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ORIGINAL RESEARCH

Shrinkage simplex-centroid designs for a quadratic mixture model

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Abstract A simplex-centroid design for q mixture components comprises of all possible subsets of the q components, present in equal proportions. The design does not contain full mixture blends except the overall centroid. In real-life situations, all mixture blends comprise of at least a minimum proportion of each component. Here, we introduce simplex-centroid designs which contain complete blends but with some loss in D-efficiency and stability in G-efficiency. We call such designs as shrinkage simplex-centroid designs. Furthermore, we use the proposed designs to generate component-amount designs by their projection.

Keywords Mixture experiment · Simplex-centroid design · D-optimality · G-optimality

Introduction

In a mixture experiment with q components, the proportion of ingredients may be denoted by $x_1, x_2, \ldots, x_q$, where $x_i \geq 0$ for $i = 1, 2, \ldots, q$ and $\sum_{i=1}^{q} x_i = 1$. The response depends only on the mixture and not on the total amount of the mixture. The factor space is a $(q-1)$-dimensional regular simplex S_{q-1}:

$$S_{q-1} = \left\{ x : (x_1, x_2, \ldots, x_q) \middle| \sum_{i=1}^{q} x_i = 1, x_i \geq 0 \right\}.$$

There are various mixture models available in the literature, and for the estimation of their parameters, many mixture designs have been proposed. Simplex-centroid design is the simplest and widely used mixture design by the practitioners. The main feature of this article is the construction of simplex-centroid mixture design with the real mixture blends, though with less efficiency. The design points in a simplex-centroid design do not completely explore the whole mixture region, whereas in our proposed designs, the design points fall uniformly inside the mixture space and explore it in true sense. Prescott (1998) shrunk the co-ordinates of design points towards the centroid after their re-parameterization, using orthogonally blocked mixture designs. The resulting designs were composed of full mixture blends, falling inside the simplex. The designs were less efficient as compared to the original design.

The article is organized as follows. First simplex-centroid design is reviewed. Next, the concept of shrinkage design and the re-parameterization of co-ordinate system for three and fours components are discussed. Then, the shrinkage simplex-centroid designs for three and four components are constructed and their D- and G-efficiencies are compared. The proposed designs are further used to develop component-amount designs by their projection. Finally, the application of shrinkage design is given and conclusions are made.

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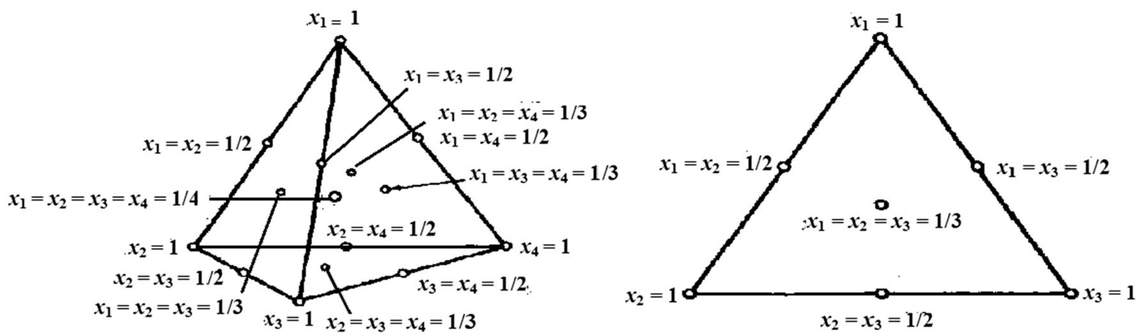

Fig. 1 Simplex-centroid designs for three and four components

Table 1 Simplex-centroid design for three components

Run	x_1	x_2	x_3
1	1	0	0
2	0	1	0
3	0	0	1
4	1/2	1/2	0
5	1/2	0	1/2
6	0	1/2	1/2
7	1/3	1/3	1/3

