Food Quality and Preference 101 (2022) 104620

Contents lists available at ScienceDirect

Food Quality and Preference

journal homepage: www.elsevier.com/locate/foodqual

I-optimal design of split-plot mixture-process variable experiments: A case study on potato crisps

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ARTICLE INFO

Keywords:
Mixture-process variable experiment
I-optimal design
Potato crisps
Covariates
Pre-specified factor level combinations

ABSTRACT

Designed experiments are powerful tools when developing new products or processes. They allow changing the settings in a systematic way such that a minimal experimental effort results in maximal information. In the food industry, performing designed experiments can be challenging because products and processes are often complex and involve many factors of different kinds. Moreover, some of the factors (for instance, the ingredients of a new formulation) may have intrinsic properties that can be measured but cannot be changed. These are referred to as pre-specified factors or covariates. In this paper, we discuss a representative case study and show how these complications often encountered in the food industry are handled. The goal of the case study is to lower the lipid content of potato crisps by means of a split-plot type of designed experiment involving a complex mixture of ingredients, various constraints on the proportions of the mixture ingredients, and a limited number of batches that each come with a set of covariates. We show how our approach provides insight into the most important factors and allows the product quality to be optimized in an efficient way.

1. Introduction

When developing new products or processes, the optimal combination of factors (inputs) that result in the most desirable response (output) needs to be identified. This combination can be found by iteratively changing one or more inputs and investigating the effect these changes have on the response. While this process is relatively simple when only one or two factors are involved, it rapidly becomes more complex when a larger number of factors are under consideration. The field of Design Of Experiments (DOE) provides an answer to the question how to change the inputs in the most efficient way so that the optimal output can be identified with the least experimental effort (De Ketelaere, Goos, & Brijs, 2011). Since the early work on DOE almost a century ago (Fisher, 1935), the field has advanced considerably so as to be able to accommodate many practical constraints that make the original classical experimental plans unsuited for a large portion of the real-life applications. In general, the state-of-the-art in DOE has moved from the definition of a catalog of rigid, pre-defined experimental designs to the generation of tailored experimental designs that take into account the specificities of the application as much as possible. Examples of such specificities are the experimental budget (how many experimental runs are feasible), blocking factors, the possible presence of constraints that make some combinations of the factors undesired or impossible, and the presence of covariates (Goos & Jones, 2011). This flexibility is enabled by the paradigm of optimal experimental design, which searches for experimental designs consisting of factor combinations that are optimal according to a criterion measuring somehow the information content of the experiment. The most commonly used criteria are the estimation-

Abbreviations: AM, amylose; CW, crisp weight; dm, dry matter; DOE, design of experiments; DP_w, average degree of polymerization on a weight average basis; E-AM, extractable amylose; E-S, extractable starch; LC, lipid content; MC, moisture content; PF, potato flake; PS, average particle size; RVA, Rapid Visco Analyzer; RVA_{CPV}, cold paste viscosity during Rapid Visco Analysis; RVA_{PV}, peak viscosity during Rapid Visco Analysis; SH, stack height.

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https://doi.org/10.1016/j.foodqual.2022.104620
Received 20 December 2021; Received in revised form 13 April 2022; Accepted 30 April 2022
Available online 6 May 2022
0950-3293/

oriented D-optimality criterion and the prediction-oriented I-optimality criterion. D-optimal designs search for factor combinations that maximize the determinant of the information matrix, whereas I-optimal designs seek to minimize the average prediction variance over the design space (Goos & Jones, 2011). Generally speaking, two different design construction approaches can be distinguished, *i.e.* the point exchange and the coordinate exchange algorithm, respectively. The point exchange algorithm starts from a large list of candidate points and tries to improve the chosen optimality criterion by exchanging points in the candidate set (Federov, 1972). The coordinate exchange algorithm (Meyer & Nachtsheim, 1995), conversely, does not require the definition of a candidate set of points but rather starts with a completely random combination of factor level combinations within the defined design space. The algorithm then attempts to improve the level of each factor at each run of the starting design, one by one. When a better value is found for a level of one of the factors in a run, then that level is changed to the better value. This coordinate-wise improvement procedure is continued until none of the individual factor levels can be further improved.

Research in the food industry is often presented with complex experimental constraints. Most often, foodstuffs comprise multi-component recipes where, in many instances, the flexibility in the factor levels that can be used is limited. This particular setting requires mixture experimental designs that take these constraints into account. Mixture designs are characterized by the fact that the sum of all components adds to 1, causing dependence amongst the experimental factors (Cornell, 2002). In this setting, specific experimental designs as well as analysis methodologies are required. The general framework of optimal experimental designs is still applicable here and allows for single or multiple component constraints. From the analysis point of view, specific mixture models have been proposed in literature and are often referred to as Scheffé polynomials (Cornell, 2002). When a recipe not only involves mixture components but also process variables, a mixture-process variable setting is obtained and its analysis is performed in general by crossing a Scheffé-type of model for the mixture components with a classical response surface model for the process variables (Cornell, 2002).

An additional challenge that is often encountered in the food industry is the presence of so-called pre-specified factors, which are also referred to as covariates. These covariates quantify inherent characteristics of a given sample. The experimenter cannot change their values, but does have the ability to measure them prior to the experiment and thus exploit these sample characteristics when designing and analyzing the experiment. Pre-specified factor settings in a DOE context have been described by several authors (Anderson-Cook & Robinson, 2009; De Ketelaere et al., 2011; Goos & Jones, 2011; Nachtsheim, 1989; Sexton, Anthony, Lewis, Please, & Keane, 2006). All of these authors focus on the case where all experimental tests are independent. This requires that the individual samples used in the experiment are all independent. Generally, however, the samples used in an experiment originate from a limited set of batches, and each batch gives rise to multiple observations. Consequently, it is generally unrealistic to assume that the experimental tests are independent. This complication is discussed by Jones and Goos (2015), who describe the similarities between the resulting experimental design problems and split-plot designs and present an algorithm to optimally design experiments involving pre-specified factors in the event multiple samples are obtained from various batches. Their algorithm has been implemented in the JMP statistical software. The case study we present here is the first published real-life application of the methodology developed by Jones and Goos (2015) for experiments with dependent tests due to the use of multiple samples from a limited set of batches. An additional innovative feature of our work is that we apply the methodology in the context of a mixture experiment.

