surface allowing the growth of moulds.

615 Though observations were few in number, milk powder in PA/EVOH/PA/PE failed due to
616 discolouration, caking and loss of solubility at the end of the shelf life. This points towards
617 insufficient protection for storage in conditions of high humidity. Meanwhile, milk powder in
618 PE bags (WVTR 3g H₂O/m²/d) did not show any moisture-related signs of failure. However,
619 off-odours were formed due to lipid oxidation in this type of bag. Thus, OTR should certainly
620 be lower than 1464cc O₂/m²/d. The packaging types with drastically lower OTR (0.1 - 1.2cc
621 O₂/m²/d) showed no signs of failure only due to lipid oxidation. Thus, an adequate packaging
622 material should provide an OTR lower than 1464cc O₂/m²/d, but on the basis of these
623 experiments the highest acceptable OTR could not be determined.

624 Conclusions

625 Investigations on cookies were focused on water vapour permeability of the packaging
626 materials. Texture measurements proved a valuable tool in determining shelf life of cookies,
627 despite large variations in the measurements. Texture was not significantly improved by the
628 improved moisture barrier that the reference film (4.2g H₂O/m²/d) provided over the PP film
629 (5g H₂O/m²/d). Thus, recyclability of this packaging system may be improved by choosing the
630 PP film, which is a monomaterial, for a trade-off of a fraction of the shelf life. The tenfold

631 superior moisture barrier of the MPP film (0.4g H₂O/m²/d) did provide an extension of the shelf
632 life based on texture, but isolated failures were still present.

633 Two shortcomings of the current test set-up on milk powders should be pointed out: the limited
634 amount of replications per treatment, and incomplete follow-up of in-package oxygen levels.
635 Missing data on in-package oxygen levels in the current study was due to opaque packages and
636 insufficient package headspace, which interfered with our measurement methods. Anyhow,
637 some important observations can still be made from the current study. If high humidity is to be
638 expected during the storage of milk powder, for example due to export to tropical regions, a
639 tightly sealed bag providing moisture and oxygen barrier is insurmountable. If a stapled or glued
640 bag is used, moisture can easily enter the bag leading to crystallisation of lactose, caking,
641 discolouration and promotion of oxidation, severely limiting the shelf life. Shelf life of milk
642 powder stored in PE bags was cut short only due to formation of off-odours as a result of lipid
643 oxidation. This shows that the water barrier was adequate, but a higher gas barrier was required.
644 PA/EVOH/PA/PE bags also did not provide adequate protection, as the barrier properties of the
645 polar layers were compromised due to high humidity conditions during storage.
646 PE/PA/EVOH/PA/PE bags and aluminum laminate bags both provided superior protection.
647 With material reduction (which can be linked to sustainability and cost-efficiency) in mind, the
648 PE/PA/EVOH/PA/PE film is definitely to be preferred for long-term storage of milk powder in
649 humid environments as it is more material-efficient (56µm less thick) and does not include the
650 aluminum component.

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657 Tables

658 Table 1: Specifications of the cookie packaging films

Material	Thickness (μm)	OTR ^a ($\text{cc}/\text{m}^2/\text{d}$)	WVTR ^b ($\text{g}/\text{m}^2/\text{d}$)
PP (Jindal Films, Virton, Belgium)	25	1307.1 \pm 28.0	5.0
Acrylic – PVdC coated PP (Jindal Films, Virton, Belgium)	26	21.8 \pm 0.3	4.2
Metallized PP (Taghleef GmbH, Holzhausen an der Heide, Germany)	30	31.2 \pm 1.8	0.4

659 ^aMeasured at 23°C, 50%RH at the outside of the material, 0%RH at the inside. Measured with 99.9% O₂ gas flow660 ^bMeasured at 38°C and 90%RH. Data gathered from the material's technical sheet

661 Table 2: Specifications of the milk powder packaging bags

	Thickness (μm)	OTR ($\text{cc}/\text{m}^2/\text{d}$)	WVTR ($\text{g}/\text{m}^2/\text{d}$)
Unsealed PE (Mondi, Poperinge, Belgium)	80	∞	∞
PE (Mondi, Poperinge, Belgium)	80	1464 \pm 43 ^a	3.00 \pm 0.03 ^c
PE/PA/EVOH/PA/PE (Mondi, Poperinge, Belgium)	80	0.25 \pm 0.02 ^a	0.95 \pm 0.01 ^c
PA/EVOH/PA/PE (Obermühle GmbH, Pößneck, Germany)	80	1.24 \pm 0.06 ^a	8.00 \pm 0.23 ^c
BOPP/Al/LDPE (Goglio S.p.A., Milan, Italy)	136	0.1 ^b	0.1 ^d

