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Optimal orthogonal block designs for three-component symmetric general blending models in mixture experiment

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ABSTRACT

In mixture experiments, the observed response is determined by the relative proportions of the components, consequently rendering the experimental region a simplex. This paper focuses primarily on the optimal designs of mixture experiments that involve process variables. Prior research has extensively delved into optimal orthogonal block designs for some classic mixture models with process variables. Based on the framework of general blending models, this paper proposes a class of symmetric linear mixture models, which can be regarded as a generalization of many existing ones. Under the orthogonal blocking conditions, orthogonal block designs are devised through Latin squares in the presence of process variables. The *D*-, *A*-, and *E*-optimality criteria are utilized to obtain optimal designs at the boundary of the simplex in the case of 3 components. As the values of the exponents change, numerically derived optimal design points are presented to illustrate the pattern of their variations, and to verify the consistency of the results with previous research on some specific symmetric general blending models.

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1. Introduction

Mixture experiments are typically used to investigate the effects of the proportions of mixing components on the response variable, and are common in fields such as chemistry, pharmaceuticals, material science and food industry (see e.g., Atkinson et al., 2007; Wu & Hamada, 2021). In mixture experiments, the basic constraint is that the total proportion of all components adds up to 1, and the experimental region is a regular $(q-1)$-dimensional simplex.

Models are powerful tools for analysing data from mixture experiments. There is no constant term in mixture models, because a constant term can always be replaced through the linear constraints (see also Cornell, 2002). Scheffé polynomials, also known as canonical polynomials, originated from re-parameterization of the standard polynomials by Scheffé (1958), and have played a significant role in the development of mixture experiments since then. Three homogeneous models were then suggested by Becker (1968), which could describe more complex interactions than Scheffé polynomials. Brown et al. (2015) introduced general blending models (GBM) to describe complicated curvilinear effects, which were general cases

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of several mixture models, including Scheffé polynomials and Becker's H2 and H3 models. GBM is non-linear and its coefficients are challenging to estimate, but it often demonstrates good fit in data analysis, particularly in instances where there exist unknown non-additive effects of the components (Bao, 2021).

Designs are useful methodologies for collecting data scientifically in mixture experiments. The choice of designs plays a critical role in ensuring the quality and efficiency of statistical analysis. Kiefer (1959) proposed the concept of optimal designs, and the well-known D-, A- and E-optimality criteria were defined, to minimize the variance of the coefficients in parameter estimation. For more information on optimal designs, please refer to Pukelsheim (2006). In recent years, the optimal problems have become increasingly complex, involving the application of new models or optimality criteria. See also Dette et al. (2022) and Zhou et al. (2024). Along these lines, a series of noteworthy papers, Peter et al. (2016), Duarte et al. (2021), Gong et al. (2022) and Li et al. (2024), developed a range of optimal designs in mixture experiments.

Process variables refer to qualitative factors other than mixture components, typically encompassing operational conditions or settings, such as temperature and pressure, which can potentially influence the response (see also Donev, 1989; Sinha et al., 2014). The problem of mixture experiments involving process variables was introduced by Scheffé (1963). Subsequently, Nigam (1970) and Nigam (1976) constructed block designs through mutually orthogonal Latin squares, and gave orthogonal blocking conditions for quadratic Scheffé polynomial. Following this, Czitrom (1988), Lewis et al. (1994), Aggarwal et al. (2002) and Mario and Peter (2023) sequentially obtained optimal orthogonal block designs for various mixture models under different criteria. Aggarwal et al. (2013) used F-squares to construct orthogonal block designs for Becker's models of 3 and 4 components.

In this paper, we introduce the symmetric general blending model as a class of mixture models, and develop orthogonal block designs aimed at mitigating the impact of process variables on the estimation of parameters. We take into account D-, A-, and E-optimality criteria, and provide theoretical and numerical results in the case of 3 components.

2. Symmetric general blending model

In this chapter, we will introduce several commonly used models in mixture experiments, all of which are second-order, with q single terms and $\binom{q}{2}$ binary terms, where q is the number of mixture components. Let y denote the response, and $x_1, x_2, \dots, x_q$ denote the proportions of the q components. Remain that the experimental region is a regular $(q-1)$-dimensional simplex, i.e.,

$$S_{q-1} = \left\{ (x_1, x_2, \dots, x_q) \middle| \sum_{i=1}^{q} x_i = 1, \quad x_i \geq 0, \; i = 1, 2, \dots, q \right\}. \tag{1}$$

We begin with a general form of mixture models of order 2, that is,

$$E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q} \beta_{ij} h(x_i, x_j), \tag{2}$$

where $h(x_i, x_j)$ is a function of x_i and x_j that characterizes the joint effects between them, and β_i and β_{ij} are linear coefficients to be estimated, $i, j = 1, 2, \dots, q$.

Definition 2.1: (Liu & Neudecker, 1997; Li et al., 2017) A mixture model is symmetric, when it is symmetrically invariant with respect to the components $x_1, x_2, \ldots, x_q$. In other words, for $x = (x_1, x_2, \ldots, x_q)$, the function value remains unchanged upon swapping any of its components.

By Definition 2.1, a second-order mixture model (2) is symmetric, when its binary terms satisfy that, $\sum_{i<j}^{q} h(x_i, x_j)$ is a symmetric function of $x_1, x_2, \ldots, x_q$. It is obvious that, three classic mixture models, i.e., the quadratic Scheffé polynomial (3) and Becker's H2 (4) and H3 models (5) are symmetric, respectively:

$$E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q} \beta_{ij} x_i x_j, \tag{3}$$

$$E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q} \beta_{ij} \frac{x_i x_j}{x_i + x_j}, \tag{4}$$

$$E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q} \beta_{ij} \sqrt{x_i x_j}. \tag{5}$$

The general blending model (GBM) (Brown et al., 2015) considers more complex joint effects, which is structured as follows:

$$E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q} \beta_{ij} \frac{x_i^{r_{ij}} x_j^{r_{ji}}}{(x_i + x_j)^{s_{ij}}}, \tag{6}$$

where r_{ij}, r_{ji} and s_{ij} $(i < j,\ i, j = 1, 2, \ldots, q)$ are exponents.

Based on the structure of GBM (6), we define a class of linear mixture models, named symmetric general blending model (SGBM), which retains flexible utilization of the exponents and also satisfies the symmetry properties defined in Definition 2.1.

Definition 2.2: A second-order SGBM can be formulated by

$$E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q} \beta_{ij} \frac{(x_i x_j)^r}{(x_i + x_j)^s}, \tag{7}$$

where r and s are exponents to be assigned. Typically, we consider that $r > 0$ and $s \geq 0$. To avoid meaningless results, we set $h(0, 0) = 0$, where $h(x_i, x_j) = \frac{(x_i x_j)^r}{(x_i + x_j)^s}$, for $i, j = 1, 2, \ldots, q$.