This case study deals with the typical constraints defined above, and relates to 'potato *crisp*' making. In contrast to what is the case for 'potato *chips*', which are produced by deep-frying thin slices of fresh potato (Garayo & Moreira, 2002; Pedreschi & Moyano, 2005), potato crisps are manufactured from dry potato derivatives such as potato flakes (PFs; dry potato particles typically smaller than 500 μm). In the process, PFs are mixed with water and emulsifier into dough that is then sheeted, cut into individual pieces, and deep-fried (Bouchon & Pyle, 2004; Pedreschi, Mariotti, & Cortés, 2016; Reyniers, De Brier, Matthijs, Brijs, & Delcour, 2019). Besides the potato component, cereal and/or legume-based alternatives [starch(es) and/or flour(s)] are frequently part of the recipe (Nath & Chattopadhyay, 2007; Prestini, 2007; Reyniers, De Brier, et al., 2020; Reyniers, Vluymans, et al., 2020; Villagran & Boiano, 2013). It is known that biological- and/or processing-related variations in the properties of the raw materials, especially those amongst different PFs, impact their functionality during crisp making (Reyniers, De Brier, et al., 2020). Therefore, when studying the quality characteristics of potato crisps, it is crucial to incorporate the properties of raw materials in the experimental plan, as pre-specified factor level combinations or covariates.

Table 1
Reference recipe (% *w/w*) for crisp making and constraints on the proportions of the ingredients in the design of 256 samples. The compositions of the 256 dough samples are presented in Supplementary Table S1.

	Reference recipe	Constraints I-optimal design
Dry starchy ingredients	63.9	56.9–69.0
Potato flakes	34.5	22.6–69.0
Wheat starch	9.3	0.0–20.7
Parboiled rice flour	6.7	0.0–34.5
Extruded rice flour	3.2	0.0–34.5
Corn flour	10.2	0.0–34.5
Maltodextrin solution (8.3% *w/w*)	35.0	30.0–40.0
Emulsifier	1.1	1.0–3.1

In recent years, the consumer search for calorie-reduced food products has stimulated research to lower oil uptake during crisp production. While recent efforts to control crisp lipid content (LC) by tailoring PF properties have proven to be successful (Reyniers et al., 2019; Reyniers, Vluymans, et al., 2020), the potential of altering the proportions of the different ingredients in the recipe remains to be explored. In this article, we therefore aim to minimize the LC of crisps by optimizing the proportions of the different ingredients in the recipe using an I-optimal mixture experiment, taking into account the PFs' inherent properties as pre-specified factor level combinations or covariates.

2. Materials and methods

2.1. Recipe overview

Potato crisps are produced starting from a mixture of five components: PFs, wheat starch, parboiled and extruded rice flour and corn flour. We refer to these ingredients as starchy ingredients. They make up 56.9 to 69.0 (% *w/w)* of the total crisp ingredient weight. Besides, the crisp recipes include a maltodextrin solution (30.0 to 40.0% *w/w*) and an emulsifier (1.0 to 3.1% *w/w*). Table 1 presents an overview of these ingredients and related constraints.

2.2. Materials

Commercial samples of PFs were provided by Clarebout (Nieuwkerke, Belgium), Lamb Weston/Meijer (Kruiningen, The Netherlands), Agrarfrost (Wildeshausen, Germany) and Farm Frites (Lommel, Belgium). Wheat starch was from Tereos Syral (Aalst, Belgium) and parboiled and extruded rice flour were from Herba Ingredients (Schoten, Belgium). Corn flour was from Maselis (Roeselare, Belgium). Commercial maltodextrin (Maldex® 190) and (monoacylglycerol and diacylglycerol based) emulsifier were provided by Tereos Syral and Aarhus Karlshamn (Hull, UK), respectively. Sunflower oil was from Cargill

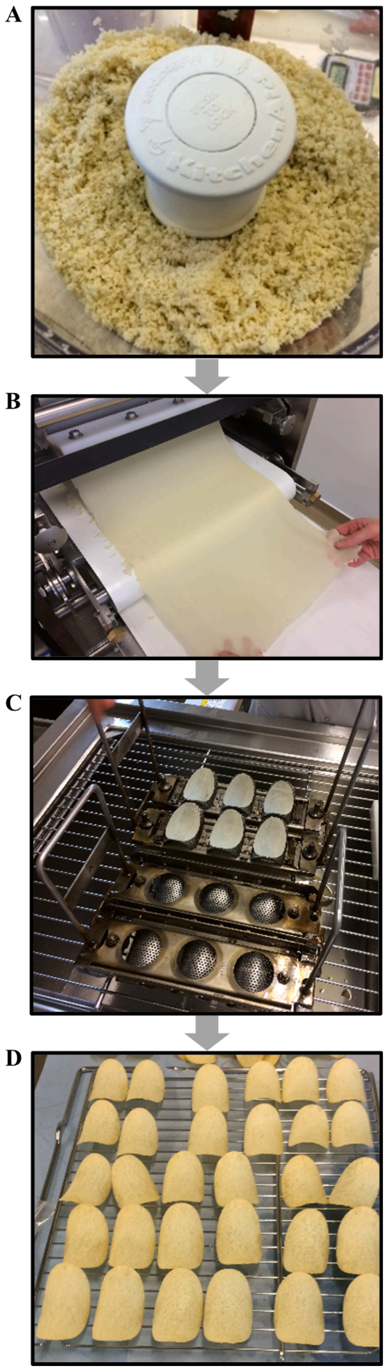

Fig. 1. Production of potato crisps. The dry starchy blend is mixed with maltodextrin solution and emulsifier into a crumbly dough (A) that is immediately sheeted (B). Oval-shaped dough pieces are cut out, placed in a stainless steel mold (C) and finally deep-fried (D) (reprinted from Reyniers et al. 2019, with permission from Elsevier).

