Optimal designs for multi-response generalized linear models with applications in thermal spraying

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Abstract

We consider the problem of designing experiments for investigating particle in-flight properties in thermal spraying. Observations are available on an extensive design for an initial day and thereafter in limited number for any particular day. Generalized linear models including additional day effects are used for analyzing the process, where the models vary with respect to different responses. We construct robust D-optimal designs to collect additional data on any current day, which are efficient for the estimation of the parameters in all models under consideration. These designs improve a reference fractional factorial design substantially. We also investigate designs, which maximize the power of the test for an additional day effect. The results are used to design additional experiments of the thermal spraying process and a comparison of the statistical analysis based on a reference design as well as on a selected D-optimal design is performed.

Keywords and Phrases: Generalized linear models, day effects, optimal designs, thermal spraying

1 Introduction

Response surface methodology is a widely used tool to analyze the influence of experimental conditions on a response by an adequate selection of a design and subsequent fitting of a model. It is nowadays used in a variety of applications, such as physics, chemistry, biology or engineering to name just a few. The precision of the estimates can be substantially improved by the choice of an experimental design and numerous designs which improve the statistical accuracy have been derived for the standard linear model [see Myers and Montgomery (1995), Khuri and Cornell (1996)]. Most of the literature refers to the situation, where it is assumed that the data generating process does not change during the experiment, however, there are many situations where this assumption may not be reasonable. We recently encountered such a situation in the context of thermal spraying, where experiments are conducted at different days and the process is highly influenced by latent day specific effects such as temperature or humidity. This specific application is described in detail in Section 2, but similar problems appear frequently in industrial practice, whenever some latent variables change because experiments are conducted at different days. A response surface model is estimated on the basis of the available data from the first day. The experiment is continued at another day where a limited number of additional experiments can be performed. In order to address the problem of different experimental conditions additional day effects are included in the model. While the design of experiment for the initial day can be obtained from standard methodology, we are interested in an optimal design of experiment for the necessary additional experimental runs.

Linear models with continuous (quite often, normally distributed) responses as assumed in standard response surface methodology are inappropriate in the context of thermal spraying and it is demonstrated in Tillmann et al. (2012) that generalized linear models turn out to be more suitable for describing the in-flight properties in thermal spraying. This class of models contains the "classical" approach as special case and also provides models for situations in which the response is not necessarily normal, but follows a distribution of an exponential family where the mean is modeled as a function of the predictor. Unlike the linear regression case, optimal designs then may depend on the unknown parameter value as well as the specifically chosen model components. So far, optimal designs for this situation are rarely treated in the literature and if they are mostly with an emphasis on binary or Poisson response variables. Khuri et al. (2006) give a very nice review of the most common approaches to handle the so-called design dependency problem, namely locally optimal designs, sequential designs, Bayesian designs and quantile dispersion graphs. Woods et al. (2006) develop a "compromise" design selection criterion that takes uncertainties in the parameters as well as in the link function and the predictor into account by averaging over a chosen parameter and model space. With regard to this generation of "compromise" designs Dror and Steinberg (2006) present a heuristic using K-means clustering over local D-optimal designs that is robust against the mentioned uncertainties.

The design problem investigated in this paper differs from the problems discussed in the literature in several perspectives. Firstly, the response in the thermal spraying process is multivariate, while the literature usually discusses designs for a univariate response. Secondly, we investigate the situation where a part of the data has been already observed on an initial day and a

design is required for collecting additional data on any current day, which has good properties to estimate the parameters in the presence of a likely day-effect, describing the difference in the spraying between two days. Hence, model selection for each component of the response can be performed on the basis of the initial design, but a compromise design has to be found for the models corresponding to the different components of the response, which additionally addresses the problem of uncertainty with respect to the model parameters. While we use the D-optimality criterion for determining an efficient design for estimating all parameters in the models including the additional day effect, this criterion might not be appropriate for the purpose of detecting differences between days. Therefore we also consider alternative criteria which are particulary designed for model discrimination.
The remaining part of the paper is organized as follows. In Section 2 we give an introduction to the problem of thermal spraying and motivate the application of generalized linear models (GLM) in this context. For the sake of transparency, we concentrate on Gamma-distributed responses and avoid most of the general notation of GLM. Section 3 is devoted to optimal design problems and we discuss locally, multi-objective or compromise designs and optimal designs for identifying an additional day effect. In Section 4 we return to the problem of designing additional experiments for the analysis of the in-flight properties in the thermal spraying problem. In particular, we demonstrate that a reference design can be substantially improved with respect to its efficiency of estimating all parameters while moderate improvements can be achieved for testing for an additional day effect. We also develop designs with good efficiencies for both purposes.
The results are illustrated by designing real new thermal spraying experiments on a different day. In particular, by performing four additional experiments under a reference and a Bayesian D-optimal design, respectively, it is demonstrated that the Bayesian D-optimal design improves the reference design with respect to the determinant criterion for all investigated in-flight properties. Finally, optimal designs for particular models and additional material are presented in an entire Appendix.

2 Statistical modeling of thermal spraying

Thermal spraying technology is widely used in industry to apply coatings on surfaces, aiming e.g. at better wear protection or durable medical instruments. However, due to uncontrollable factors thermal spraying processes are often lacking in reproducibility, especially if the same process is repeated on different days. Furthermore an immediate analysis of the coating quality is usually not feasible as it requires time and results in destruction. A solution to this problem possibly lies in measuring properties of particles in flight based on the assumption that they carry the needed information of uncontrollable day effects [Tillmann et al. (2010)]. We next introduce the analysed thermal spraying process and then the applied class of generalized linear models.