Review of simplex-centroid design

Scheffé (1963) introduced the simplex-centroid design, where only such mixtures are considered in which the components presented have equal proportions. It has $2^q - 1$ design points. These design points have $\overset{q}{\underset{1}{C}}$ permutations of $(1, 0, \ldots, 0)$, the $\overset{q}{\underset{2}{C}}$ permutations of $\left(\frac{1}{2}, \frac{1}{2}, 0, \ldots, 0\right)$, the $\overset{q}{\underset{3}{C}}$ permutations of $\left(\frac{1}{3}, \frac{1}{3}, \frac{1}{3}, 0, \ldots, 0\right)$, and so on, and the overall centroid $\left(\frac{1}{q}, \frac{1}{q}, \frac{1}{q}, \ldots, \frac{1}{q}\right)$. Such mixtures are located at the centroid of the $(q-1)$-dimensional simplex and the centroids of all the lower dimensional simplexes within the $(q-1)$-dimensional simplex. Responses are collected at the design points and a polynomial is fitted. The design points in the simplex-centroid design will support the polynomial equation. The general form of the quadratic Scheffé polynomial equation is

$$E(Y) = \sum_{i=1}^{q} \beta_i X_i + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \beta_{ij} X_i X_j.$$

Simplex-centroid designs for three and four components are given in Tables 1 and 2, and are depicted by Fig. 1a and b.

In real-life situations, we always use a mixture that has at least a minimum proportion of all the ingredients present. Therefore, we need to construct the optimal designs with complete mixture blends. The standard simplex design is a boundary point design with the exception of the overall centroid that is all the other points are on the boundary of the simplex. We propose the construction of three and four components' simplex-centroid designs by the shrinkage of design points towards its centroid and call them shrinkage simplex-centroid designs.

Table 2 Simplex-centroid design for four components

Run	x_1	x_2	x_3	x_4
1	1	0	0	0
2	0	1	0	0
3	0	0	1	0
4	0	0	0	1
5	1/2	1/2	0	0
6	1/2	0	1/2	0
7	1/2	0	0	1/2
8	0	1/2	1/2	0
9	0	1/2	0	1/2
10	0	0	1/2	1/2
11	1/3	1/3	1/3	0
12	1/3	1/3	0	1/3
13	1/3	0	1/3	1/3
14	0	1/3	1/3	1/3
15	1/4	1/4	1/4	1/4

Shrinkage design

The pioneering work on this issue was done by Prescott (1998). He constructed nearly optimal designs for Scheffé's quadratic mixture model with three and four components, using Latin square-based orthogonal blocking scheme. Such optimal designs although have complete mixture compositions, but are less efficient. Aggarwal et al. (2011) proposed nearly D-, A-, and E-optimal designs for Scheffé's, Kronecker, Becker's, and Darroch and Waller's quadratic mixture models in four components, using F-square-based orthogonal blocking scheme. Hasan and

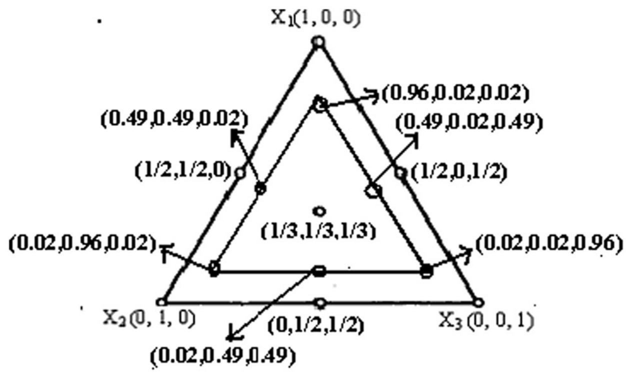

Fig. 2 Simplex-centroid design and shrinkage simplex-centroid design with $s = 0.05$

Khan (2011, 2012) proposed nearly D-, A-, and E-optimal orthogonally blocked designs in three components using Scheffé's and Kronecker mixture models. All the above references deal with the construction of shrinkage designs in orthogonal blocks. We have adapted the same methodology for the construction of shrinkage optimal design using simplex-centroid design, where the mixture blends are not in orthogonal blocks.

Re-parameterization of the co-ordinate system

Consider a mixture region in $(q - 1)$ dimensions and denote the co-ordinates in the region by the symbols $a, b, c, \ldots$, such that $a + b + c + \cdots = 1$. The orthogonal blocks containing pairs of Latin squares provide mathematical expression for the information matrix $X'X$ in terms of the symbols $a, b, c, \ldots$ Using any optimal criteria, we can determine the optimal values of a, b, $c, \ldots$ Prescott (1998) discussed the re-parameterization of co-ordinates of points in a mixture region and constructed nearly D-optimal designs. We review this re-parameterization method for the construction of Shrinkage simplex-centroid design in three and four components.