(Izegem, Belgium). All chemicals were at least of analytical grade and from VWR International (Leuven, Belgium), unless indicated otherwise.

2.3. Composition and physical properties of starchy ingredients

Moisture (MC) and ash contents were analyzed according to AACC methods 44-15a and 08–12, respectively (AACC-I, 1999). Starch contents were calculated as 0.9 times the glucose contents, which were determined in triplicate by gas chromatography of alditol acetates obtained by prior acid hydrolysis of starch, followed by reduction and acetylation of the resulting glucose units (Courtin, Van den Broeck, & Delcour, 2000). Protein contents were determined by the Dumas combustion method, an automated protein analysis system (EAS, VarioMax N/CN, Elt, Gouda, The Netherlands) in line with AOAC Official Method 990.03 (AOAC, 1995). Dietary fiber contents were analyzed with AOAC Official Method 991.43 (AOAC, 1995). Lipids were extracted with hexane and water-saturated butan-1-ol (Chem-Lab, Zedelgem, Belgium) and quantified gravimetrically after solvent evaporation (Reyniers et al., 2019). Differential scanning calorimetry was executed as described by Bosmans et al. (2012).

The functionality of PFs relies on their extractable starch (E-S) and amylose (E-AM) contents, the chain length distribution of their E-AM, their peak (RVA_{PV}) and cold paste (RVA_{CPV}) viscosities during Rapid Visco Analysis and their average particle sizes (PSs) (Reyniers, De Brier, Matthijs, Brijs, & Delcour, 2018; Reyniers et al., 2019; Reyniers, De Brier, et al., 2020; Reyniers, Vluymans, et al., 2020). These characteristics were evaluated for the different PF samples allowing us to model their impact on the crisps' quality attributes (discussed in Section 2.8). More specifically, the PF E-S and E-AM contents were determined as described by Reyniers et al. (2018), the average degrees of polymerization on a weight average basis (DP_w) of the PF E-AM were calculated from the E-AM chain length distributions essentially as described by Dries, Gomand, Delcour, and Goderis (2016), the RVA_{PV}s and RVA_{CPV}s of the PFs were examined with a Rapid Visco Analyzer (RVA) as described by Reyniers et al. (2018) and the PF PSs were analyzed as described by Reyniers et al. (2018). These characteristics are presented in Supplementary Table S1.

2.4. Production of potato crisps

Potato crisps were produced with an in-house procedure using the following ingredients: PFs (MCs ranging from 4.0 to 9.4%), wheat starch (MC 11.1%), parboiled rice flour (MC 11.8%), extruded rice flour (MC 9.6%), corn flour (MC 9.7%), maltodextrin (MC 6.0%) solution in tap water (8.3% *w/w*) and emulsifier (Reyniers et al., 2019; Reyniers, De Brier, et al., 2020; Reyniers, Vluymans, et al., 2020). Their proportions were varied during the study, taking into account the constraints discussed in Section 2.7. First, the dry starchy ingredients [400.0 g dry matter (dm)] were blended in a Hobart (Troy, OH, USA) N50 5-Quart Mixer for 2 min. The maltodextrin solution was heated to 68 °C. The emulsifier was then added to it and the mixture was homogenized by vigorous stirring. Next, the emulsion was mixed with the starchy blend for 1 min in a Magimix (Montceau-en-Bourgogne, France) 4200 XL into crumbly dough (Fig. 1A), *i.e.* a mass of loose aggregates (<2 mm). The crumbly dough was then sheeted into a continuous dough sheet (Fig. 1B) with a single pair of counter-rotating rolls (Haas-Meincke, Etten-Leur, The Netherlands). The front (2.35 rpm) and back (2.40 rpm) rolls both had a diameter of 40 cm, the roll gap was 80 μm. Individual oval-shaped dough pieces (86 × 50 mm) were placed in a stainless steel mold (40% porosity) (Fig. 1C) and deep-fried for exactly 12 s at 190 °C. After draining (12 s) and cooling to room temperature, crisp (bulk) density was determined from the weight and volume of 40 stacked crisps. The latter was calculated from the dimensions of the metal cutter used to cut the dough sheet and from the height of 40 stacked crisps, which was determined using calipers. Finally, deep-fried crisps (Fig. 1D) were hermetically packed prior to further analysis.

2.5. Specific dough sheet strength

The specific strength of dough sheets was determined as described by Reyniers et al. (2019). Twelve rectangular (82 × 41 mm) pieces were cut from the dough sheets and weighed before determining their thickness using calipers. The strength of the dough sheets was then evaluated using a TA-XT plus Texture Analyzer equipped with the self-tightening roller grips fixture (Stable Micro Systems, Surrey, UK). The initial distance between the grips and the test speed were 35 mm and 10 mm.s^{-1},

Table 2
Minimum and maximum values of the covariates related to the 40 potato flake (PF) batches. E-AM, extractable amylose; E-S, extractable starch; dm, dry matter; DP_w, average degree of polymerization on a weight average basis; RVA_{CPV}, cold paste viscosity during Rapid Visco Analysis; RVA_{PV}, peak viscosity during Rapid Visco Analysis; PS, average particle size.

	Minimum	Maximum
E-AM (AU)	6.6	16.6
E-S (% of dm)	11.7	22.1
DP_W of E-AM (amount of glucose units)	1322	2873
RVA_{CPV} (mPa.s)	543	1099
RVA_{PV} (mPa.s)	655	2322
PS (μm)	241	427

respectively. The maximum load (N) is a measure for the strength of the dough sheet and was corrected for its density (henceforth termed specific strength, and expressed in $N.cm^3.g^{-1}$).