2.1 Thermal spraying

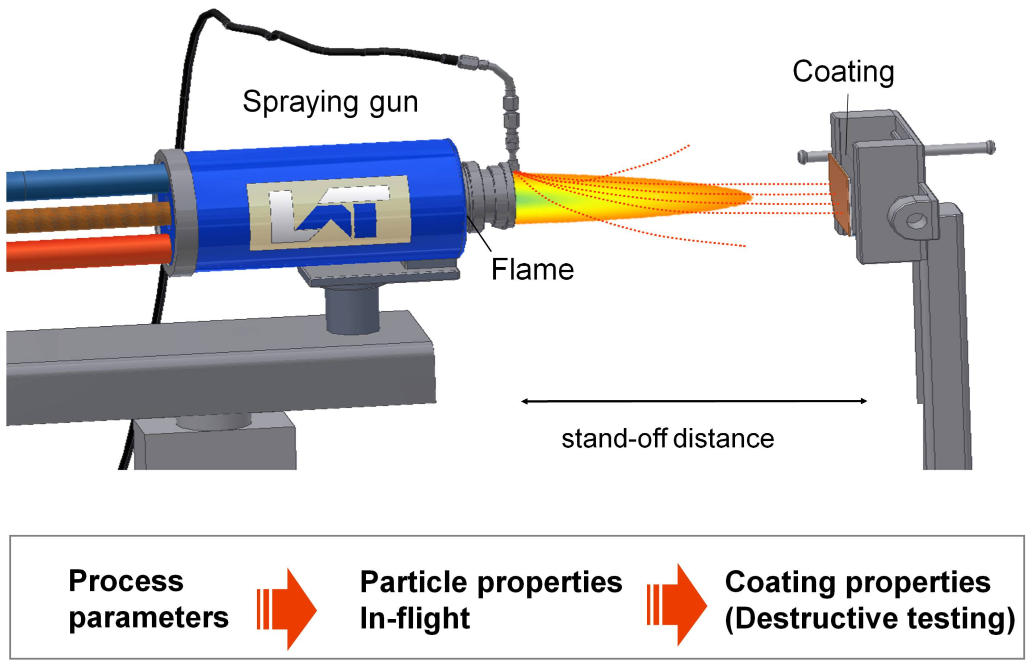

Figure 2.1: *Thermal spraying process*

As application a HVOF (high-velocity oxygen-fuel spray) spraying process is regarded where WC-Co powder is melted and at high-speed applied to a surface by a spraying gun. The influence of process parameters on in-flight properties of the coating powder is of interest. Figure 2.1 depicts the thermal spraying process. Preliminary screening experiments [Tillmann et al. (2010)] identify four relevant process parameters: The amount of kerosine (K) in liter per hour used, the ratio lambda of kerosine to oxygen (L) and the feeder disc velocity (FDV) as well as the stand-off-distance (D). The last parameter describes the distance from the spraying gun to the component which is coated and thereby also to the device measuring properties of the particles in-flight. The device measures the temperature and velocity of properties in-flight as well as flame width and flame intensity. The considered process parameters and in-flight properties are summarized in Table 2.1. Summary statistics of the in-flight measurements provide responses

process parameters	in-flight properties
stand-off-distance (D)	temperature
amount of kerosine (K)	velocity
ratio of kerosine to oxygen (L)	flame width
feeder disc velocity (FDV)	flame intensity

Table 2.1: *Process parameters and in-flight properties in thermal spraying*

which have successfully been modeled by generalized linear models with Gamma distribution and different link functions based on central composite designs [Tillmann et al. (2012); Rehage et al. (2012)]. To capture the effect of unobservable day specific influences, e.g. created by room temperature and moisture, day effects have been added to the linear predictor of the models [Tillmann et al. (2012); Rehage et al. (2012)]. These effects have to be estimated from few additional experiments on any current day. Therefore it is of high interest to determine optimal experimental designs for this specific task.

2.2 Measuring information in generalized linear models

In this section we give some background on generalized linear models which are used to model the thermal spraying process. As usually, we denote the real valued response by Y and the predictor by a q-dimensional variable x. In the application Y presents either the temperature, velocity, flame width or the flame intensity, while the predictor is a three- or four-dimensional variable containing some combination of the machine parameters stand-off-distance, amount of kerosine, ratio of kerosine to oxygen and feeder disc velocity.
Let (Y_i, x_i), $i = 1, \cdots, n$, be a sample of observations where $x_i = (x_{1i}, \cdots, x_{qi})^T \in \mathbb{R}^q$ are explanatory variables and $Y_i \in \mathbb{R}$ is the response at experimental condition x_i $(i = 1, \ldots, n)$. In contrast to linear models the response described by a generalized linear model may follow a distribution of the exponential family. Tillmann et al. (2012) and Rehage et al. (2012) showed that the in-flight properties in the thermal spraying application can be adequately modeled by generalized linear models with Gamma distributed response. These models are defined by the density

$$f(y|x, \beta) = \frac{1}{\Gamma(\nu)} \left(\frac{\nu}{\mu}\right)^{\nu} y^{\nu-1} e^{-\frac{\nu}{\mu} y}, \quad y \geq 0, \tag{2.1}$$

and mean

$$\mu = E(Y|x) = g^{-1}\left(z^T \beta\right) \tag{2.2}$$

where $g(\cdot)$ is an appropriate (known) link function, $z = z(x) \in \mathbb{R}^p$ is a vector of regression functions depending on the explanatory variables x, $\beta \in \mathbb{R}^p$ denotes an unknown parameter vector and $\mu > 0$ and $\nu > 0$ denote the mean and shape parameter, respectively [see Fahrmeir and Tutz (2001)]. Common link functions for the Gamma distribution include the identity $g(\mu) = \mu$, the canonical link $g(\mu) = -1/\mu$ and the log link $g(\mu) = \log(\mu)$. For the first two link functions restrictions regarding the parameter β have to be made such that the conditional expectation μ is non-negative.
If n independent observations at experimental conditions $x_1, \ldots, x_n$ are available and the inverse of the link function g^{-1} is twice continuously differentiable, it follows by a straightforward calculation that the Fisher information matrix for the parameter β is given by

$$I(\beta) = \nu^2 \sum_{i=1}^{n} w(z_i^T \beta) z_i z_i^T \in \mathbb{R}^{p \times p}, \tag{2.3}$$

where the weight function is defined by

$$w(\mu) = ((\log g^{-1}(\mu))')^2 = \frac{1}{(g'(g^{-1}(\mu))g^{-1}(\mu))^2}.$$

The covariance matrix of the maximum likelihood estimator for the parameter β can be approximated by the inverse of the information matrix $I(\beta)$. Note that for the different link functions the corresponding information matrices differ only with respect to the weight $w(\mu)$, and the weights corresponding to the Gamma distribution for the named link functions are shown in Table 2.2.

link function $g(\cdot)$	weight in (2.3)
$g(\mu) = \mu$	$1/(z_i^T\beta)^2$
$g(\mu) = 1/\mu$	$1/(z_i^T\beta)^2$
$g(\mu) = \log(\mu)$	1

Table 2.2: *Weights in the information matrix* (2.3) *for the Gamma distribution with identity, canonical and log link*

In each case the information matrix depends on the sample size n, the link function g, the vector of regression functions $z(x)$ and especially on the parameter β. Throughout this paper we consider a quadratic response function for $g(E[Y|x])$, that is

$$z^T\beta = \beta_0 + \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q} \sum_{j\geq i}^{q} \beta_{ij} x_i x_j. \tag{2.4}$$

3 Optimal designs for generalized linear models

Optimal designs maximize a functional, say Φ, of the Fisher information matrix with respect to the choice of the experimental conditions $x_1, \ldots, x_n$, and numerous criteria have been proposed in the literature to discriminate between competing designs [see Pukelsheim (2006)]. The commonly used optimality criteria (such as the D-, A- or E-optimality criterion) are positively homogenous, that is $\Phi(\lambda I(\beta)) = \lambda\Phi(I(\beta))$ whenever $\lambda \geq 0$ [see Pukelsheim (2006)]. Consequently, an optimal design maximizing a functional of the Fisher information matrix (2.3) will not depend on the parameter ν, but it will depend on the parameter β. Designs depending on unknown parameters of the model are called locally optimal designs and were at first discussed by Chernoff (1953). Since this fundamental paper many authors have worked in the construction of locally optimal designs. We refer to some recent work in this direction by Yang and Stufken (2009), Yang (2010a) and Dette and Melas (2011), who discuss admissible classes of locally optimal designs for nonlinear regression models with a one-dimensional predictor.

In situations where preliminary knowledge regarding the unknown parameters of a nonlinear model is available, the application of locally optimal designs is well justified. A typical example are phase II clinical dose finding trials, where some useful knowledge is already available from phase I [see Dette et al. (2008)]. A further example is given by the thermal spraying problem introduced in Section 2. Here a couple of experiments were already performed on the basis of a central composite design, and new experiments have to be planned for further investigations. On the basis of the available observations parameter estimates and standard deviations are available, which can be used in the corresponding local optimality criteria. Locally D-optimal designs for the generalized linear model introduced by (2.1), (2.2), (2.4) will be defined in Section 3.1 and discussed in 4.1.