(1) Three components' mixture

Consider a two-dimensional simplex formed by three components, as given in Fig. 2. Take any design point P (a, b, c) in the simplex, such that $a \geq b \geq c$. Express the co-ordinates a, b, and c in terms of s and f, where s is the shrinkage parameter with f be a co-ordinate of the point on the edge of simplex, and (1/3, 1/3, 1/3) is the centroid of the simplex:

$$
\begin{aligned}
a &= (1 - s)f + \frac{s}{3} \\
b &= (1 - s)(1 - f) + \frac{s}{3} \\
c &= \frac{s}{3}.
\end{aligned}
\tag{1}
$$

For instance, when $s = 0$, the point P (a, b, c) reduces to P (f, $1 - f$, 0, 0), falling on the edge of the simplex.

(2) Four components' mixture

We re-parameterize a point P with the co-ordinates (a, b, c, d) in a three-dimensional simplex, where $a \geq b \geq c \geq d$. As a special case when $c = d$, the co-ordinates (a, b, c, d) of the point P, expressed as a function of (f, s), are given as follows:

$$
\begin{aligned}
a &= (1 - s)f + s/4 \\
b &= (1 - s)(1 - f) + s/4 \\
c &= d = s/4.
\end{aligned}
\tag{2}
$$

The extension of re-parameterization to mixture experiments with more than four components is very simple.

Shrinkage simplex-centroid designs for three and four components

We construct shrinkage simplex-centroid design by considering quadratic Scheffé's mixture models in three and four components. The D-efficiency of shrinkage designs is obtained with regard to the D-criterion, where $D = |X'X|^{1/P}$ and P denotes the number of parameters in the model:

$$\text{D-efficiency} = |X'X|^{1/P} / |X'X|_o^{1/P} \times 100. \tag{3}$$

Here, $|X'X|_o^{1/P}$ is the value of D for non-shrinkage design. Furthermore, the prediction capability of a design can be assessed using G-optimality criterion, which searches for the design that minimizes the maximum prediction variance over the experimental region. A design criterion related to G-optimality is G-efficiency, defined as

$$\text{G-efficiency} = 100 \times \left(\frac{p/n}{\text{MPV}} \right), \tag{4}$$

where p/n is the average prediction variance (APV) and MPV is the maximum prediction variance. We evaluate D- and G-efficiencies of shrinkage designs for several values of parameter s, for a quadratic Scheffé's model. The three components' quadratic Scheffé's model is

$$
\begin{aligned}
E(y) = {} & \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 \\
& + \beta_{23} x_2 x_3.
\end{aligned}
\tag{5}
$$

The value of D for simplex-centroid design, given in Table 1, for three component quadratic Scheffé's model is 0.27049. Now, consider that the shrinkage parameter $s = 0.05$. Using re-parameterized co-ordinates given in Eq. (1), the shrinkage simplex-centroid design is obtained as follows.

Table 3 Shrinkage simplex-centroid design with the parameter $s = 0.05$

Run	x_1	x_2	x_3
1	0.96	0.02	0.02
2	0.02	0.96	0.02
3	0.02	0.02	0.96
4	0.49	0.49	0.02
5	0.49	0.02	0.49
6	0.02	0.49	0.49
7	1/3	1/3	1/3

Table 4 *D*-efficiency of shrinkage simplex-centroid designs for $q = 3$

s	$\lvert X'X\rvert^{1/p}$	$\lvert X'X\rvert_0^{1/p}$	*D*-efficiency
0.00	0.270492	0.270492	100
0.05	0.229272	0.270492	85
0.10	0.201085	0.270492	74

Table 5 *G*-efficiency of shrinkage simplex-centroid designs for $q = 3$

s	APV	MPV	*G*-efficiency
0.00	0.857	0.992	86.4
0.05	0.857	0.992	86.4
0.10	0.857	0.992	86.4