2.6. Crisp quality assessment

The crisp quality was assessed through its LC, its MC, individual crisp weight (CW) as well as the stack height (SH). Crisps were ground in a mortar prior to determining their MC as described in Section 2.3 Lipids were sequentially extracted in triplicate from ground crisps (0.50 g) with hexane and water-saturated butan-1-ol (Chem-Lam, Zedelgem, Belgium) and quantified gravimetrically after solvent evaporation, as described by Reyniers et al. (2019). Individual CW was calculated from the weight of 40 stacked crisps, while SH was defined as the height of 40 stacked crisps, which was determined using calipers (see Section 2.4).

2.7. Experimental constraints

To investigate the impact of the proportions of the different ingredients on crisp quality attributes and to identify the recipe that minimizes the LC, we used an I-optimal experimental design of experiments involving seven mixture ingredients and six covariates. The level of dry starchy ingredients in the dough recipe (x_{dry}) ranged from 56.9 to 69.0% *w/w*, that of maltodextrin solution from 30.0 to 40.0% *w/w* and that of emulsifier from 1.0 to 3.1% *w/w*. Relative to x_{dry}, the level of PFs (x_{PF}) ranged from 40 to 100% *w/w*, that of wheat starch (x_{wheat}) from 0 to 30% *w/w* and that of extruded ($x_{rice,e}$) and parboiled ($x_{rice,p}$) rice flour as well as that of corn flour (x_{corn}) from 0 to 50% *w/w*. Table 1 provides an overview of these constraints on the proportions of the different ingredients taken into account when creating the I-optimal experimental design. In this table, the constraints for the starchy ingredients were expressed relative to the total weight of the dough rather than relative to the total weight of starchy ingredients.

In order to design the experiment, a total of 256 experimental runs was considered feasible. These experimental runs were based on the availability of 40 different batches of PFs, each characterized by 6 covariates (E-AM, E-S, DP_W, RVA_{PV}, RVA_{CPV}, and PS). The minimum and maximum values for these covariates, which served as pre-specified factor level combinations, are presented in Table 2. The compositions of the 256 dough samples are presented in Supplementary Table S1.

2.8. Modelling crisp quality attributes

The model we used for the experimental data involved seven mixture variables (five composing the dry starchy ingredients, maltodextrin solution and emulsifier) and six covariates. Modeling data from experiments that combine mixture variables and non-mixture variables, such as process variables or covariates, can be performed by crossing Scheffé-types of mixture models with response surface models. The main drawback of this approach is that the number of parameters that need to be estimated becomes prohibitively large when the number of mixture components and non-mixture variables increases. To overcome this problem, Kowalski, Cornell, and Vining (2000) suggested to remove all higher order terms in the crossed model (*i.e.* all terms of third order or higher), and replace them with second order interaction terms and pure quadratic terms in the non-mixture variables.

For the general situation involving q mixture components and m non-mixture variables, the model proposed by Kowalski et al. (2000) is given by.

$$\widehat{Y} = \sum_{i=1}^{q}\beta_i x_i + \sum_{i=1}^{q-1}\sum_{j=i+1}^{q}\beta_{ij}x_i x_j + \sum_{i=1}^{q}\sum_{k=1}^{m}\gamma_{ik}x_i p_k + \sum_{k=1}^{m-1}\sum_{l=k+1}^{m}\delta_{kl}p_k p_l + \sum_{k=1}^{m}\delta_{kk}p_k^2 \quad (1)$$

where x_i denotes the proportion of component i in the mixture and p_k represents the value of the k^{th} non-mixture variable.

An additional complication in the potato crisp experiment was that it involved dependent tests and thus correlated responses. Because only 40 batches of PFs were available for performing the 256 experimental runs, multiple runs were carried out using samples from a single batch. The common origin of the runs from one batch implies they are not independent. In order to take the dependence into account, we adopted the approach suggested by Jones and Goos (2015) that allows for multiple yet varying numbers of runs for each batch. In order to do so, we added random block effects to the model in Eq. (1), one for each batch.

In our case study, the non-mixture variables are the six covariates, so that $m = 6$. Since there are seven ingredients in the mixture, $q = 7$.

2.9. Designing the experiment

Taking into account the restrictions imposed on ingredients, the dependence between observations originating from the same batch and the random effects discussed in Section 2.8, a 13-factor I-optimal design was generated using the JMP software (JMP Pro, Version 14, The SAS Institute, Cary, NC, USA). The first seven factors, related to the mixture, were designated as mixture factors, while the last six were designated as covariates (due to the fact that they were pre-specified). To indicate that multiple runs originate from each given batch and therefore yield correlated responses (as in a split-plot experimental design), the six covariates were designated as hard to change in the JMP software. The seven mixture factors had to be designated as easy to change (which is the default for any experimental factor in JMP).

As explained by Jones and Goos (2015), the algorithm that is invoked combines a coordinate exchange procedure to optimize the settings of the experimental factors (the mixture components), and a point exchange procedure for choosing the optimal values of the covariates from the pre-specified list of 40 PF batches. In our work, we used an I-optimal experimental design rather than a D-optimal design, since, for mixture experiments, it is common that I-optimal designs produce much more precise predictions than D-optimal designs (Goos, Jones, & Syafitri, 2016).

In order to include the five constraints from Section 2.7 in JMP, we first wrote the dry starchy fraction x_{dry} as a sum of the individual fractions.

$$x_{dry} = \left(x_{PF} + x_{wheat} + x_{rice,p} + x_{rice,e} + x_{corn}\right). \quad (2)$$

Table 3
Minimum and maximum observed values of the quality attributes [*i.e.* stack height (mm), moisture content (%), weight (g) and lipid content (%) of crisps] in the I-optimal experiment involving 256 samples. The values for the 256 crisp samples are presented in Supplementary Table S1.