On the other hand, locally optimal designs are often used as benchmarks for commonly proposed designs (see also the discussion in Section 4). Moreover, they are the basis for more sophisticated design strategies, which require less precise knowledge about the model parameters, such as sequential, Bayesian or standardized maximin optimality criteria [see Pronzato and Walter (1985), Chaloner and Verdinelli (1995) and Dette (1997) among others]. Optimal designs with respect to the latter criteria are called robust designs and will be discussed in Section 3.2. Finally, optimal designs for investigating the existence of an additional day effect are introduced in Section 3.3.

3.1 Locally D-Optimal designs

As Myers et al. (2002) point out, the D-optimality criterion is a commonly used design selection criterion especially for industrial experiments. To be precise, consider a link function g and a regression model of the form (2.4) defined with corresponding vector $z = z(x)$ and parameter β. We collect the model information in the vector $s = (g, z, \beta)$. In order to reflect the dependency of the Fisher information matrix in (2.3) on the design and on the particular model specified by the link function g and corresponding parameter β we introduce the notation

$$I(\mathbf{X}, s) = \sum_{i=1}^{n} w(z_i, \beta) z_i z_i^{'} \qquad (3.1)$$

for the Fisher information matrix, where $\mathbf{X} = (x_1, \cdots, x_n)$ denotes the design and $z_i = z(x_i)$ $(i = 1, \ldots, n)$. Following Chernoff (1953) we call a design $\mathbf{X}_s^*$ locally D-optimal if it maximizes the determinant of the Fisher information matrix

$$\Phi_D(\mathbf{X}|s) = |I(\mathbf{X}, s)|^{1/p(s)}, \qquad (3.2)$$

where $p(s)$ denotes the number of parameters in model s. Note that the locally D-optimal design depends on the link function g, the model z and the corresponding unknown parameter vector β, which justifies our notation $\mathbf{X}_s^*$ $(s = (g, z, \beta))$. It is usually assumed that information regarding the unknown parameter in a specific fixed model is available [see for example Ford et al. (1992), Biedermann et al. (2006b), Fang and Hedayat (2008), Dette et al. (2010) among many others]. Locally D-optimal designs (and other optimal designs with respect to local optimality criteria) have been criticized because of their dependence on the specific choice of the parameter β. However, there are numerous situations where preliminary knowledge regarding the unknown parameters is available, such that the application of locally optimal designs is well justified (see the discussion at the beginning of this section). A further common criticism of the criterion (3.2) is that it requires the specification of the model and the link function and there are several situations where a design for a specific model is not efficient for an alternative competing model [see Dette et al. (2008)]. For example in the case of thermal

spraying, different models turn out to be appropriate for analyzing the temperature, velocity, flame width and intensity but data can only be collected according to one design. In Section 4.1 we demonstrate that a locally D-optimal design for analyzing one particular in-flight property might be inefficient for analyzing another one.
In the following sections we briefly discuss different approaches to find D-optimal designs which are less sensitive with respect to a misspecification of link, model and parameter vector β.

3.2 Multi-objective designs

The problem of addressing model uncertainty (with respect to the form of the regression function or prior information regarding the unknown parameter) has a long history. Läuter (1974a) proposed a criterion which is based on a product of the determinants of the information matrices in the various models under consideration and yields designs which are efficient for a class of given models. Lau and Studden (1985) and Dette (1990) determine optimal designs with respect to Läuter's criterion for a class of trigonometric and polynomial regression models, respectively. In the case where the form of the model is fixed and there is uncertainty about the nonlinear parameter Läuter (1974b) and Chaloner and Larntz (1989) propose a Bayesian D-optimality criterion which maximizes an expected value of the D-optimality criterion with respect to a prior distribution for the unknown parameter [see also Pronzato and Walter (1985), who call the corresponding designs robust designs, or Chaloner and Verdinelli (1995) for comprehensive reviews of this approach]. Since its introduction Bayesian optimal designs have found considerable attention in the literature [see Haines (1995), Mukhopadhyaya and Haines (1995), Dette and Neugebauer (1997), Han and Chaloner (2004) among others]. Biedermann et al. (2006a) determined efficient designs for binary response models, when there is uncertainty about the form of the link function (e.g. Probit or Logit model) and the parameters. Recently, Woods et al. (2006) used this approach for finding D-optimal designs in the case of uncertainty concerning the parameter vector β as well as the linear predictor $\eta = z^{'}\beta$ and the link function $g(\cdot)$. For this purpose these authors propose a multi-objective criterion [see Cook and Wong (1994)] for the selection of a design. Most of the optimality criteria in these references are based on the average of given optimality criteria $\Phi(\mathbf{X}|s)$ (such as the D-optimality criterion) over the space $\mathcal{M}$ of the possible models, which takes the model uncertainty into account. In the present context the elements of the set $\mathcal{M}$ are of the form $s = (g, z, \beta)$ corresponding to uncertainty with respect to the link function g, the regression function $z = z(x)$ and the parameter β.
To be precise, let $\mathcal{G}$ denote a class of possible link functions. For each $g \in \mathcal{G}$ let $\mathcal{N}_g$ denote a class of vector-valued functions $z(x)$ and finally define for each pair (g, z) with $z \in \mathcal{N}_g$ a parameter space $\mathcal{B}_{g,z}$. With $\mathcal{M} = \{(g, z, \beta) : g \in \mathcal{G}, z \in \mathcal{N}_g, \beta \in \mathcal{B}_{g,z}\}$ the robust optimality criterion is given by

$$\Phi_B(\mathbf{X}|\mathcal{M}) = \int_{\mathcal{M}} \text{eff}_D(\mathbf{X}|s) dh_1(\beta|g, z) dh_2(z|g) dh_3(g), \qquad (3.3)$$

where the efficiency is defined by

$$\text{eff}_D(\mathbf{X}|s) = \frac{\Phi_D(\mathbf{X}|s)}{\Phi_D(\mathbf{X}_s^*|s)}, \tag{3.4}$$

$\mathbf{X}_s^*$ is the locally D-optimal design for model $s \in \mathcal{M}$ and h_1, h_2 and h_3 represent cumulative distribution functions reflecting the importance of the particular constellation (g, z, β).
As an alternative to the Bayesian criterion Dette (1997) proposed a standardized maximin D-optimality criterion, which determines a design maximizing the worst efficiency over the class $\mathcal{M}$ of possible models [see also Müller and Pázman (1998)]. Since its introduction this criterion has found considerable attention in the literature. To be precise, assume that $\mathcal{M}$ is a set of possible values $s = (g, z, \beta)$ for the link function, model and parameter vector and recall the definition of the relative efficiency of the design $\mathbf{X}$ with respect to the locally optimal design $\mathbf{X}_s^*$ defined by (3.4). The standardized maximin optimal design $\mathbf{X}^*$ is defined as the solution of the optimization problem

$$\max_{\mathbf{X}} \min_{s \in \mathcal{M}} \text{eff}_D\left(\mathbf{X}|s\right).$$

Therefore this design maximizes the minimal relative efficiency calculated over the set $\mathcal{M}$, and it can be expected that such a design has reasonable efficiency for any choice of the parameter $s \in \mathcal{M}$.
Standardized maximin optimal designs are extremely difficult to find and for this reason we will mainly consider optimal designs with respect to the Bayesian-type criterion (3.3). Some explicit results for models with a one-dimensional predictor can be found in Imhof (2001) as well as Dette et al. (2007).