Through the exponents in SGBM (7), we are able to flexibly describe the interaction of the components in different mixture models. It is clear that, Models (3)–(5) are all particular cases of SGBM, with exponents assigned to $(r = 1, s = 0)$, $(r = 1, s = 1)$ and $(r = \frac{1}{2}, s = 0)$, respectively. Note that Becker's H1 model (also known as the minimum polynomial) is not a special case of SGBM, and thereby the optimal conclusions proposed in this paper are not applicable to the H1 model.

3. Orthogonal block designs

Process variables, introduced as factors that affect the response independent of the components in mixture experiments, are addressed by blocking the design to mitigate their impact on parameter estimation.

Let n denote the total number of runs, and m denote the number of process variables. A second-order SGBM (7) with process variables can be written as

$$E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q} \beta_{ij} \frac{(x_i x_j)^r}{(x_i + x_j)^s} + \sum_{k=1}^{m} \alpha_k z_k, \tag{8}$$

where z_k's are process variables, and α_k's are corresponding block difference parameters, for $k = 1, 2, \ldots, m$. Model (8) can also be expressed in the matrix notation:

$$E[\boldsymbol{y}] = \boldsymbol{X\beta} + \boldsymbol{Z\alpha}, \tag{9}$$

where $\boldsymbol{y}$ is the response vector of length n, $\boldsymbol{X}$ is the $n \times \frac{q(q+1)}{2}$ (extended) design matrix, $\boldsymbol{Z} = (\boldsymbol{z}_1, \boldsymbol{z}_2, \ldots, \boldsymbol{z}_m)$ is the $n \times m$ block matrix, $\boldsymbol{z}_1, \boldsymbol{z}_2, \ldots, \boldsymbol{z}_m$ are process vectors of length n, $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$ are both parameter column vectors of length m and $\frac{q(q+1)}{2}$, respectively.

According to the orthogonal blocking condition for quadratic Scheffé polynomial given by Nigam (1970), we define similar conditions for SGBM's (9) to ensure that the effects of mixture components can be unbiased estimated and not affected by the effects of process variables.

Definition 3.1 (Orthogonal blocking condition): A block design for SGBM with process variables (9) satisfies the orthogonal blocking condition, when the estimates of the coefficients are independent of the block effects, that is

$$\boldsymbol{X}^\top \boldsymbol{Z} = \boldsymbol{0}_{\frac{q(q+1)}{2} \times m}, \tag{10}$$

where $\boldsymbol{0}_{\frac{q(q+1)}{2} \times m}$ is a matrix of zeros.

For each block, the orthogonal blocking condition (10) is equivalent to

$$\sum_w x_{iw} = l_i; \quad \sum_w \frac{\left(x_{iw} x_{jw}\right)^r}{\left(x_{iw} + x_{jw}\right)^s} = l_{ij},$$

where l_i and l_{ij} are constants for $i < j$, $i, j = 1, 2, \ldots, q$, and the range of w depends on the entries of each block.

Block designs can be generated through some well-established construction methods, and we utilize mutually orthogonal Latin squares (MOLS) to achieve the orthogonal blocking condition (10). If q is a prime power, there exist $(q-1)$ MOLS of order q, denoted $q-1$ MOLS(q). Referring to the construction by Bose (1938), we construct $q-1$ MOLS(q), given in the forms of $\boldsymbol{L}_1, \boldsymbol{L}_2, \ldots, \boldsymbol{L}_{q-1}$, as follows. For $1 \le k \le q-1$ and $1 \le i, j \le q$, the element in the i-th row and j-th column of $\boldsymbol{L}_k$ is derived from

$$\boldsymbol{L}_{k,ij} = (k \cdot (j-1) + i - 1) \mod q. \tag{11}$$

This construction makes sure that the first column of each Latin square is always $(0, 1, 2, \ldots, q-1)^\top$, and there are no duplicate runs among $q-1$ MOLS(q). Then, we can replace the numbers 0, 1, 2, …, in order with the parameters $a,\ b,\ c, \dots$, and add the centroid of the simplex S_{q-1}, i.e., $(\frac{1}{q}, \frac{1}{q}, \dots, \frac{1}{q})$, into each Latin square, to ensure a non-singular information matrix $\frac{1}{n}\boldsymbol{X}^\top \boldsymbol{X}$. Overall, an orthogonal block design with $(q^2 - q + 1)$ support points will be subsequently obtained.

The number of process variables and their corresponding levels determine the number of blocks. When $q = 3$, we can construct an exact design with 8 runs throughout 2 MOLS(3), for an SGBM including a 2-level process variable (8):

$$\begin{pmatrix} \boldsymbol{p_1} & \boldsymbol{p_2} & \cdots & \boldsymbol{p_6} & \boldsymbol{p_7} \\ \frac{1}{8} & \frac{1}{8} & \cdots & \frac{1}{8} & \frac{1}{4} \end{pmatrix}, \tag{12}$$

where points $\boldsymbol{p_1}, \boldsymbol{p_2}, \dots, \boldsymbol{p_6}$ come from 2 MOLS(3), with parameters $0 \le a, b, c \le 1$ $(a + b + c = 1)$, and $\boldsymbol{p_7} = (\frac{1}{3}, \frac{1}{3}, \frac{1}{3})$ is the centroid of S_2, which is arranged into both blocks to enhance the balance of the design (hence, the number of replicates is 2 for $\boldsymbol{p_7}$):

$$\boldsymbol{B_1} = \begin{array}{c} \\ \boldsymbol{p_1} \\ \boldsymbol{p_2} \\ \boldsymbol{p_3} \\ \boldsymbol{p_7} \end{array} \begin{array}{c} \begin{matrix} x_1 & x_2 & x_3 \end{matrix} \\ \begin{pmatrix} a & b & c \\ b & c & a \\ c & a & b \\ \frac{1}{3} & \frac{1}{3} & \frac{1}{3} \end{pmatrix} \end{array}, \quad \boldsymbol{B_2} = \begin{array}{c} \\ \boldsymbol{p_4} \\ \boldsymbol{p_5} \\ \boldsymbol{p_6} \\ \boldsymbol{p_7} \end{array} \begin{array}{c} \begin{matrix} x_1 & x_2 & x_3 \end{matrix} \\ \begin{pmatrix} a & c & b \\ b & a & c \\ c & b & a \\ \frac{1}{3} & \frac{1}{3} & \frac{1}{3} \end{pmatrix} \end{array}.$$

When it comes to m 2-level process variables, we can divide the design for components into 2^m blocks. For each block, the design of the components is a repetition 2^{m-1} of $\boldsymbol{B_1}$ and $\boldsymbol{B_2}$. Simultaneously, the process variables are constructed by full factorial designs, which ensures that all process variables take the same level combination in the same block. We provide examples for $m = 1, 2$ as follows.