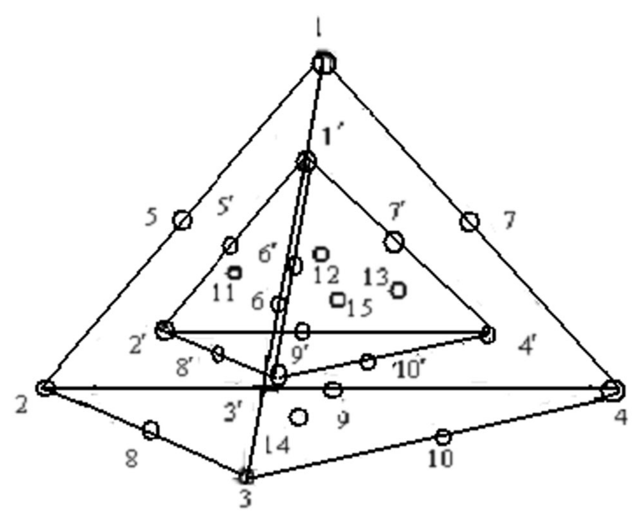

Fig. 3 Simplex-centroid design and shrinkage simplex-centroid design for $q = 4$ with $s = 0.05$ (simple number shows runs in simplex-centroid design and the numbers with primes show runs in shrinkage simplex-centroid design)

The value of D for shrinkage simplex-centroid design given in Table 3 for quadratic Scheffé's model is 0.2293. Using *D*-efficiency criterion given in Eq. (3), the efficiency of the above design is 85%. Comparing the design in Table 3 with the simplex-centroid design in Table 1, it is clear that the constructed design is a true mixture design with full mixture blends, though less efficient. Table 4 gives *D*-efficiency of shrinkage simplex-centroid designs for several values of s. In Table 5, we have *G*-efficiency of the designs for different values of shrinkage parameter s (Fig. 3).

Table 6 Shrinkage simplex-centroid design for four components with $s = 0.05$

Run	x_1	x_2	x_3	x_4
1	0.9625	0.0125	0.0125	0.0125
2	0.0125	0.9625	0.0125	0.0125
3	0.0125	0.0125	0.9625	0.0125
4	0.0125	0.0125	0.0125	0.9625
5	0.4875	0.4875	0.0125	0.0125
6	0.4875	0.0125	0.4875	0.0125
7	0.4875	0.0125	0.0125	0.4875
8	0.0125	0.4875	0.4875	0.0125
9	0.0125	0.4875	0.0125	0.4875
10	0.0125	0.0125	0.4875	0.4875
11	0.32915	0.32915	0.3215	0.0125
12	0.32915	0.32915	0.0125	0.32915
13	0.32915	0.0125	0.32915	0.32915
14	0.0125	0.32915	0.32915	0.32915
15	0.2500	0.2500	0.2500	0.2500

Table 7 *D*-efficiency of shrinkage simplex-centroid designs for $q = 4$

s	$\lvert X'X\rvert^{1/p}$	$\lvert X'X\rvert_0^{1/p}$	*D*-efficiency
0.00	0.232169	0.232169	100
0.05	0.179471	0.232169	77.3
0.10	0.169251	0.232169	73.0

Table 8 *G*-efficiency of shrinkage simplex-centroid designs for $q = 4$

s	APV	MPV	*G*-efficiency
0.00	0.667	0.978	68.2
0.05	0.667	0.977	68.2
0.10	0.667	0.977	68.2

Next, we consider four components' quadratic Scheffé mixture model:

$$E(y) = \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_4 x_4 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{14} x_1 x_4 + \beta_{23} x_2 x_3 + \beta_{24} x_2 x_4 + \beta_{34} x_3 x_4. \quad (6)$$

The value of D for simplex-centroid design, given in Table 2, for four components' quadratic Scheffé's model is 0.2322. Now, shrink design in Table 2 is towards the centroid, with the shrinkage parameter $s = 0.05$ and reparameterizes the co-ordinates using Eq. (2). The resulting shrinkage simplex-centroid design is given as follows.

The value of D for shrinkage simplex-centroid design, given in Table 6, for quadratic Scheffé's model is 0.1795. Using D-efficiency criterion given in Eq. (3), the efficiency of the above design is 77.3%. Tables 7 and 8 provide D- and G-efficiencies, respectively, for shrinkage simplex-centroid designs with several values of s.