3.3 Design criteria in the presence of an additional day-effect

Recall the motivating example discussed at the end of Section 2, where observations are taken at two different days. In order to address this situation in the generalized linear model we replace the regression model $z(x)$ and the parameter β in (2.2) by the vectors

$$z^*(x, t) = (z(x)^T, t)^T \ ; \quad \beta^* = (\beta^T, \gamma)^T$$

respectively, where the parameter t can attain the values 0 and 1 corresponding to different experimental conditions caused by a possible day effect. Thus the expected response at a particular experimental condition satisfies

$$g(E[Y|x]) = \begin{cases} z^T(x)\beta & \text{if} \quad t = 0 \\ \gamma + z^T(x)\beta & \text{if} \quad t = 1. \end{cases} \tag{3.5}$$

We assume that n observations are taken at the initial day at experimental conditions $x_1, \dots, x_n$. This corresponds to the choice $t = 0$ and a generalized linear model without the day effect γ is fitted to the data. Additional experiments can be made at any further day at experimental conditions $x_{n+1}, \dots, x_{n+m}$ which corresponds to the choice $t = 1$. The Fisher information for a specific model, weight function (corresponding to the generalized linear model) and parameter is then given by

$$I(\mathbf{X}, s) = \sum_{i=1}^{n+m} w(z_i^{*T}\beta^*) z_i^* z_i^{*T} \in \mathbb{R}^{p(s)+1 \times p(s)+1} \tag{3.6}$$

where $z_i^* = z(x_i, t_i)$ denotes the vector of regression functions corresponding to the i-th observation $(i = 1, \ldots, n+m)$ and the weight function is defined by

$$\frac{1}{\left(z_i^{*T}\beta^*\right)^2}\,,\quad \frac{1}{\left(z_i^{*T}\beta^*\right)^2}\,,\quad 1$$

for the identity, inverse and log-link, respectively. Note that in the matrix

$$\mathbf{X} = (\mathbf{X}^{(1)}, \mathbf{X}^{(2)}) = (x_1, \ldots, x_n, x_{n+1}, \ldots, x_{n+m})$$

the elements in the matrix $\mathbf{X}^{(1)} = (x_1, \ldots, x_n)$ are fixed (because they correspond to observations from the initial day) and the criteria are optimized with respect to the experimental conditions $\mathbf{X}^{(2)} = (x_{n+1}, \ldots, x_{n+m})$ for the experiments at a different day. We reflect this fact by the notation

$$\Phi_D(\mathbf{X}^{(2)}|s) = \Phi_D((\mathbf{X}^{(1)}, \mathbf{X}^{(2)})|s) \tag{3.7}$$

$$\Phi_B(\mathbf{X}^{(2)}|\mathcal{M}) = \Phi_B((\mathbf{X}^{(1)}, \mathbf{X}^{(2)})|\mathcal{M}) \tag{3.8}$$

for the criteria (3.2) and (3.3), respectively. The corresponding locally optimal designs are denoted by $\mathbf{X}_s^{*(2)}$ and the analogue of the efficiency (3.4) is given by

$$\mathrm{eff}_D(\mathbf{X}^{(2)}|s) = \left(\frac{|I((\mathbf{X}^{(1)}, \mathbf{X}^{(2)}), s)|}{|I((\mathbf{X}^{(1)}, \mathbf{X}_s^{*(2)}), s)|} \right)^{1/p(s)+1}, \tag{3.9}$$

where the Fisher information matrix I is defined in (3.6). The D-optimality criterion is well justified if the main goal is to estimate all parameters in the presence of such day effects.
On the other hand other optimality criteria should be used if the only goal of the experiment is the investigation of an additional day effect. For this purpose a likelihood ratio test for the hypothesis

$$H_0 : \gamma = 0 \tag{3.10}$$

on the basis of all $n+m$ observations is usually performed. Standard results on the asymptotic properties of the likelihood ratio test show that the power of the test for the hypothesis (3.10) in a model $s = (g, z, \beta)$ is an increasing function of the quantity

$$\Phi_{D_1}(\mathbf{X}^{(2)}|s) = (e_{p(s)+1}^T I^{-1}(\mathbf{X}, s) e_{p(s)+1})^{-1} \tag{3.11}$$

where $\mathbf{X} = (\mathbf{X}^{(1)}, \mathbf{X}^{(2)})$, $\mathbf{X}^{(1)} = (x_1, \ldots, x_n)$, $\mathbf{X}^{(2)} = (x_{n+1}, \ldots, x_{n+m})$ and $e_{p(s)+1} = (0, \ldots, 0, 1)^T$ denotes the $(p(s)+1)$-th unit vector in $\mathbb{R}^{p(s)+1}$ [see Dette et al. (2008)]. Consequently, an optimal design for investigating the existence of an additional day effect if a particular model $s = (g, z, \beta)$ is used for the data analysis maximizes the function $\Phi_{D_1}(\mathbf{X}^{(2)}|s)$ with respect to the choice of the experimental conditions $\mathbf{X}^{(2)} = (x_{n+1}, \ldots, x_{n+m})$ for the m observations taken at any further day. The criterion defined by (3.11) is called D_1-optimality criterion in the literature. D_1-optimal designs have been studied by several authors in the context of linear and nonlinear regression models [see Studden (1980), Dette et al. (2005) or Dette et al. (2010) among others], but less work can be found on D_1-optimal designs for generalized linear models. In order to address uncertainty with respect to the model assumptions we denote by $\mathbf{X}_s^{*(2)}$ the locally D_1-optimal design maximizing the criterion defined in (3.11) and define the D_1-efficiency of a design $\mathbf{X} = (\mathbf{X}^{(1)}, \mathbf{X}^{(2)})$ in model $s = (g, z, \beta)$ by

$$\text{eff}_{D_1}(\mathbf{X}^{(2)}|s) = \frac{\Phi_{D_1}(\mathbf{X}^{(2)}|s)}{\Phi_{D_1}(\mathbf{X}_s^{*(2)}|s)}. \tag{3.12}$$

The Bayesian D_1-optimality criterion is finally defined by

$$\Phi_{B_1}(\mathbf{X}^{(2)}|\mathcal{M}) = \int_{\mathcal{M}} \text{eff}_{D_1}(\mathbf{X}^{(2)}|s) dh_1(\beta|g, z) dh_2(z|g) dh_3(g), \tag{3.13}$$

where h_1, h_2 and h_3 represent again cumulative distribution functions reflecting the importance of the particular constellation (g, z, β). Criteria of this type have been discussed by several authors in the case of linear regression models [see Dette (1994), Dette and Haller (1998)].

4 Optimal designs for thermal spraying

	temperature	velocity	flame width	flame intensity
Main effects	L, K, D	L, K, D, FDV	L, K, D, FDV	L, K, D, FDV
Squared effects	K^2	K^2	K^2	L^2, K^2, FDV^2
Interaction terms	–	$L \cdot K$	–	$D \cdot FDV$
Link	identity	logistic	inverse	identity
BIC	245.744	196.979	99.749	106.148

Table 4.1: *The generalized linear models chosen by the BIC for the four responses observed in the thermal spraying process.*

We return to the problem of designing additional experiments for the thermal spraying process. In the application the design space for each variable is the interval $[-2, 2]$ and 30 observations have already been made on the basis of a central composite design $\mathbf{X}_C^{(1)}$ (see Table A.1 in Appendix A.1) while a small number of additional experiments, e.g. four, are to be conducted for the investigation of an additional day effect. For each response (temperature, velocity, flame width, flame intensity) the data from the initial day has been used to identify a generalized linear model in the class of all models with the three link functions specified in Section 2.2 and different forms for the vector z on the basis of the BIC. The corresponding results are listed in Table 4.1. For each response the parameter estimates corresponding to the model chosen by the BIC are shown in Table A.2 in Appendix A.1. For example, for the

temperature	velocity	flame width	flame intensity
80.03%	71.13%	68.11%	64.85%
81.62%	72.94%	74.48%	69.91%
79.56%	74.58%	74.58%	72.21%