Example 3.2: The case of one process variable. Take the process variable at -1 level for Block 1 and at +1 level for Block 2, and the corresponding 8×6 design matrix and 8×1 block matrix under Design (12) are

$$\boldsymbol{X} = \begin{pmatrix} x_1 & x_2 & x_3 & x_2 : x_3 & x_1 : x_3 & x_1 : x_2 \\ a & b & c & h(b,c) & h(a,c) & h(a,b) \\ b & c & a & h(a,c) & h(a,b) & h(b,c) \\ c & a & b & h(a,b) & h(b,c) & h(a,c) \\ \frac{1}{3} & \frac{1}{3} & \frac{1}{3} & h(\frac{1}{3},\frac{1}{3}) & h(\frac{1}{3},\frac{1}{3}) & h(\frac{1}{3},\frac{1}{3}) \\ a & c & b & h(b,c) & h(a,b) & h(a,c) \\ b & a & c & h(a,c) & h(b,c) & h(a,b) \\ c & b & a & h(a,b) & h(a,c) & h(b,c) \\ \frac{1}{3} & \frac{1}{3} & \frac{1}{3} & h(\frac{1}{3},\frac{1}{3}) & h(\frac{1}{3},\frac{1}{3}) & h(\frac{1}{3},\frac{1}{3}) \end{pmatrix}, \quad \boldsymbol{Z} = \begin{pmatrix} -1 \\ -1 \\ -1 \\ -1 \\ +1 \\ +1 \\ +1 \\ +1 \end{pmatrix}, \tag{13}$$

where $h(x_i, x_j) = \frac{(x_i x_j)^r}{(x_i + x_j)^s}$, for $i \neq j,\ i, j = 1, 2, 3$.

Example 3.3: The case of two process variables. Based on the design matrix (13) for $m = 1$, the 16×6 design matrix and 16×2 block matrix for $m = 2$ can be written as

$$\boldsymbol{X}' = \begin{pmatrix} \boldsymbol{X} \\ \hline \boldsymbol{X} \end{pmatrix}, \quad \boldsymbol{Z}' = \left(\boldsymbol{z_1} \mid \boldsymbol{z_2} \right) = \left(\begin{array}{c|c} -\boldsymbol{1}_4 & -\boldsymbol{1}_4 \\ +\boldsymbol{1}_4 & -\boldsymbol{1}_4 \\ \hline +\boldsymbol{1}_4 & +\boldsymbol{1}_4 \\ -\boldsymbol{1}_4 & +\boldsymbol{1}_4 \end{array}\right) \tag{14}$$

where $\mathbf{1}_4$ is a 4-dimensional column vector of ones. Note that, $\boldsymbol{z}_1^\top \boldsymbol{z}_2 = 0$ and $\frac{1}{16}(\boldsymbol{X}')^\top \boldsymbol{X}' = \frac{1}{16} \cdot 2\boldsymbol{X}^\top \boldsymbol{X} = \frac{1}{8}\boldsymbol{X}^\top \boldsymbol{X}$, i.e., the information matrix remains invariant despite the change of m.

Then we can prove that Design (12) satisfies the orthogonal blocking conditions with any number of m process variables.

Theorem 3.4: *Design (12) is an orthogonal block design for 3-component SGBM with 2-level process variables.*

Proof: We start with the case of one 2-level process variable. Certainly, the sufficient condition for orthogonal blocking is satisfied: $\boldsymbol{X}^\top \boldsymbol{Z} = \mathbf{0}_{6\times 1}$. When $m = 2$, it can be shown from (14) that, $(\boldsymbol{X}')^\top \boldsymbol{z}_1 = \mathbf{0}_{6\times 1}$, $(\boldsymbol{X}')^\top \boldsymbol{z}_2 = \mathbf{0}_{6\times 1}$, and therefore $(\boldsymbol{X}')^\top \boldsymbol{Z}' = \mathbf{0}_{6\times 2}$. In the case of a general m, the $2^{m+2} \times 6$ design matrix and $2^{m+2} \times m$ block matrix under Design (12) can always be expressed by $\boldsymbol{X}$ in (13) and orthogonal vectors $\boldsymbol{z}_1, \boldsymbol{z}_2, \dots, \boldsymbol{z}_m$, and similarly to $m = 2$, orthogonal blocking condition can be verified. ■

Observations at or near the boundary of the experimental region exert a more significant influence on both the estimation of coefficients and the variance of predictions, compared to observations collected deep within the simplex (Cornell, 2002). Whether the optimal orthogonal block design achieves optimal values at the edge of the simplex has been a topic of discussion in previous studies, and researchers have verified that some optimality criteria tend to achieve their minimun at the boundary for some specific mixture models (see, e.g., Aggarwal et al., 2002; Chan, 1999; Czitrom, 1988). In this paper, we mainly focus on the orthogonal block designs for the 3-component SGBM with a 2-level process variable, with some support points located at the edge of the simplex. Without loss of generality, we assume that $c = 0,\ 0 \le a \le b \le 1$ (and hence, $a + b = 1$), and define ξ as any design on S_{q-1} that satisfies the structure of Design (12) with $c = 0$, and Ξ as the set of all such ξ's.

4. Optimal designs

In this chapter, the optimal ξ in Ξ will be studied, using D-, A- and E-optimality criteria, in order to minimize the variance of the estimator in Model (9) based on least squares estimation. The following standard assumption should be mentioned: the random errors are assumed to be identically independently normally distributed, with 0 expectation and constant variance. We begin with the study of the information matrix, which associates design ξ with the optimal criteria and directly determines the magnitude of the variance of the estimator, given by $\frac{1}{n}\boldsymbol{X}^\top \boldsymbol{X}$. Denote

$$\boldsymbol{M} = \boldsymbol{X}^\top \boldsymbol{X} = \begin{pmatrix} (A-B)\boldsymbol{I}_{3\times 3} + B\boldsymbol{J}_{3\times 3} & (E-F)\boldsymbol{I}_{3\times 3} + F\boldsymbol{J}_{3\times 3} \\ (E-F)\boldsymbol{I}_{3\times 3} + F\boldsymbol{J}_{3\times 3} & (C-D)\boldsymbol{I}_{3\times 3} + D\boldsymbol{J}_{3\times 3} \end{pmatrix}, \tag{15}$$

where $\boldsymbol{I}_{3\times 3}$ is the identity matrix of order 3, $\boldsymbol{J}_{3\times 3}$ is a 3×3 matrix of ones, and

$$A = 2\left(a^2 + b^2 + \frac{1}{9}\right), \quad B = 2\left(ab + \frac{1}{9}\right), \; C = 2\left((ab)^{2r} + \frac{3^{2s-4r}}{2^{2s}}\right),$$

$$D = \frac{3^{2s-4r}}{2^{2s-1}}, \quad E = \frac{3^{s-2r-1}}{2^{s-1}}, \; F = (ab)^r + \frac{3^{s-2r-1}}{2^{s-1}}. \tag{16}$$