	Minimum	Maximum
Stack height	52	140
Moisture content	0.9	10.3
Weight	1.16	4.54
Lipid content	13.0	40.0

This allowed us to express the five constraints as.

$$\begin{aligned} &x_{PF} \geq 0.4\left(x_{PF} + x_{wheat} + x_{rice,p} + x_{rice,e} + x_{corn}\right); \\ &x_{wheat} \leq 0.3\left(x_{PF} + x_{wheat} + x_{rice,p} + x_{rice,e} + x_{corn}\right); \\ &x_{rice,p} \leq 0.5\left(x_{PF} + x_{wheat} + x_{rice,p} + x_{rice,e} + x_{corn}\right); \\ &x_{rice,e} \leq 0.5\left(x_{PF} + x_{wheat} + x_{rice,p} + x_{rice,e} + x_{corn}\right); \\ &x_{corn} \leq 0.5\left(x_{PF} + x_{wheat} + x_{rice,p} + x_{rice,e} + x_{corn}\right). \end{aligned} \tag{3}$$

Finally, bringing all terms in these inequalities to their left hand sides yields the types of linear inequalities that can be entered into JMP:

$$\begin{aligned} &-0.6x_{PF} + 0.4x_{wheat} + 0.4x_{rice,p} + 0.4x_{rice,e} + 0.4x_{corn} \leq 0; \\ &-0.3x_{PF} + 0.7x_{wheat} - 0.3x_{rice,p} - 0.3x_{rice,e} - 0.3x_{corn} \leq 0; \\ &-0.5x_{PF} - 0.5x_{wheat} + 0.5x_{rice,p} - 0.5x_{rice,e} - 0.5x_{corn} \leq 0; \\ &-0.5x_{PF} - 0.5x_{wheat} - 0.5x_{rice,p} + 0.5x_{rice,e} - 0.5x_{corn} \leq 0; \\ &-0.5x_{PF} - 0.5x_{wheat} - 0.5x_{rice,p} - 0.5x_{rice,e} + 0.5x_{corn} \leq 0. \end{aligned} \tag{4}$$

These constraints, in combination with the information about the considered model (Eq. (1)), are used to generate the I-optimal design, using 1000 random starts.

2.10. Model estimation and selection for crisp quality attributes

For each of the quality attributes (*i.e.* CW, SH, MC, and LC), we started by fitting the full random block effects model discussed in Section 2.8, using generalized least squares for the effects of the seven mixture ingredients and the six covariates and restricted maximum likelihood for the variances of the random effects and the error term (Goos & Jones, 2011). Non-significant terms were then removed one by one using a backward elimination procedure and a 5% significance level. A logit transformation was applied for the MC and the LC because these responses were bounded between 0 and 1. For CW and SH, which are bounded by zero only, a logarithmic transformation was used.

2.11. Quality optimization of crisps

As outlined above, the main goal of this study was to determine the proportions of the different ingredients in the recipe that minimize the LC of deep-fried crisps. To ensure that the quality of the crisps with a reduced LC would be comparable to that of crisps made according to a reference recipe (see Table 1), additional constraints were imposed on the other quality attributes:

(i) 95 mm $\leq$ SH $\leq$ 105 mm;
(ii) MC $<$ 2.5%; and
(iii) 2.0 g $\leq$ weight $\leq$ 2.2 g.

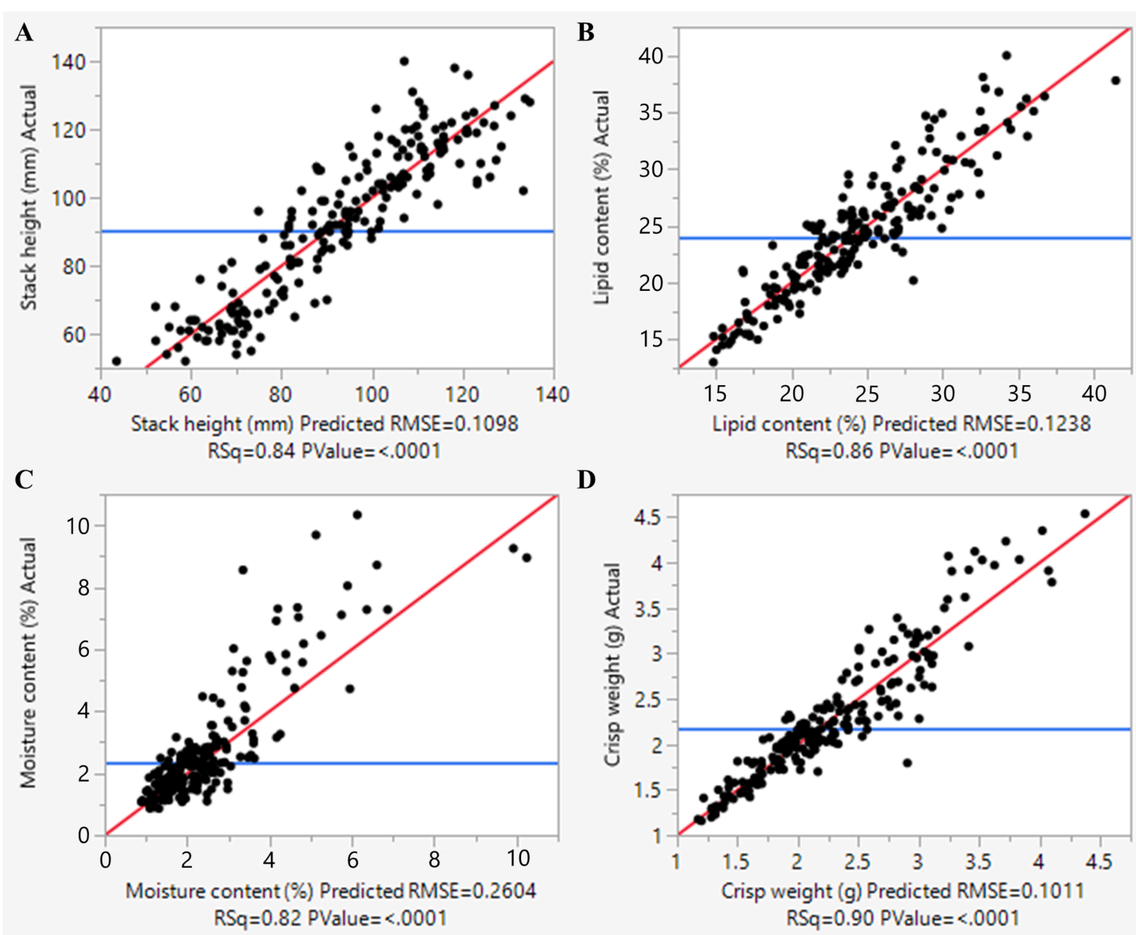

Fig. 2. Actual vs predicted plot for (A) stack height (mm), (B) lipid content (%), (C) moisture content (%), and (D) crisp weight (g).