Table 4.2: *First row: D-efficiencies of the reference design. Second row: D-efficiencies of the reference design* $\mathbf{X}_R^{(2)}$ *with respect to the design* $\mathbf{X}_B^{*(2)}$ *maximizing the multi objective criterion* (3.8)*, where* γ *has been fixed. Third row: D-efficiencies of the reference design* $\mathbf{X}_R^{(2)}$ *with respect to the design* $\mathbf{X}_B^{*(2)}$ *maximizing the multi objective D-criterion* (3.8)*, where uncertainty with respect to the parameter* γ *has been addressed.*

temperature the BIC selects the generalized linear model with gamma distribution and identity link where the linear part of the model is given by

$$z^T(x)\beta = \beta_0 + \beta_1 L + \beta_2 K + \beta_3 D + \beta_4 K^2.$$

The estimated values of the parameters $(\beta_0, \ldots, \beta_4)$ can be obtained from Table A.2. For the investigation of the existence of an additive day effect a reference design $\mathbf{X}_R^{(2)} = (x_{31}, \ldots, x_{34})$ for the four additional experiments was proposed, which is shown in Table A.6. In order to investigate the efficiency of this design we have calculated the best locally D-optimal designs for the models which were identified by the BIC for modeling the four responses with an additional day effect. These designs require the specification of the unknown parameters and we used the available information from the first 30 experiments of the first day to estimate β (see Table A.2), while the parameter γ for the additional day effect was chosen (on the basis of information from similar experiments) as $\gamma = -16$, $\gamma = 0.01$, $\gamma = 0.002$ and $\gamma = 0.09$ in the models for temperature, velocity, flame width and flame intensity, respectively.
All designs presented in this section are calculated by Particle Swarm Optimization (PSO) which was introduced by Eberhart and Kennedy (1995). We also refer to the monographs Clerc (2006) and Yang (2010b) for the general methodology.

4.1 D-optimal designs

The locally D-optimal designs are shown in Table A.3 in Appendix A.2, while the corresponding D-efficiencies

$$\mathrm{eff}_D(\mathbf{X}_R^{(2)}|s) = \left(\frac{|I(\mathbf{X}_R, s)|}{|I(\mathbf{X}_s^*, s)|} \right)^{1/(p(s)+1)}$$

for the designs $\mathbf{X}_R = (\mathbf{X}_C^{(1)}, \mathbf{X}_R^{(2)})$ and $\mathbf{X}_s^* = (\mathbf{X}_C^{(1)}, \mathbf{X}_s^{*(2)})$ are depicted in the first row of Table 4.2. Here $I(\mathbf{X}, s)$ is the Fisher information in the generalized linear model including the day effects and $p(s) + 1$ denotes the number of parameters in the corresponding model where $p(s)$ parameters appear in regression function $z^T(x)\beta$. We observe that for each type of response the corresponding locally D-optimal design yields a substantial improvement of the reference design. The efficiency of the reference design varies between 65% - 80%.

Because an important goal of the experiment is to answer the question of additional day

temperature	velocity	flame width	flame intensity
100.17%	195.57%	76.31%	202.41%
132.76%	129.17%	85.39%	515.47%
132.76%	131.51%	87.92%	508.25%

Table 4.3: *First row: D_1-efficiencies of the reference design with respect to the locally D-optimal designs for estimating the parameter γ (see formula (4.1)). Second row: D_1-efficiencies of the reference design $\mathbf{X}_R^{(2)}$ with respect to the design $\mathbf{X}_B^{*(2)}$ maximizing the multi objective D-criterion (3.8), where γ has been fixed. Third row: D_1-efficiencies of the reference design $\mathbf{X}_R^{(2)}$ with respect to the design $\mathbf{X}_B^{*(2)}$ maximizing the multi objective D-criterion (3.8), where uncertainty with respect to the parameter γ has been addressed.*

Locally D-optimal design for	Model			
	temperature	velocity	flame width	flame intensity
temperature	100.00%	85.19%	80.88%	72.91%
velocity	96.15%	100.00%	84.81%	93.05%
flame width	91.79%	84.29%	100.00%	73.05%
flame intensity	96.19%	87.93%	65.77%	100.00%

Table 4.4: *The efficiencies of the locally D-optimal designs for the different models.*

effects we display in Table 4.3 the D_1-efficiencies

$$\mathrm{eff}_{D_1}(\mathbf{X}_R^{(2)}, \mathbf{X}_s^{*(2)}|s) = \frac{\Phi_{D_1}(\mathbf{X}_R^{(2)}|s)}{\Phi_{D_1}(\mathbf{X}_s^{*(2)}|s)} \tag{4.1}$$

of the reference design $\mathbf{X}_R = (\mathbf{X}_C^{(1)}, \mathbf{X}_R^{(2)})$ with respect to the locally D-optimal design $(\mathbf{X}_C^{(1)}, \mathbf{X}_s^{(*2)})$ for estimating the parameter γ. Most of the D_1-efficiencies of the locally D-optimal designs are larger than 100% compared to the reference design. The locally D-optimal design only performs better if flame width is concerned. Summarizing this means that the locally D-optimal design does not yield an improvement of the reference design when the only goal of the experiment is a most precise estimation of the additional day effect. Therefore we also investigate locally D_1-optimal designs in Section 4.2 in order to optimize the power of the test for an additional day effect.

Note that the selected models for the four responses differ and it is not clear if a locally D-optimal design for a particular model (for example the model used for temperature) has good properties in the models used for the other responses. In Table 4.4 we show the D-efficiencies if a locally D-optimal design for a particular model is used for a different model. We observe a substantial loss of efficiency. For example if the locally D-optimal design for the flame intensity is used its D-efficiency for analyzing the flame width is only 65.77%. In order to address this problem we have used the multi-objective criterion (3.8) to find a design $\mathbf{X}^{(2)}$ for the observations on a different day with reasonable D-efficiencies in all models under consideration. We begin considering only uncertainty with respect to the model in the criterion (3.4), while all the parameters are fixed. We used equal weights for all four models from Table 4.1 as prior distribution and the resulting design is given in the left part of Table 4.5.

Run	L	K	D	FDV	L	K	D	FDV
1	2	2	2	2	2	2	2	-2
2	2	-2	2	-2	2	-2	-2	-2
3	-2	0.34	-2	2	-2	0.37	-2	2
4	-2	-2	-2	-2	-2	-2	2	2

Table 4.5: *Bayesian D-optimal designs with respect to the criterion* (3.8) *for the four generalized linear models specified in Table 4.1. Left part: parameter of the day effect γ is fixed; right part: three values for the parameter of the day effect γ, $\gamma \pm 10\%\gamma$.*