Lemma 4.1: *The eigenvalues of* $\boldsymbol{M}$ *are*

$$\lambda_1, \lambda_2 = \frac{T_1 \pm \sqrt{T_1^2 - 4T_2}}{2}, \quad \lambda_3, \lambda_4 = \frac{T_3 \pm \sqrt{T_3^2 - 4T_4}}{2}, \tag{17}$$

where λ_1 *and* λ_2 *are both of multiplicity 2, and*

$$\begin{aligned} T_1 &= (A - B) + (C - D), \\ T_2 &= (A - B)(C - D) - (E - F)^2, \\ T_3 &= (A + 2B) + (C + 2D), \\ T_4 &= (A + 2B)(C + 2D) - (E + 2F)^2. \end{aligned} \tag{18}$$

Proof: Using the properties of the block matrix (note that $(\lambda - A + B)\boldsymbol{I}_{3\times3} - B\boldsymbol{J}_{3\times3}$ should be non-singular), the characteristic equation of $\boldsymbol{M}$ can be simplified through algebraic operations:

$$|\lambda \boldsymbol{I}_{6\times6} - \boldsymbol{M}| = (\lambda^2 - T_1\lambda + T_2)^2(\lambda^2 - T_3\lambda + T_4) = 0, \tag{19}$$

and the eigenvalues can thus be derived. ■

Following Lemma 4.1, the criteria of optimality can be expressed in terms of the eigenvalues:

$$\Phi(\boldsymbol{M}) = \begin{cases} |\boldsymbol{M}^{-1}| = \lambda_1^{-2}\lambda_2^{-2}\lambda_3^{-1}\lambda_4^{-1} = T_2^{-2}T_4^{-1}, & (20) \\ \operatorname{tr}(\boldsymbol{M}^{-1}) = 2\lambda_1^{-1} + 2\lambda_2^{-1} + \lambda_3^{-1} + \lambda_4^{-1} = 2\dfrac{T_1}{T_2} + \dfrac{T_3}{T_4}, & (21) \\ \lambda_{\max}(\boldsymbol{M}^{-1}) = \max(\lambda_1^{-1}, \lambda_2^{-1}, \lambda_3^{-1}, \lambda_4^{-1}), & (22) \end{cases}$$

where Equations (20)-(22) correspond to the D-, A- and E-optimality criteria, respectively, and minimizing them allows for the derivation of the optimal design. In the subsequent sections, we will systematically discuss three optimal criteria in detail.

4.1. D-Optimality

The D-optimal criterion aims to minimize the generalized variance of the estimators, i.e., $|\operatorname{Cov}(\hat{\boldsymbol{\beta}})|$, which is proportional to the determinant of $\boldsymbol{M}^{-1}$. Thus, the D-optimal orthogonal block design (denoted as ξ_D^*) in Ξ is obtained by maximizing $|\boldsymbol{M}|$. Recall that, we have set $b = 1 - a$, $c = 0$, and consequently, $|\boldsymbol{M}|$ can be simplified to a function of single variable a $(0 \leq a \leq \frac{1}{2})$, that is

$$\begin{aligned} |\boldsymbol{M}| &= T_2^2 T_4 \\ &= \left((A - B)(C - D) - (E - F)^2\right)^2 \\ &\quad \times \left((A + 2B)(C + 2D) - (E + 2F)^2\right) \\ &= 192a^{4r}(1 - a)^{4r}\left(a - \frac{1}{2}\right)^4\left((a - a^2)^r - \frac{3^{s-2r+1}}{2^s}\right)^2. \end{aligned} \tag{23}$$

Let $u = a - a^2$ $(0 \le u \le \frac{1}{4})$, and $t = \frac{3^{s-2r+1}}{2^s}$. Then we have

$$|\boldsymbol{M}(u)| = 192u^{4r}\left(u - \frac{1}{4}\right)^2 (u^r - t)^2. \tag{24}$$

The theorem below provides the necessary condition for ξ to be D-optimal.

Theorem 4.2: *The D-optimal parameter u_D^* must be obtained at the endpoints and critical points of $|\boldsymbol{M}(u)|$. Given values of the exponents r and s, u_D^* necessarily satisfies the following equation:*

$$r(3u^r - 2t)(1 - 4u) - 4u(u^r - t) = 0. \tag{25}$$

The solution to Equation (25) that maximizes $|\boldsymbol{M}(u)|$ is the D-optimal u. Consequently, ξ_D^* will be computed from $a_D^* = \frac{1}{2} - \sqrt{\frac{1}{4} - u_D^*}$.

Proof: $|\boldsymbol{M}(u)|$ is continuous and differentiable within $[0, \frac{1}{4}]$. Hence, its maximum value can either be found at the endpoints (0 and $\frac{1}{4}$) or the critical points where its derivative equals zero, i.e., $\frac{\partial |\boldsymbol{M}(u)|}{\partial u} = 0$. Note that, $|\boldsymbol{M}(u)|$ reaches its minimum, 0, at the endpoints $u = 0$ and $\frac{1}{4}$, so the D-optimal u must satisfy $\frac{\partial |\boldsymbol{M}(u)|}{\partial u} = 0$, which can be simplified as

$$u^{4r}\left(u - \frac{1}{4}\right)(u^r - t)(r(3u^r - 2t)(1 - 4u) - 4u(u^r - t)) = 0. \tag{26}$$

Equation (25) is derived after eliminating $u = 0$, $\frac{1}{4}$ and $t^{\frac{1}{r}}$, which maximize $|\boldsymbol{M}(u)|$. ■

Theorem 4.2 enables us to systematically compute the values of unknown parameters a for various combinations of r and s, which could be illustrated through an example provided below.

Example 4.3: D-optimal orthogonal block design for quadratic Scheffé polynomial. When $r = 1$ and $s = 0$, the SGBM turns into the quadratic Scheffé polynomial (3), and Equation (25) becomes

$$8u^2 - \frac{7}{2}u + \frac{1}{3} = 0. \tag{27}$$

The approximate solution to Equation (27) in $[0, \frac{1}{4}]$ is 0.1401, and upon verification, $u_D^* = 0.1401$ is the point that maximizes $|\boldsymbol{M}(u)|$. Accordingly, ξ is D-optimal when $a = 0.1685$,$b = 0.8315$ and $c = 0$, and this result is consistent with Czitrom (1988)'s work.