In order to cope with these four simultaneous optimization goals, we used the desirability function approach of Derringer and Suich (1980), as implemented in JMP. Desirability functions map responses onto the [0,1] interval, where desirabilities close to zero indicate highly undesirable outcomes and desirabilities close to 1 indicate highly desirable outcomes. Afterwards, the individual desirabilities are combined in an overall desirability measure by taking their geometric mean.

2.12. Validation

In order to validate the optimal recipe that was found using the desirability function approach, ten additional experimental runs were executed using five different PF batches. These batches were selected based on a principal components analysis (PCA, based on covariance) (Jolliffe, 2002) performed on the six covariates that described each batch. Four extreme PF batches out of the available 40 were selected, as well as the batch that was closest to the average PF batch. Each of these five PF batches was used in duplicate, yielding ten validation runs for which new measurements were performed and responses were recorded. A simple correlation analysis was then used to check how well the predicted responses (based on the models built before) were aligned with these validation runs.

3. Results and discussion

The observed crisp quality attributes [*i.e.* SH (mm), LC (%), MC (%), and CW (g)] in the experiment varied substantially (Table 3 and Supplementary Table S1). For 47 of the 256 experimental tests, the particular combination of mixture variables and covariates did not result in a cohesive dough sheet and, therefore, did not allow the production of crisps. As a result, it was not possible to record the quality attributes for these samples.

To investigate the factors driving dough making failure, we performed a logistic regression in which the binary response was whether or not the dough was processable (*i.e.* whether or not the crumbly dough could be sheeted into a cohesive dough sheet). The explanatory variables were the ingredients of the dough. Four dough ingredients turned out to have a significant impact on dough processability. More specifically, mixtures having a large proportion of emulsifier in combination with small proportions of water, extruded rice flour and wheat starch lead to unprocessable doughs.

The structure of PF doughs primarily relies on the formation of a continuous starchy [mainly amylose (AM)] network during dough mixing (Reyniers, Ooms, & Delcour, 2020). Water is required for forming such a starch-based gel network (Delcour & Hoseney, 2010). Addition of too low amounts of water during ingredient mixing may limit gel formation and, hence, impede the formation of a cohesive dough sheet. In addition to the evident non-gelatinized nature of native wheat starch, the starch in the extruded rice flour used in this case study was largely non-gelatinized (results not shown). The functional role of gelatinized starch during potato dough making is quite well understood as recently reviewed by Reyniers, Ooms, and Delcour (2020). In contrast, the role of native starch in potato dough making is less understood. In the context of potato doughs, native starch granules have been described as filler particles that are embedded in the polymer network (van der Sman & Broeze, 2013). The prominent impact of the levels of extruded rice flour and wheat starch used in this case study on dough processing is rather unclear and would be an interesting topic for further work. Emulsifiers form single helical complexes with AM (Eliasson & Krog, 1985) and in doing so impact AM gel network formation. Indeed, addition of such complexing agents hinders the formation of AM double helices because the formation of single helical structures is favored (Blazek & Copeland, 2009). The reduced availability of AM for intermolecular hydrogen bonding hinders long distance interactions in the starchy gel, which in turn results in globular aggregates rather than a cohesive structure (Blazek & Copeland, 2009; Richardson, Kidman, Langton, & Hermansson, 2004; Tang & Copeland, 2007). Based on the above, we hypothesize that using high emulsifier levels may well result in dough making failure. As a matter of fact, 23 of the 47 unprocessable doughs were made with the highest emulsifier level evaluated in this designed experiment, *i.e.* 3.1% *w/w* (Supplementary Table S1). This shows that already when added in relatively low amounts emulsifier has a tremendous impact on dough strength and product quality.

Out of the 210 remaining samples, only 11 produced response values that were within the ranges defined in Section 2.11.

3.1. Models for stack height, lipid content, moisture content and weight

3.1.1. Model for stack height

Our final model for the crisps' SH, obtained from the backward model selection procedure, involved 24 terms. A plot of the actual vs predicted SH is given in Fig. 2A. Parameter estimates can be found in Supplementary Table S2. The composition of the dry starchy blend significantly impacted the SH, as did the amounts of emulsifier and maltodextrin solution. According to the model, large proportions of extruded rice flour, wheat starch and maltodextrin solution lead to a large SH, while higher proportions of emulsifier lead to lower SHs. These results can be explained as follows. The crisp SH is directly related to dough expansion during deep-frying. Dough expansion during deep-frying is positively related to its MC. Also, physical starch transformations during deep-frying may contribute to dough expansion. Isolated starch granules rapidly swell and gelatinize at typical deep-frying temperatures (Aguilera, Cadoche, López, & Gutierrez, 2001) to a degree that depends on the MC of the food matrix (Chen et al., 2018). Reyniers, Ooms, and Delcour (2020) already suggested that starch granule swelling as a result of deep-frying induced gelatinization can contribute to dough expansion. Both the native wheat starch and the starch in the extruded rice flour used in this case study were non-gelatinized (*cf. supra*). In line with the reasoning of Reyniers, Ooms, and Delcour (2020), we here hypothesize that gelatinization of wheat and rice starch and the accompanying swelling of the starch granules during deep-frying added to the expansion caused by water evaporation. Emulsifiers form complexes with AM (*cf. supra*) and in doing so impact AM gel network formation (Villagran & Wooten, 2003) such that the dough sheet is weakened. Fitting our random block effects model to the specific dough sheet strength (results not shown) indeed showed that the emulsifier had a significant strongly negative impact on the specific strength of dough sheets. Reyniers, Vluymans et al. (2020) showed that weakening the dough structure (by partially hydrolyzing the starch in the network by amylase) to a certain extent allowed more expansion. However, too intense dough weakening caused the dough structure to collapse resulting in strongly reduced expansion (Reyniers, Vluymans, et al., 2020). Similarly, high levels of emulsifier most likely weakened the dough sheet and caused structure collapse during deep-frying resulting in dense crisps with a low SH. All covariates were found to be significant, with the E-S being most important, though their role was smaller when compared to the composition of the starchy blend.