The corresponding efficiencies

$$\mathrm{eff}_D(\mathbf{X}_R^{(2)}, \mathbf{X}_B^{*(2)}|s) = \Big(\frac{|I(\mathbf{X}_R, s)|}{|I(\mathbf{X}_B^*, s)|}\Big)^{1/(p(s)+1)} \tag{4.2}$$

$$\mathrm{eff}_{D_1}(\mathbf{X}_R^{(2)}, \mathbf{X}_B^{*(2)}|s) = \frac{\Phi_{D_1}(\mathbf{X}_R^{(2)}|s)}{\Phi_{D_1}(\mathbf{X}_B^{*(2)}|s)} \tag{4.3}$$

of the reference design $\mathbf{X}_R = (\mathbf{X}_C^{(1)}, \mathbf{X}_R^{(2)})$ with respect to the Bayesian D-optimal design $\mathbf{X}_B^* = (\mathbf{X}_C^{(1)}, \mathbf{X}_B^{*(2)})$ are presented in the second line of Table 4.2 and 4.3, respectively. We observe a similar improvement with respect to D-efficiency as obtained by the locally D-optimal designs. From this table we can also easily calculate the D-efficiencies of the design ${\mathbf{X}_B^*}^{(2)}$, which are given by 98.04%, 97.52%, 91.45%, 92.77% in the models for the temperature, velocity, flame width and flame intensity, respectively. Similarly, the efficiencies $\mathrm{eff}_{D_1}(\mathbf{X}_B^{*(2)}, \mathbf{X}_s^{*(2)})$ of the design $\mathbf{X}_B^*$ with respect to the locally D-optimal designs for estimating the parameter γ are obtained as 132.53%, 66.05%, 111.90%, 254.67%. This means that the compromise improves the reference design in all models with respect to parameter estimation. On the other hand for the estimation of the day effect only an improvement in the model for flame width is achieved (note that the design ${\mathbf{X}_B^*}^{(2)}$ is not constructed for this purpose).
While rather precise information is available for the parameter β from the first 30 observations, the designs and its properties might be sensitive with respect to the specification of the parameter γ for the additional day effect. In order to construct designs, which address this uncertainty we can also use the criterion (3.4), where we now also allow for uncertainty with respect to the parameter γ in the criterion. More precisely, for each of the four models we consider three possible values for γ, namely the value used in the local D-optimality criterion and 90% and 110% of this value (for example for the temperature model we used 14.4, 16, and 17.6 as possible values of γ). The resulting criterion (3.4) therefore consists of a sum of 12 terms and the maximizing design is depicted in the right part of Table 4.5. The structure of the two Bayesian D-optimal designs is very similar, since both designs put most of the design points in the edges of the design space. The D- and D_1-efficiencies are presented in the third rows of Table 4.2 and 4.3, respectively. Because of the similarity of the two Bayesian D- optimal designs the efficiences have nearly the same values.
These investigations show that the D-optimal designs yield a substantial improvement of the reference design if all parameters in the model (3.5) have to be estimated. On the other hand, if

temperature	velocity	flame width	flame intensity
90.51%	86.51%	63.81%	90.85%
90.55%	86.61%	64.68%	94.70%
90.64%	86.69%	66.05%	94.37%

Table 4.6: *First row: D_1-efficiencies of the reference design. Second row: D_1-efficiencies of the reference design $\mathbf{X}_R^{(2)}$ with respect to the design $\mathbf{X}_{B_1}^{*(2)}$ maximizing the multi objective criterion* (3.13)*, where γ has been fixed. Third row: D_1-efficiencies of the reference design $\mathbf{X}_R^{(2)}$ with respect to the design $\mathbf{X}_{B_1}^{*(2)}$ maximizing the multi objective criterion* (3.13)*, where uncertainty with respect to the parameter γ has been addressed.*

the only interest of the experiment is the estimation of a day effect, the reference design yields a more precise estimate of the parameter γ in the models for temperature, velocity and flame intensity than optimal designs based on D-optimality criteria.

4.2 Optimal designs for testing for an additional day effect

If the main interest of the experiment is the investigation of the existence of an additional day effect the design can be constructed such that the test for the hypothesis $H_0 : \gamma = 0$ is most powerful, which is reflected by the criterion Φ_{D_1} defined in (3.11). The corresponding multi-objective criterion addressing uncertainty with respect to the regression model, link function and parameters is given by (3.13). The locally D_1-optimal designs for the four models in Table 4.1 are presented in Table A.4 in Appendix A.2. We observe that in contrast to D-optimal designs D_1-optimal designs do not use the edges of the design space. The efficiencies of the reference designs $\mathbf{X}_R^{(2)}$ are given in the first row of Table 4.6. For the temperature and velocity the D_1-efficiencies of the reference design are about 90%. On the other hand an improvement of the reference designs can be observed for velocity and flame width (here the efficiencies are 63.81% and 86.51%, respectively).

As in the previous section we construct a robust design for testing for an additional day effect by maximizing the multi objective criterion (3.13), where all parameters have been fixed (β is obtained from Table A.2, while information from other experiments was used for the parameter γ for the construction of the locally optimal designs, that is $\gamma = -16$, $\gamma = 0.01$, $\gamma = 0.002$ and $\gamma = 0.09$ in the models for temperature, velocity, flame width and flame intensity, respectively). The resulting design is shown in the left part of Table 4.7 and its efficiencies are presented in the second row of Table 4.6. We observe a similar improvement of the reference designs as obtained by the locally D_1-optimal designs. Finally, we consider designs addressing the fact that the parameter γ cannot be estimated from the data of the initial day. If we address the uncertainty about this parameter in the same way as described in the previous section we obtain the design presented in the right part of Table 4.7. The D_1-efficiencies of the reference designs $\mathbf{X}_R^{(2)}$ with respect to this design are shown in the third row of Table 4.6.
Both Bayesian D_1-optimal designs are similar but differ substantially from the two Bayesian D-optimal designs in Table 4.5. The D_1-optimal designs use more experimental conditions from the interior of the design space $[-2, 2]^4$. Their efficiencies are very similar with respect to

Run	L	K	D	FDV	L	K	D	FDV
1	0.03	-1.62	2.00	-0.65	-0.57	0.13	-1.15	-0.96
2	0.90	0.36	0.44	-0.57	0.46	-1.53	1.97	-0.61
3	1.11	0.53	-2.00	-0.70	-1.37	0.45	-0.61	1.83
4	-1.83	0.54	-0.53	2.00	1.42	0.84	-0.27	-0.60

Table 4.7: *Bayesian D_1-optimal designs with respect to the criterion* (3.13) *for the four generalized linear models specified in Table 4.1. Left part: parameter of the day effect γ is fixed; right part: three values for the parameter of the day effect, γ, $\gamma \pm 10\%\gamma$.*

the locally D_1-optimal designs and range between 65% and 95%. Whereas the reference design performs nearly as well as the two Bayesian D_1-optimal designs in the cases of temperature and velocity, in the cases of flame width and flame intensity the Bayesian D_1-optimal yields more precise estimates as to the reference designs.

On the other hand the D-efficiencies of the Bayesian D_1-optimal designs are given by 85.14%, 73.69%, 79.58% and 69.23% for the temperature velocity, flame width and flame intensity, respectively, and therefore these designs are not very efficient for estimating all parameters in a generalized linear model with an additional day effect.

4.3 Efficient designs for estimating and testing

The numerical results of Section 4.1 and 4.2 show that different objectives such as estimation of all parameters and testing for an additional day effect result in rather different experimental designs, and optimal designs for one particular task (such as maximization of the power) are usually not efficient for the other (estimation of parameters). In order to construct efficient designs for these contradicting tasks we consider in a final step a compromise criterion of the form

$$\alpha\Phi_B(\mathbf{X}^{(2)}|\mathcal{M}) + (1-\alpha)\Phi_{B_1}(\mathbf{X}^{(2)}|\mathcal{M}), \tag{4.4}$$

where Φ_B and Φ_{B_1} are defined in (3.8) and (3.13), respectively, and $\alpha \in [0,1]$ is a pre-determined constant reflecting the importance of the different goals estimation and testing. The resulting design for four additional experiments is shown in Table 4.8 for $\alpha = 0.5$, where we use in both criteria the same prior distributions as described in Section 4.1 and 4.2. The corresponding efficiencies are depicted in Table 4.9 and we observe that this design yields high D_1-efficiencies in all four models under consideration and additionally a substantial improvement with respect to the D-efficiencies, which vary between 76% and 99%. The D-efficiencies could be increased if larger values of α are used in the criterion (4.4) at the expense of smaller D_1-efficiencies. For the sake of brevity these results are not depicted.