662 ^aMeasured at 23°C, 50%RH at the outside of the material, 0%RH at the inside. Measured with 99.9% O₂ gas flow663 ^bFrom technical sheet. Measured according to ASTM D3985-95 at 23°C and 0%RH664 ^cMeasured at 38°C, 90%RH at the outside of the material, 0%RH at the inside.665 ^dFrom technical sheet. Measured according to ASTM F1249-90 at 38°C and 90%RH

666 Table 3: Fatty acid composition of the cookies, average of 6 measurements

Fatty acid	Percentage
C14:0	1.0 \pm 0.1 %
C16:0	39.4 \pm 0.7 %
C18:0	6.5 \pm 1.1 %
C18:1	40.5 \pm 1.0 %
C18:2	12.1 \pm 0.2 %

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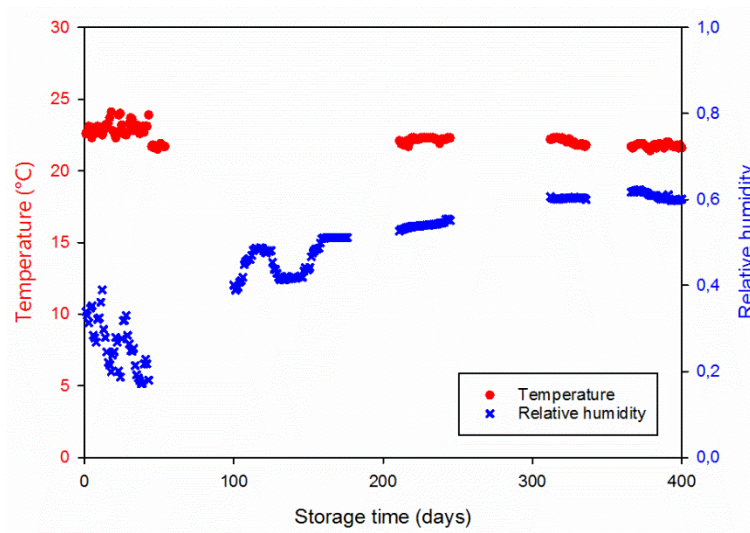
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Table 4: Fatty acid composition of the milk powder, average over 5 measurements

Fatty acid	Percentage	
C4:0	3.8	± 0.2 %
C6:0	3.3	± 0.2 %
C8:0	1.0	± 0.2 %
C10:0	3.4	± 0.1 %
C12:0	4.1	± 0.1 %
C14:0	12.5	± 0.1 %
C14:1	1.1	± 0.1 %
C16:0	33.6	± 0.3 %
C16:1	1.6	± 0.1 %
C18:0	11.1	± 0.2 %
C18:1	22.4	± 0.4 %
C18:2	1.8	± 0.1 %
C18:3	0.6	± 0.1 %

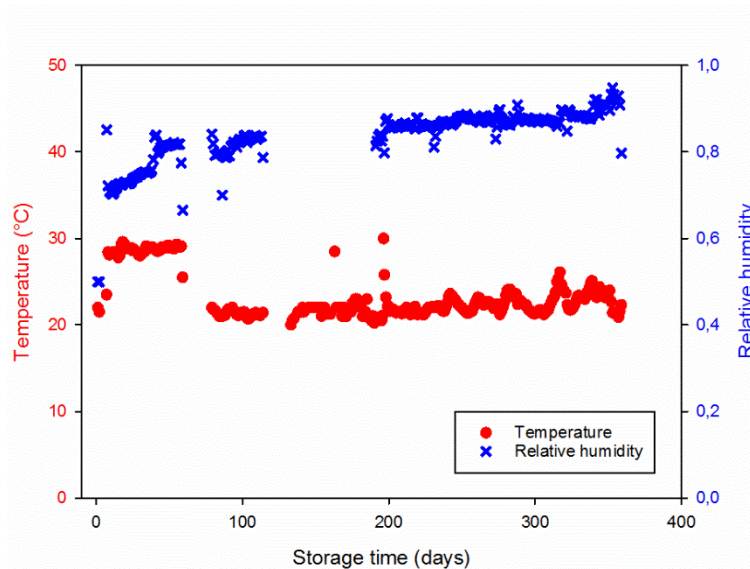
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Figure 16: Temperature (●) and humidity (×) conditions during the cookie storage test



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Figure 17: Temperature and humidity conditions during the milk powder storage test

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