3.1.2. Model for lipid content

After applying the backward elimination procedure, 29 significant model term were retained. A plot of the actual vs predicted LC is given in Fig. 2B. Parameter estimates can be found in Supplementary Table S2. As with the SH, the proportion of maltodextrin solution as well as the composition of the dry starchy blend significantly affected the crisps' LC. More specifically, doughs containing large proportions of maltodextrin solution, and thus having a high water content, resulted in crisps with a high LC. During deep-frying, intensive water evaporation results in dough expansion (see Section 3.1.1), which in turn allows oil penetration into the product. It is generally accepted that increased water evaporation results in higher levels of oil uptake (Mellema, 2003; Ziaiifar, Achir, Courtois, Trezzani, & Trystram, 2008). In addition, the proportion of PFs was the most important factor in the composition of

Table 4
Reference and optimal recipes taking into account the constraints defined in Section 2.11. Quality attributes of crisps made from potato flake sample 9 using the reference recipe and optimal recipe, respectively, are also presented.

	Reference recipe	Optimal recipe
Dry starchy ingredients	63.9	64.9
Potato flakes	34.5	30.4
Wheat starch	9.3	7.9
Parboiled rice flour	6.7	7.4
Extruded rice flour	3.2	14.1
Corn flour	10.2	5.1
Maltodextrin solution (8.3% *w/w*)	35.0	33.7
Emulsifier	1.1	1.5
Lipid content (%)	25.1	22.9
Stack height (mm)	99	95
Moisture content (%)	1.9	2.5
Weight (g)	2.05	2.22

the dry starchy blend, followed by the fraction of extruded rice flour. In general, high amounts of PFs resulted in crisps with a low LC. The PF specific characteristics (*i.e.* the covariates linked to the individual PF batches) all played a significant role with E-S and RVA_C being most influential.

3.1.3. Model for moisture content

The MC of the crisps was described using 23 significant model terms. A plot of the actual vs predicted MC is given in Fig. 2C. Parameter estimates can be found in Supplementary Table S2. The emulsifier content played a prominent role, as well as the composition of the dry starchy fraction. The strength of the starchy gel network in the dough sheet determines its water holding capacity and, as a result, the rate of water evaporation during deep-frying (Reyniers, De Brier, et al., 2020). Here, the ratios of the different starchy ingredients and the level of emulsifier clearly affected the strength of the dough sheet, which in turn determined its water holding capacity. Stronger dough sheets have a higher water holding capacity which in turn slows down water evaporation. This can result in crisps with a higher MC if water is retained in the dough to such an extent that substantial amounts remain after deep-frying. As a matter of fact, a plot of crisp MC vs specific dough sheet strength (data can be found in Supplementary Table S1) supports the above hypothesis as it shows that stronger dough sheets generally resulted in higher moisture levels remaining in the deep-fried crisps. Remarkably, the level of maltodextrin solution and, hence, the initial water content of the dough was a significant though not dominant factor. This suggests that the water binding by the (starchy) ingredients in the dough sheet rather than its content determines its rate of evaporation during deep-frying. All covariates significantly contributed to the model, but their role was minor.