Run	L	K	D	FDV
1	0.10	0.17	2.00	-0.76
2	0.29	-2.00	-2.00	-1.05
3	1.75	0.58	2.00	-0.08
4	-2.00	0.41	-2.00	2.00

Table 4.8: *The compromise design maximizing the criterion* (4.4)*.The Bayesian criteria Φ_B and Φ_{B_1} only consider model uncertainty.*

	model			
	temperature	velocity	flame width	flame intensity
D-efficiency	91.15%	77.84%	85.38%	76.12%
D_1-efficiency	99.17%	98.50%	97.89%	91.02%

Table 4.9: *D- and D_1-efficiences of the compromise design maximizing the criterion* (4.4).

4.4 Experimental results

A further series of eight experiments was conducted to improve the understanding of the thermal spraying process, where four runs were performed under a reference and an optimal design, respectively. Because the goal was to estimate all parameters in the models for temperature, velocity, flame width and intensity (including the day effect) a Bayesian D-optimal design was calculated. In order to address the uncertainty with respect to the day effect γ an average over five possible values for γ was calculated, i.e. γ, $\gamma \pm 10\%\gamma$, $\gamma \pm 20\%\gamma$. The calculated reference and Bayesian D-optimal design are depicted in Table 4.10. The Bayesian D-optimal design advices the experimenter to use experimental conditions at the boundary of the design space whereas the reference design naturally stays more towards the center (i.e. it only contains coded -1, 1 of the input parameters).
The observed data are given in Table A.5 and A.6 in Appendix A.3 and it can be seen that the particle properties at the extreme experimental conditions differ substantially from the original data. For example, in run number 2 the temperature and the value of flame intensity are only 1298.81 and 9.84, respectively. As a consequence adding the four additional runs to the initial data leads to noticeable changes in the parameter estimates for the model of temperature if the optimal design is used. All other parameter estimates are only marginally altered (these results are not displayed for the sake of brevity).
The D-efficiencies (4.2) of the reference design with respect to this Bayesian D-optimal design are displayed in the first row of Table 4.11 and are given by 79.53%, 75.35%, 78.14% and 69.03%. It is also of interest to compare these "theoretical" values (based on the 30 initial observations) with the "observed" D-efficiencies (based on the estimated covariance matrices from the 30 initial plus four additional observations), which are shown in the second line of Table 4.11. These results show a substantial improvement between 39% (temperature) and 86% (flame width) with respect to the D-criterion, if the 4 additionals runs are performed according to the Bayesian D-optimal design.

It might also be of interest to compare both designs with respect to their prediction properties. On the same day of the eight new experiments, fourteen additional experiments were conducted with different aims. The design and data are shown on Table A.7. We investigate how the estimated generalized linear models from the two designs perform with respect to prediction of these measured in flight properties. The lower part of Table 4.11 presents the mean squared error between predicted and measured particle properties, and we observe that the reference design leads to smaller mean squared errors for temperature and velocity whereas the Bayesian D-optimal design yields better predictions of the flame width and intensity.

Run	L	K	D	FDV	L	K	D	FDV
1	2	2	2	-0.53	1	1	-1	-1
2	-2	-2	2	-2	-1	1	1	-1
3	-2	0.31	-2	2	1	1	1	1
4	2	-2	-2	-2	1	-1	1	-1

Table 4.10: *Right part: Bayesian D-optimal design with respect to the criterion* (3.8) *with five values for the parameter of the day effect γ, $\gamma \pm 10\%\gamma$, $\gamma \pm 20\%$. Left part: The reference design.*

	Temperature	Velocity	Flame Width	Flame Intensity
theoretical D-efficiency	79.53%	75.35%	78.14%	69.34%
observed D-efficiency	39.34%	85.89%	86.46%	57.05%
MSE optimal design	27.40	12.65	1.43	1.89
MSE reference design	16.97	8.91	4.10	2.73

Table 4.11: *Upper part: D-efficiencies (theoretical and observed) of the reference design with respect to the Bayesian D-optimal design. Lower part: MSE for the prediction of* 14 *runs from (A.7) using four additional observations from an optimal and a reference design.*

5 Concluding remarks

In this paper we have investigated optimal designs for analyzing thermal spraying processes on the basis of generalized linear models, where observations are available from experiments conducted at two different days. While a central composite design is used for the experiments on the first day, optimal designs for the experiments on a further day are constructed, which also allow for testing the existence of an additional additive day effect in the generalized linear model. Uncertainty with respect to the model assumptions occurs from several perspectives and is addressed in the optimality criteria used for the construction of efficient designs. Firstly, one design is constructed for analyzing various in-flight properties as temperature, velocity, flame width and flame intensity with different generalized linear models. Secondly, the criteria also address the problem uncertainty with respect to unknown day effect.

We consider D- and D_1-optimality criteria which yield designs minimizing the volume of the ellipsoid of concentration for the vector of all parameters and maximizing the power of the likelihood ratio test for the existence of an additional day effect, respectively. Bayesian D- and D_1-optimal designs (addressing the problem of model uncertainty and imprecise information about the unknown day effect) are determined and investigated with respect to their statistical efficiencies. D-optimal designs use the edges of the design space and it is demonstrated that these designs improve a reference design substantially with respect to the efficiency for estimating all parameters in the generalized linear model. On the other hand, in many cases the reference design yields more power for the likelihood ratio test of an additional day effect than the D-optimal designs and the reference design can be improved by a Bayesian D_1-optimal design. Therefore, if the only goal of the additional experiments is the investigation of an additional day effect Bayesian D_1-optimal designs should be used. These designs advice the experimenter to take more observations in the interior of the design space. Thus the two ob-

jectives estimation of all parameters and testing for an additional day effect result in rather different experimental designs and the goals of the experiment have to be carefully defined before optimal designs are constructed for the analysis of thermal spraying processes with generalized linear models. If this is not possible, a compromise design criterion can be developed, which yields designs with reasonable efficiencies for estimating all parameters and testing for an additional day effect under model uncertainty.
Finally, we use the results of this paper to design new experiments for the analysis of the thermal spraying process and demonstrate that a Bayesian D-optimal design improves a reference design with respect to the D-optimality criterion. On the other hand, for the prediction of 14 additional experiments the superiority of the Bayesian D-optimal design is only visible for the responses flame width and intensity.

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A Appendix: Data, designs and and estimates

A.1 Estimates in the identified models and the standard design

In this section we display the parameter estimates in the models identified by the BIC for the four responses. The values are obtained from the 30 observations of the initial day and are used in the local optimality criteria to construct the optimal design for the additional four runs on the next day.