We have calculated the values of the D-optimal parameter a_D^* for certain specific combinations of r and s, and the results are presented in Table A1 in Appendix. We can observe that, when $0 < r \le 1$, with r held constant, a_D^* gradually increases as s increases, and similarly, with s held fixed, the values tend to gradually increase with the rise of r. When $r > 1$, the value of a_D^* locally increases with s, but the change in the value of a_D^* becomes more complex, and an intuitive pattern is no longer evident.

The D-optimal values of a in the orthogonal block designs for the three classic mixture models with process variable have been numerically collected in Table A1 and the results are presented in Figure 1.

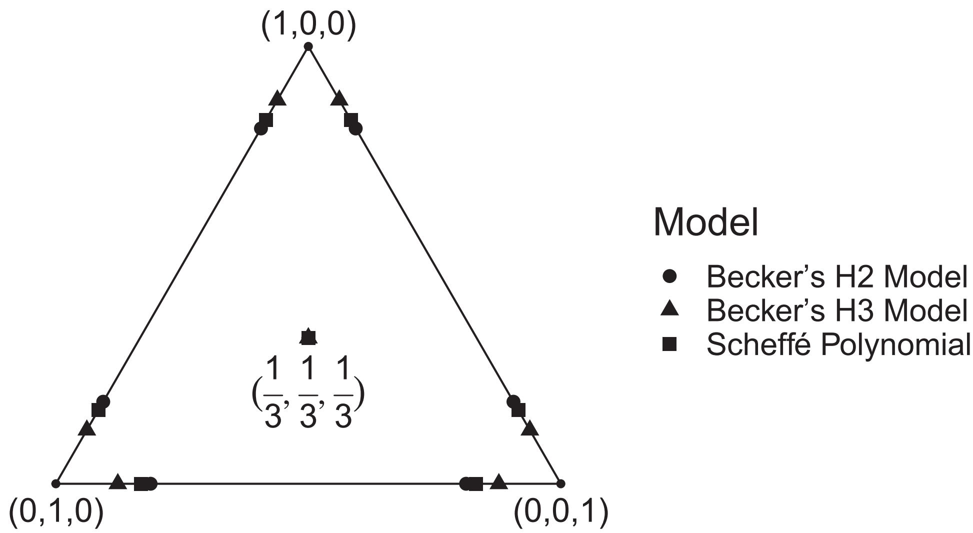

Figure 1. *D*-optimal orthogonal block designs for the classic mixture models.

4.2. A-optimality

The A-optimal criterion focuses on minimizing the average variance of the parameters to estimate, which is proportional to the trace of $\boldsymbol{M}^{-1}$, i.e.,

$$\begin{aligned}\mathrm{tr}(\boldsymbol{M}^{-1}) &= 2\frac{T_1}{T_2} + \frac{T_3}{T_4}\\ &= 2\frac{A-B+C-D}{(A-B)(C-D)-(E-F)^2} + \frac{A+2B+C+2D}{(A+2B)(C+2D)-(E+2F)^2}\\ &= 4\frac{(a(1-a))^{2r}-3a(1-a)+1}{3(a(1-a))^{2r}(2a-1)^2} + \frac{3(a(1-a))^{2r}+4+(\frac{3^{s-2r+1}}{2^s})^2}{2(a(1-a)^r-\frac{3^{s-2r+1}}{2^s})^2}.\end{aligned} \tag{28}$$

Let $u = a - a^2$, $t = \frac{3^{s-2r+1}}{2^s}$ $(0 \le u \le \frac{1}{4},\ u \ne t^{\frac{1}{r}})$. Then

$$\mathrm{tr}(\boldsymbol{M}^{-1}(u)) = 4\frac{u^{2r}-3u+1}{3u^{2r}(1-4u)} + \frac{3u^{2r}+4+t^2}{2(u^r-t)^2}. \tag{29}$$

A-optimal orthogonal block design will be obtained by the following necessary condition.

Theorem 4.4: *The A-optimal parameter u_A^* must be obtained at the critical points of* $\mathrm{tr}(\boldsymbol{M}^{-1}(u))$. *Given values of the exponents r and s, u_A^* necessarily satisfies the following equation:*

$$4(4u^{2r+1}-24ru^2+14ru+u-2r)(u^r-t)^3 - 3ru^{3r}(3tu^r+t^2+4)(1-4u)^2 = 0. \tag{30}$$

The solution to Equation (30) that minimizes $\mathrm{tr}(\boldsymbol{M}^{-1}(u))$ *is the A-optimal u. Consequently, ξ_A^* will be generated from $a_A^* = \frac{1}{2} - \sqrt{\frac{1}{4} - u_A^*}$.*

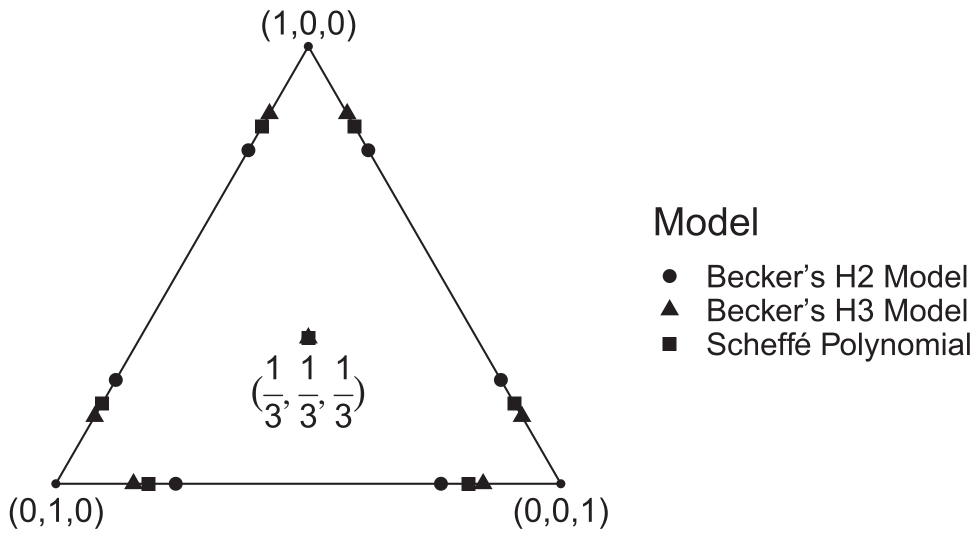

Figure 2. *A*-optimal orthogonal block designs for the classic mixture models.

Proof: The proof is similar to that of Theorem 4.2. Note that $\mathrm{tr}(\boldsymbol{M}^{-1}(u)) \to \infty$ when $u \to 0$ and $\frac{1}{4}$, and therefore, only the critical points could minimize $\mathrm{tr}(\boldsymbol{M}^{-1}(u))$. Thereby, Equation (30) is simplified from $\frac{\partial \mathrm{tr}(\boldsymbol{M}^{-1}(u))}{\partial u} = 0$. ■

Theorem 4.4 offers a structured approach to constructing A-optimal designs tailored to different exponent settings, and it could be directly illustrated through the following example.