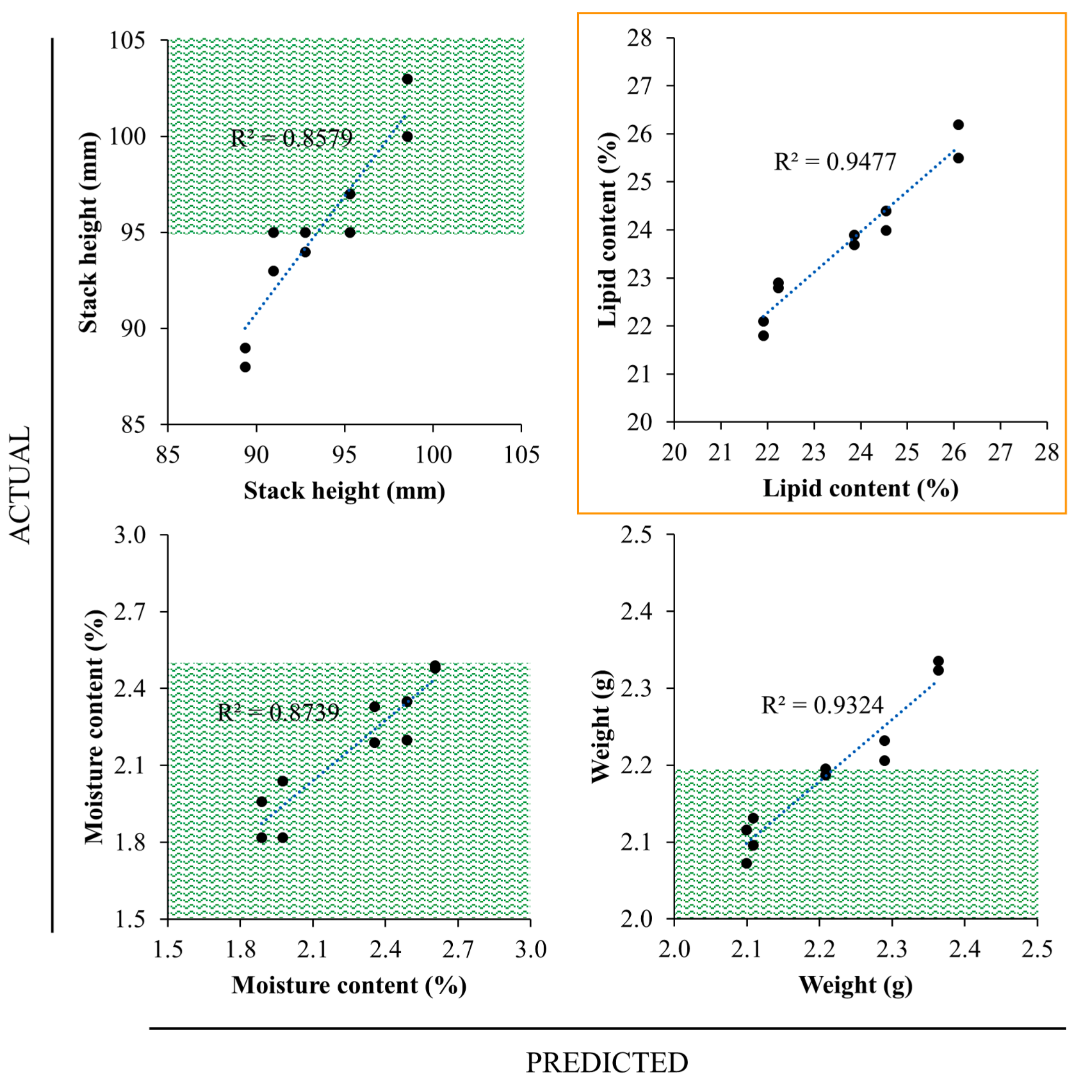

Fig. 3. Validation of the models. Actual by predicted plots for the four quality attributes based on ten confirmatory tests. The shaded areas indicate the acceptable ranges for the stack height, moisture content and weight.

3.1.4. Model for weight

The CW model included 28 model terms. A plot of the actual vs predicted CW is given in Fig. 2D. Parameter estimates can be found in Supplementary Table S2. Increasing the levels of emulsifier and maltodextrin solution resulted in a lower CW. While emulsifier forms AM-lipid complexes (see Section 3.1.1), water acts as a plasticizer (van der Sman & Broeze, 2013). As they both moderate the viscoelastic properties of the dough, they increased its sheeting ability resulting in thinner dough sheets and, hence, crisps with a lower weight. The composition of the dry starchy fraction was a main driver as well, with increasing wheat starch or parboiled rice flour resulting in a higher CW. The covariates only played a minor role for the CW.

3.2. Quality optimization

With the different models in place, we aimed at optimizing the quality of the crisps and searched for a recipe that minimized the LC whilst still adhering to the constraints for SH, MC and CW using the desirability approach (see Section 2.11). A recipe adhering to these constraints is given in Table 4. Compared to the reference recipe, that recipe resulted in a reduction of the crisps' LC by about 2%, for any given PF batch.

3.3. Model validation

The models reported in Sections 3.1.1 through 3.1.4 and the new recipe mentioned in Section 3.2 were validated by means of ten validation runs (Supplementary Table S3) selected using the approach mentioned in Section 2.12. Fig. 3 shows the actual vs predicted values for these runs. Since all ten runs were based on the same optimal recipe (Table 4), the difference in measured responses is purely due to the effect of the covariates and random variation. The large spread in the responses from the validation runs highlights the importance of the batch-related covariates when optimizing crisp quality. Overall, the correlations we obtained between the predicted and observed responses of the validation runs were high, indicating that our final models for the four quality attributes adequately described the underlying processes.

4. Conclusions

In this paper, we described a practical case study concerning the production of potato crisps where a designed experiment was constructed to optimize product quality. The study involved a complex mixture with five linear constraints on the ingredient proportions and the practical limitation of having a limited stock of samples to start from. Moreover, each of these samples was characterized by a number of prespecified characteristics. Recently, Jones and Goos (2015) presented the methodology required to deal with such a complex yet practically highly relevant situation. To the best of our knowledge, our article presents the first real-life application of that methodology. We showed how the experimental and modeling approach provides the necessary insight in the formulation to optimize product quality, which was defined through multiple characteristics. As a conclusion, the recent advancements of the field of optimal experimental design allow food scientists to handle complex practical situations, and achieve new product development more efficiently.

CRediT authorship contribution statement

S. Reyniers: Conceptualization, Data curation, Investigation, Methodology, Writing – original draft, Writing – review & editing. **N. De Brier:** Conceptualization, Investigation, Methodology, Writing – review & editing. **K. Brijs:** Writing – review & editing. **B. De Ketelaere:** Conceptualization, Data curation, Investigation, Methodology, Writing – original draft, Writing – review & editing. **W. Akkermans:** Data curation, Investigation, Methodology, Writing – review & editing. **S. Matthijs:** Conceptualization, Investigation, Methodology, Writing – review & editing. **J.A. Delcour:** Writing – review & editing. **P. Goos:** Conceptualization, Investigation, Methodology, Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

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

Acknowledgements

We thank the Agentschap Innoveren & Ondernemen - Vlaanderen (VLAIO) (Brussels, Belgium) and Wimble Services Belgium (Mechelen, Belgium) for their financial support. We are also grateful to N. Schoonens, K. Oosterlinck and J. Coppens for their excellent technical assistance. B. De Ketelaere and K. Brijs acknowledge the Industrial Research Fund (KU Leuven, Leuven, Belgium) for their position as industrial research manager. This research is part of the Methusalem program "Food for the Future" (2007 to 2021) at KU Leuven.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodqual.2022.104620.

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