Run	L	K	D	FDV	Temperature	Velocity	Flame Width	Flame Intensity
1	1	-1	1	-1	1450.5706	674.1324	7.9059	13.1971
2	1	1	1	1	1500.9382	726.6706	12.4912	21.0029
3	-1	-1	1	-1	1484.8952	649.1190	8.1238	15.3929
4	-1	-1	-1	1	1534.6750	666.0781	13.5563	21.4375
5	0	0	0	0	1519.4829	709.3029	11.9629	19.7143
6	0	0	0	0	1527.6065	713.6581	12.1742	19.9419
7	-1	1	1	-1	1543.3053	730.3474	10.3711	18.3579
8	-1	1	-1	1	1574.0970	739.4212	14.9909	23.3667
9	1	1	-1	1	1536.2371	756.7057	13.7657	21.8543
10	1	-1	-1	-1	1497.6209	698.4093	8.7767	15.8093
11	0	0	0	0	1527.8571	710.8250	11.9821	19.9393
12	-1	1	-1	-1	1564.3114	753.5943	11.1229	18.7143
13	1	1	-1	-1	1528.9267	770.7367	9.5000	17.0000
14	-1	1	1	1	1546.6594	714.0031	14.8187	23.5625
15	1	-1	1	1	1484.7806	665.0000	12.3472	20.5139
16	-1	-1	1	1	1502.0265	640.9088	13.2176	21.3500
17	-1	-1	-1	-1	1525.3917	678.9194	10.0417	17.2917
18	1	1	1	-1	1508.2706	749.0647	8.5206	15.9706
19	0	0	0	0	1535.5706	714.2500	12.3294	20.1412
20	1	-1	-1	1	1504.6000	689.5364	12.6121	20.2879
21	0	0	0	0	1521.7227	708.9636	11.7977	19.6568
22	0	0	-2	0	1534.7182	726.6697	11.7939	19.2697
23	-2	0	0	0	1542.8600	688.1171	12.4971	20.2914
24	2	0	0	0	1462.0088	723.5471	9.1765	16.7735
25	0	0	0	0	1521.4765	709.1412	11.5176	19.2412
26	0	0	0	-2	1516.5378	708.6919	11.3649	19.0757
27	0	0	2	0	1491.7684	684.3026	10.4868	18.5632
28	0	2	0	0	1512.7982	755.1382	10.8436	18.8527
29	0	0	0	2	1520.6485	695.1848	14.4455	22.7879
30	0	-2	0	0	1435.7488	612.6093	8.9209	16.0163

Table A.1: *Central Composite Design used for the first* 30 *observations*

	Temperature	Velocity	Flame Width	Flame Intensity
(Intercept)	1523.2627 (2.6722)	6.5648 (0.0016)	0.0863 (0.0018)	19.4784 (0.3364)
L	-17.7423 (2.3136)	0.0136 (0.0014)	0.0053 (0.0015)	-0.8887 (0.1901)
K	19.6580 (2.2939)	0.0516 (0.0014)	-0.0044 (0.0016)	0.8646 (0.1863)
D	-13.8181 (2.3136)	-0.0171 (0.0014)	0.0029 (0.0015)	-0.3709 (0.1970)
FDV	-	-0.0078 (0.0014)	-0.0123 (0.0015)	2.1661 (0.2042)
L^2	-	-	-	-0.3096 (0.1760
K^2	-9.9897 (2.0813)	-0.0092 (0.0012)	0.0039 (0.0015)	-0.5615 (0.1925)
FDV^2	-	-	-	0.5092 (0.1699)
$L \cdot K$	-	-0.0031 (0.0017)	-	-
$D \cdot FDV$	-	-	-	0.4095 (0.2378)

Table A.2: *Parameter estimates and standard errors in brackets of the models for temperature, velocity, flame width and flame intensity chosen by the BIC.*

A.2 Optimal designs with four runs

	temperature			velocity				flame width				flame intensity			
Run	L	K	D	L	K	D	FDV	L	K	D	FDV	L	K	D	FDV
1	-2	-0.07	-2	-2	2	-2	2	2	0.37	2	2	2	2	2	-2
2	2	-0.13	2	- 2	-2	2	2	-2	2	-2	2	-2	-2	2	-2
3	2	2	-2	2	2	-2	-2	-2	0.08	-2	2	0.47	-0.95	2	-0.64
4	-2	-2	2	2	-2	2	-2	2	0.37	-2	-2	2	-2	-2	-2

Table A.3: *Locally D-optimal designs for the responses temperature (left part), velocity (middle left part), flame width (middle right part) and flame intensity (right part)*

	temperature			velocity				flame width				flame intensity			
Run	L	K	D	L	K	D	FDV	L	K	D	FDV	L	K	D	FDV
1	-1.23	-1.32	-0.05	0.00	0.96	0.64	-0.73	-1.02	0.29	1.72	-1.83	1.59	1.03	1.30	-1.075
2	0.10	0.94	0.81	-0.54	-0.62	-1.90	0.84	-1.81	0.99	0.29	1.92	0.09	-1.53	1.43	-0.51
3	0.28	0.64	0.42	0.40	-1.12	1.31	-1.25	0.07	-1.07	-1.40	-0.29	0.00	-0.61	-0.66	0.64
4	1.20	-0.64	-0.89	0.13	0.79	-0.05	1.14	1.54	0.28	-1.58	1.50	-0.85 -	0.22	-1.79	-1.11

Table A.4: *Locally D_1-optimal designs for the responses temperature (left part), velocity (middle left part), flame width (middle right part) and flame intensity (right part)*

A.3 Bayesian D-optimal and reference design for 4 additional runs

Run	L	K	D	FDV	Temperature	Velocity	Flame Width	Flame Intensity
1	2	2	2	-0.53	1466.8123	787.5585	12.3446	22.0938
2	-2	-2	2	-2	1298.8123	593.2692	10.2261	9.8646
3	-2	0.31	-2	2	1560.3545	706.1242	18.9030	28.6333
4	2	-2	-2	-2	1437.7284	687.5351	7.4500	13.8446

Table A.5: *Bayesian D-optimal design for four additional runs*

Run	L	K	D	FDV	Temperature	Velocity	Flame Width	Flame Intensity
1	1	1	-1	-1	1527.1426	778.2632	13.2456	22.3985
2	-1	1	1	-1	1493.9143	752.6063	12.4841	22.3333
3	1	1	1	1	1507.5667	752.8273	17.6909	27.9348
4	1	-1	1	-1	1443.8103	696.7851	10.4471	18.8977

Table A.6: *Reference design for four additional runs*

A.4 Additional runs and results

Run	L	K	D	FDV	Temperature	Velocity	Flame Width	Flame Intensity
1	0.01	1.09	-0.20	-1.67	1514.6610	782.1146	10.2951	19.1976
2	0.01	1.09	-0.20	-1.67	1521.8186	786.4209	10.0116	18.6930
3	1.82	-0.36	0.46	-0.58	1475.2022	734.7978	11.7778	21.1467
4	1.82	-0.36	0.46	-0.58	1488.6825	737.3925	11.1850	20.0250
5	1.27	-1.32	0.38	-0.71	1434.2327	687.7714	10.4510	18.9408
6	1.27	-1.32	0.38	-0.71	1456.8717	689.5453	10.0019	17.7623
7	0.00	-0.01	0.20	-1.73	1478.7136	743.1182	9.7227	17.1773
8	0.00	-0.01	0.20	-1.73	1520.9761	747.9326	9.3457	16.3413
9	-0.48	0.50	-0.60	1.78	1529.3061	726.5163	17.9367	27.6449
10	-0.48	0.50	-0.60	1.78	1521.4094	721.4434	17.4113	27.1906
11	1.00	-1.00	-1.00	1.00	1507.4579	698.7368	16.2667	25.5053
12	-1.00	-1.00	-1.00	-1.00	1491.3108	696.6215	12.4585	21.0092
13	-1.00	-1.00	1.00	1.00	1453.4762	661.9381	16.2333	26.2127
14	-1.00	1.00	-1.00	1.00	1552.7875	749.2734	18.3484	27.9703

Table A.7: *Additional runs used for the investigation of the prediction properties of the reference and optimal design*