Example 4.5: A-optimal orthogonal block design for Becker's H3 model.
For Becker's H3 model (5), we have $r = \frac{1}{2}$ and $s = 0$, so Equation (30) turns into

$$144u^4 + 304u^{\frac{7}{2}} - 264u^3 + 8u^{\frac{5}{2}} + 137u^2 - 169u^{\frac{3}{2}} + 40u + 24u^{\frac{1}{2}} - 8 = 0. \quad (31)$$

The approximate solution to Equation (31) for $0 < u < \frac{1}{4}$ is 0.1302, and upon verification, $u_A^* = 0.1302$ is the point that minimizes $\mathrm{tr}(\boldsymbol{M}^{-1}(u))$, and thereby the A-optimal design ξ_A^* has $(a = 0.1538, b = 0.8462, c = 0)$. This result is consistent with Aggarwal et al. (2002)'s work.

Under different combinations of specified r and s, a_A^*'s can be calculated. From Table A2 in Appendix, we can observe that, when $0 < r \le 1$, with r held constant, a_A^* increases with the growth of s, and the rate of increase gradually slows down. This is similar to the phenomenon demonstrated by the numerical results of D-optimality in Table A1. However, the difference lies in the fact that as r increases from around 0.9, the changes in a_A^* start to become complicate, making it difficult to describe the underlying patterns. Moreover, the critical values at which these changes occur are influenced by both r and s.

The A-optimal orthogonal block designs for the three classic mixture models have been included in Table A2 and are plotted in Figure 2. Comparing Figures 1 and 2, the sequence of the optimal a^* values for D- and A-optiamlity across the three models is consistent.

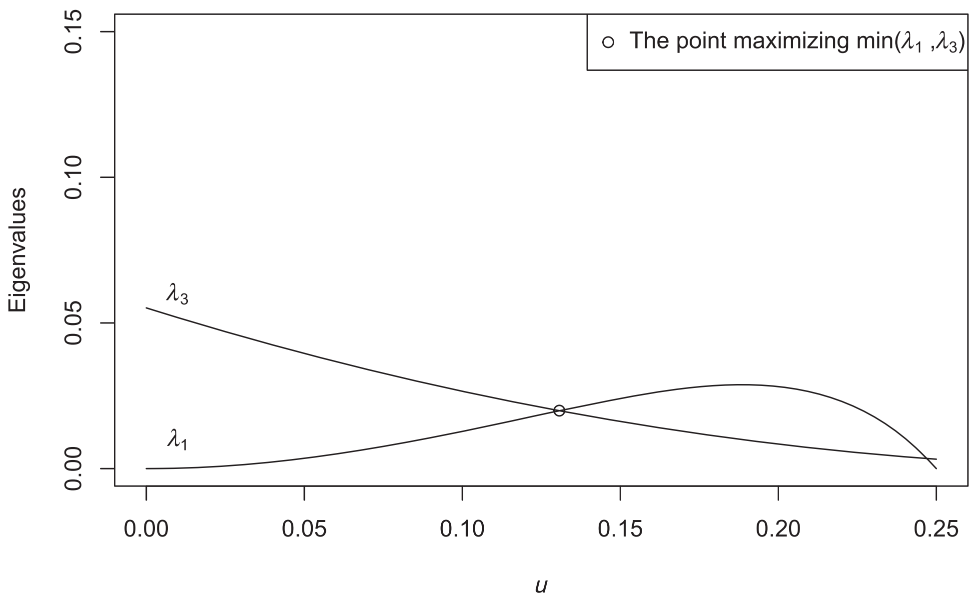

Figure 3. Graphs of λ_1 and λ_3 against u for quadratic Scheffé polynomial.

4.3. E-optimality

The E-optimal criterion aims to minimize the maximum variance of the estimators, which is equivalent to maximizing $\lambda_{\min}(\boldsymbol{M}) = \min(\lambda_1, \lambda_3)$ (for $\lambda_1 \le \lambda_2$ and $\lambda_3 \le \lambda_4$). Let $u = a - a^2$ $(0 \le u \le \frac{1}{4})$, $t = \frac{3^{s-2r+1}}{2^s}$, and we have

$$\lambda_1(u) = \frac{T_1 - \sqrt{T_1^2 - 4T_2}}{2} = u^{2r} - 3u + 1 - \sqrt{(u^{2r} + 3u - 1)^2 + u^{2r}}, \tag{32}$$

$$\lambda_3(u) = \frac{T_3 - \sqrt{T_3^2 - 4T_4}}{2} = u^{2r} + \frac{t^2 + 4}{3} - \sqrt{\left(u^{2r} + \frac{t^2 - 4}{3}\right)^2 + 4\left(u^r + \frac{t}{3}\right)^2}. \tag{33}$$

Whenever r and s are fixed, we need to compare λ_1 and λ_3 to find the point that maximizes $\lambda_{\min}(\boldsymbol{M})$. Two possible situations are illustrated below through two examples.

Example 4.6: E-optimal orthogonal block design for quadratic Scheffé polynomial. When $r = 1$ and $s = 0$, the SGBM turns into the quadratic Scheffé polynomial (3), and the graphs of $\lambda_1(u)$ and $\lambda_3(u)$ for $(0 \le u \le \frac{1}{4})$ are shown in Figure 3.

It can be clearly observed that, the E-optimal u_E^* occurs at the intersection of $\lambda_1(u)$ and $\lambda_3(u)$, that is, it satisfies

$$3u + \frac{10}{27} + \sqrt{(u^2 + 3u - 1)^2 + u^2} - \sqrt{\left(u^2 - \frac{35}{27}\right)^2 + 4\left(u + \frac{1}{27}\right)^2} = 0. \tag{34}$$

By solving Equation (34), we find that $u_E^* = 0.1307$ maximizes $\lambda_{\min}(\boldsymbol{M})$. Utilizing this value of u_E^*, we obtain the E-optimal design ξ_E^*, with $(a = 0.1546, b = 0.8454, c = 0)$. This result is consistent with Chan (1999)'s work.

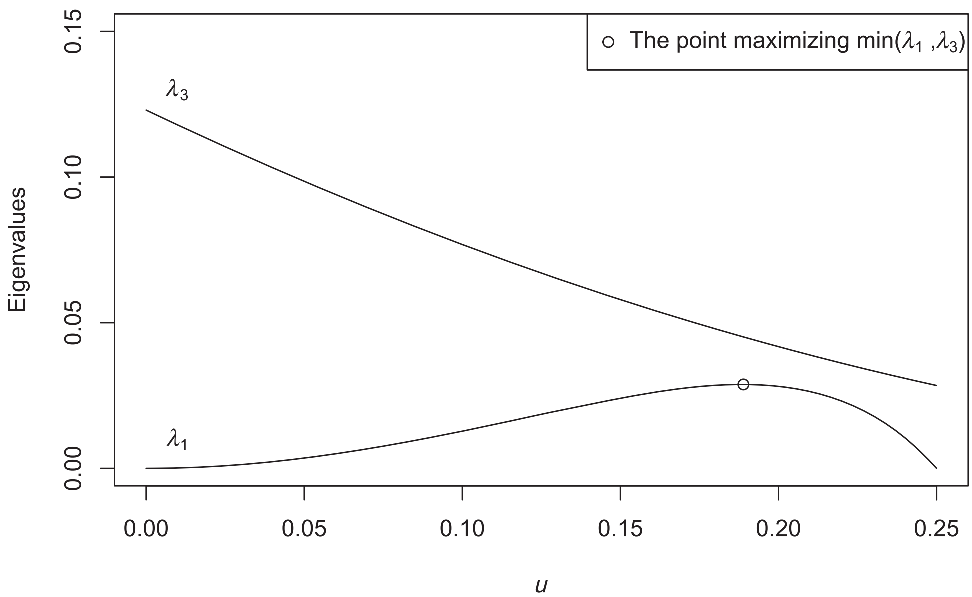

Figure 4. Graphs of λ_1 and λ_3 against u for Becker's H_2 model.

The alteration of s may lead to different outcomes. Let us proceed to the subsequent example.

Example 4.7: E-optimal orthogonal block design for Becker's H2 model. When it comes to Becker's H2 model (4), we have $r = 1$ and $s = 1$, and the graphs of $\lambda_1(u)$ and $\lambda_3(u)$ for $(0 \le u \le \frac{1}{4})$ are shown in Figure 4.

Figure 4 indicates that, $\lambda_1(u) < \lambda_3(u)$ always holds for $0 < u < \frac{1}{4}$, i.e., $\lambda_{\min}(\boldsymbol{M}) = \lambda_1(u)$. Differentiate $\lambda_1(u)$ with respect to u, and solve for $u \in [0, \frac{1}{4}]$ in the equation $\frac{\partial \lambda_1(u)}{\partial u} = 0$, which can be simplified as

$$2u - 3 - \frac{u + (2u+3)(u^2+3u-1)}{\sqrt{(u^2+3u-1)^2+u^2}} = 0. \tag{35}$$

Among the critical points and endpoints of $\lambda_1(u)$, it is found that $u_E^* = 0.1889$ maximizes $\lambda_{\min}(\boldsymbol{M})$. Utilizing this value of u_E^*, we obtain the E-optimal design ξ_E^*, corresponding to the E-optimal design ξ_E^* with $(a = 0.2527, b = 0.7473, c = 0)$. This result is consistent with Aggarwal et al. (2002)'s work.

Table A3 in Appendix displays the variations in the value of the E-optimal a_E^*'s under different combinations of r and s. Note that when r is fixed, several a_E^* values remain the same and do not vary with changes in s. This is because, as in Example 4.7, a_E^*'s are derived from the maximum points of λ_1 in (32), which are independent of s. Nonetheless, it is important to mention that the exponent s persistently shapes the curve of λ_3 in (33), consequently influencing the final value of the a_E^*'s.

The numerical pattern for E-optimality is completely different from those of D- and A-optimality. Additionally, the E-optimal orthogonal block designs for the classic mixture models in Figure 5 indicate that, the sequence of the optimal a^* values for E-optimality across the models does not align with those for D- and A-optimality.

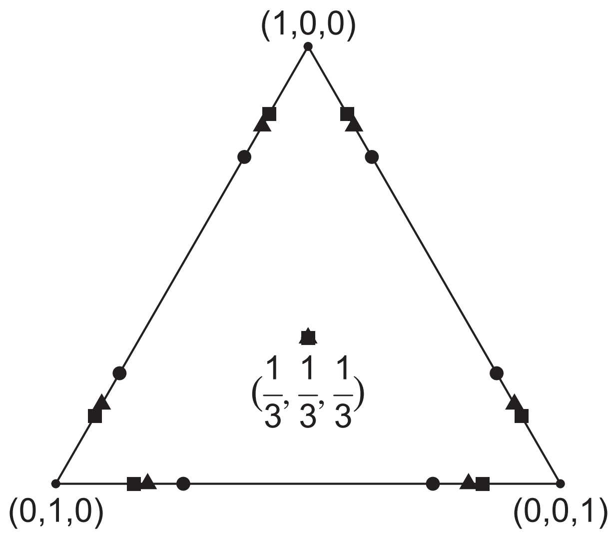

Figure 5. *E*-optimal orthogonal block designs for the classic mixture models.

5. Conclusions

In this paper, we propose the symmetric general blending model (SGBM) as a class of mixture models, on the basis of the structure of general blending models. We then introduce process variables into SGBM for practical application scenarios, and construct the orthogonal block designs to eliminate the influence of process variables on parameter estimation.

D-, *A*- and *E*-optimality criteria are considered to reduce the variance of coefficients in parameter estimation. The following are some potential future research directions for us. In previous research, orthogonal block designs were always set at the boundary of the simplex to obtain optimal designs (see, e.g., Aggarwal et al., 2013). We also impose such a constraint to facilitate the solution of optimality. It is difficult to prove that the optimal design indeed lies on the boundary (which is very likely influenced by the values of the exponents). Moreover, we are committed to extending optimal orthogonal block designs for SGBM in q components ($q \geq 4$), based on the construction (11) we provide and some feasible combinatorial design theories. See also, Wang et al. (2018) and Pang et al. (2022).

Necessary conditions with examples are provided in Section 4, offering optimality solution processes, to help obtain optimal designs for specified SGBM's more conveniently. Experimenters can also directly refer to the numerical results under various combinations of the exponents in the Appendix.

We finally describe and compare the trend of optimal design points changing with the exponents under three optimality criteria. The underlying patterns would benefit from further theoretical exploration.

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Disclosure statement

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Appendix. numerical results

Table A1. Values of a_D^* for SGBM's under different combinations of r and s.

	r													
s	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1	2	3	4	5
0	0.0353	0.0642	0.0876	0.1068	0.1227	0.1358	0.1467	0.1556	0.1628	0.1685	0.3546	0.3531	0.3652	0.3765
0.1	0.0358	0.0649	0.0886	0.1080	0.1240	0.1373	0.1484	0.1576	0.1650	0.1710	0.1794	0.3537	0.3654	0.3765
0.2	0.0362	0.0656	0.0895	0.1090	0.1253	0.1388	0.1501	0.1594	0.1671	0.1734	0.1830	0.3543	0.3655	0.3766
0.3	0.0366	0.0662	0.0903	0.1100	0.1264	0.1401	0.1516	0.1612	0.1691	0.1756	0.1866	0.3550	0.3657	0.3767
0.4	0.0370	0.0668	0.0911	0.1110	0.1275	0.1414	0.1531	0.1628	0.1710	0.1777	0.1903	0.3558	0.3659	0.3767
0.5	0.0373	0.0674	0.0918	0.1118	0.1286	0.1426	0.1544	0.1644	0.1727	0.1796	0.1939	0.3566	0.3662	0.3768
0.6	0.0377	0.0679	0.0924	0.1127	0.1295	0.1437	0.1557	0.1658	0.1743	0.1814	0.1974	0.3574	0.3664	0.3769
0.7	0.0380	0.0684	0.0931	0.1134	0.1304	0.1447	0.1568	0.1671	0.1758	0.1831	0.2010	0.3583	0.3666	0.3770
0.8	0.0382	0.0689	0.0937	0.1141	0.1312	0.1457	0.1579	0.1684	0.1772	0.1847	0.2045	0.3593	0.3669	0.3771
0.9	0.0385	0.0693	0.0942	0.1148	0.1320	0.1466	0.1590	0.1695	0.1786	0.1862	0.2079	0.3603	0.3672	0.3772
1	0.0388	0.0697	0.0948	0.1154	0.1328	0.1474	0.1599	0.1706	0.1798	0.1876	0.2113	0.3615	0.3674	0.3773
2	0.0406	0.0727	0.0986	0.1201	0.1382	0.1536	0.1669	0.1785	0.1886	0.1975	0.2393	0.3798	0.3714	0.3785
3	0.0417	0.0744	0.1009	0.1228	0.1412	0.1571	0.1708	0.1829	0.1935	0.2029	0.2556	0.2539	0.3789	0.3806
4	0.0424	0.0755	0.1022	0.1244	0.1431	0.1592	0.1732	0.1854	0.1963	0.2060	0.2642	0.2784	0.3952	0.3842
5	0.0428	0.0762	0.1031	0.1254	0.1443	0.1605	0.1746	0.1870	0.1980	0.2079	0.2689	0.2926	0.2819	0.3911

Table A2. Values of a_A^* for SGBM's under different combinations of r and s.

	r													
s	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1	2	3	4	5
0	0.0330	0.0732	0.1068	0.1334	0.1538	0.1686	0.1782	0.1832	0.1845	0.1833	0.4012	0.3732	0.3734	0.3791
0.1	0.0346	0.0752	0.1091	0.1363	0.1573	0.1730	0.1837	0.1897	0.1917	0.1908	0.4052	0.3743	0.3737	0.3792
0.2	0.0360	0.0770	0.1112	0.1387	0.1603	0.1768	0.1884	0.1955	0.1985	0.1981	0.4096	0.3754	0.3740	0.3794
0.3	0.0372	0.0785	0.1129	0.1408	0.1629	0.1800	0.1925	0.2006	0.2047	0.2051	0.1702	0.3766	0.3744	0.3795
0.4	0.0383	0.0798	0.1145	0.1425	0.1650	0.1827	0.1959	0.2051	0.2102	0.2116	0.1746	0.3779	0.3748	0.3796
0.5	0.0392	0.0809	0.1158	0.1440	0.1668	0.1849	0.1989	0.2089	0.2151	0.2176	0.1792	0.3792	0.3751	0.3797
0.6	0.0400	0.0819	0.1169	0.1453	0.1684	0.1869	0.2013	0.2121	0.2192	0.2229	0.1840	0.3806	0.3755	0.3799
0.7	0.0407	0.0828	0.1178	0.1464	0.1697	0.1885	0.2034	0.2148	0.2228	0.2274	0.1888	0.3821	0.3760	0.3800
0.8	0.0414	0.0836	0.1187	0.1474	0.1708	0.1898	0.2051	0.2170	0.2257	0.2314	0.1939	0.3837	0.3764	0.3801
0.9	0.0419	0.0842	0.1194	0.1482	0.1717	0.1910	0.2065	0.2189	0.2282	0.2347	0.1991	0.3854	0.3769	0.3803
1	0.0424	0.0848	0.1200	0.1489	0.1725	0.1919	0.2077	0.2204	0.2303	0.2374	0.2045	0.3872	0.3774	0.3805
2	0.0452	0.0880	0.1234	0.1524	0.1764	0.1964	0.2131	0.2271	0.2389	0.2488	0.2650	0.4129	0.3845	0.3826
3	0.0462	0.0891	0.1244	0.1535	0.1775	0.1975	0.2143	0.2285	0.2406	0.2508	0.3000	0.2526	0.3971	0.3863
4	0.0467	0.0896	0.1249	0.1539	0.1779	0.1979	0.2147	0.2289	0.2410	0.2513	0.3060	0.3043	0.4205	0.3927
5	0.0469	0.0898	0.1251	0.1540	0.1780	0.1980	0.2148	0.2290	0.2411	0.2514	0.3070	0.3286	0.2839	0.4044

Table A3. Values of a_E^* for SGBM's under different combinations of *r* and *s*.

	r													
s	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1	2	3	4	5
0	0.1542	0.0938	0.1297	0.1585	0.1820	0.1733	0.1667	0.1618	0.1578	0.1546	0.1398	0.3927	0.3660	0.3627
0.1	0.0337	0.0938	0.1297	0.1585	0.1820	0.2010	0.1844	0.1752	0.1688	0.1638	0.1435	0.3946	0.3671	0.3627
0.2	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2087	0.1914	0.1813	0.1741	0.1472	0.3966	0.3682	0.3627
0.3	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2123	0.1959	0.1856	0.1511	0.3986	0.3694	0.3627
0.4	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2140	0.1989	0.1552	0.4007	0.3706	0.3627
0.5	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2391	0.2147	0.1594	0.4028	0.3718	0.3627
0.6	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2428	0.2349	0.1638	0.4050	0.3731	0.3627
0.7	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2428	0.2527	0.1684	0.4073	0.3744	0.3627
0.8	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2428	0.2527	0.1731	0.4096	0.3757	0.3627
0.9	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2428	0.2527	0.1781	0.4120	0.3771	0.3627
1	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2428	0.2527	0.1833	0.4145	0.3785	0.3627
2	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2428	0.2527	0.4462	0.1928	0.3949	0.3679
3	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2428	0.2527	0.3074	0.2371	0.4162	0.3806
4	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2428	0.2527	0.3074	0.3142	0.2295	0.3969
5	0.0488	0.0938	0.1297	0.1585	0.1820	0.2014	0.2176	0.2312	0.2428	0.2527	0.3074	0.3333	0.2731	0.4179