Hence, comparing the design in Table 6 with the simplex-centroid design in Table 2, the constructed design is a true mixture design, containing full mixture blends with some compromise on its efficiency.

Construction of component-amount design via projection of shrinkage simplex-centroid design

A mixture-amount experiment is a type of mixture experiment that is performed at two or more levels of total amount. The response is assumed to be dependent upon the individual proportions of components in the blend and also on its amount. The effect on the response after varying mixture component proportions and the total amount of the mixture is measured by fitting a mixture-amount model to the design. The design for fitting mixture-amount model is called mixture-amount design, developed by Piepel and Cornell (1985). Piepel (1988) modified that model to accommodate zero-amount condition. The alternative model uses the actual amounts of the ingredients denoted by $a_1, a_2, \ldots, a_q$, such that $a_1 + a_2 + \cdots + a_q = A$. The proportions x_i are related to the amount a_i through $x_i = a_i/A$. This is called component-amount model:

$$E(y) = \alpha_0 + \sum_{i=1}^{q} \alpha_i a_i + \sum_{i=1}^{q} \left(\alpha_{ii} a_i^2 + \sum_{i\langle j}^{q} \alpha_{ij} a_i a_j \right). \qquad (7)$$

Prescott and Draper (2004, 2008) discussed the construction of designs for component-amount models by projecting standard symmetric mixture designs (simplex-lattice and simplex-centroid designs) in lower dimensions. The method involved the collapsing of the standard symmetric mixture designs by the removal of their one or more columns. It leads to a set of symmetric designs for each of the several levels of the amount A. We construct component-amount designs by the projection of shrinkage simplex-centroid designs, as given in "Shrinkage design".

The component-amount design formed by Prescott and Draper (2004, 2008), through projection of simplex-centroid design, given in Table 2, is provided in Table 9.

The above component-amount design has the value $D = 0.21825$ and its G-efficiency is 68.2%. The design is composed of incomplete mixture blends with different levels of amounts except the centroid point as a complete mixture blend. For real-life situations, we need component-amount designs which have full mixture blends with at least a minimum amount of each component in the mixture. Such designs can be obtained by the projection of shrinkage simplex-centroid designs.

We project the design, given in Table 6, to lower dimension by deleting any column. Let the column for x_4 in Table 6 is deleted. The resulting component-amount design in three components has the levels of amount $A = 0.0375$, 0.5125, 0.6708, 0.9875, and 0.7500 with replicates 1, 3, 3, 7, and 1 and is given in Table 10. This shrinkage

Table 9 Simplex-centroid design for four components

Run	a_1	a_2	a_3	A
1	1	0	0	1
2	0	1	0	1
3	0	0	1	1
4	0	0	0	0
5	1/2	1/2	0	1
6	1/2	0	1/2	1
7	1/2	0	0	1/2
8	0	1/2	1/2	1
9	0	1/2	0	1/2
10	0	0	1/2	1/2
11	1/3	1/3	1/3	1
12	1/3	1/3	0	2/3
13	1/3	0	1/3	2/3
14	0	1/3	1/3	2/3
15	1/4	1/4	1/4	3/4

Table 10 Shrinkage simplex-centroid component-amount design for three components

Run	a_1	a_2	a_3	A
1	0.9625	0.0125	0.0125	0.9875
2	0.0125	0.9625	0.0125	0.9875
3	0.0125	0.0125	0.9625	0.9875
4	0.0125	0.0125	0.0125	0.0375
5	0.4875	0.4875	0.0125	0.9875
6	0.4875	0.0125	0.4875	0.9875
7	0.4875	0.0125	0.0125	0.5125
8	0.0125	0.4875	0.4875	0.9875
9	0.0125	0.4875	0.0125	0.5125
10	0.0125	0.0125	0.4875	0.5125
11	0.32915	0.32915	0.32915	0.9875
12	0.32915	0.32915	0.0125	0.6708
13	0.32915	0.0125	0.32915	0.6708
14	0.01250	0.32915	0.32915	0.6708
15	0.2500	0.2500	0.2500	0.7500

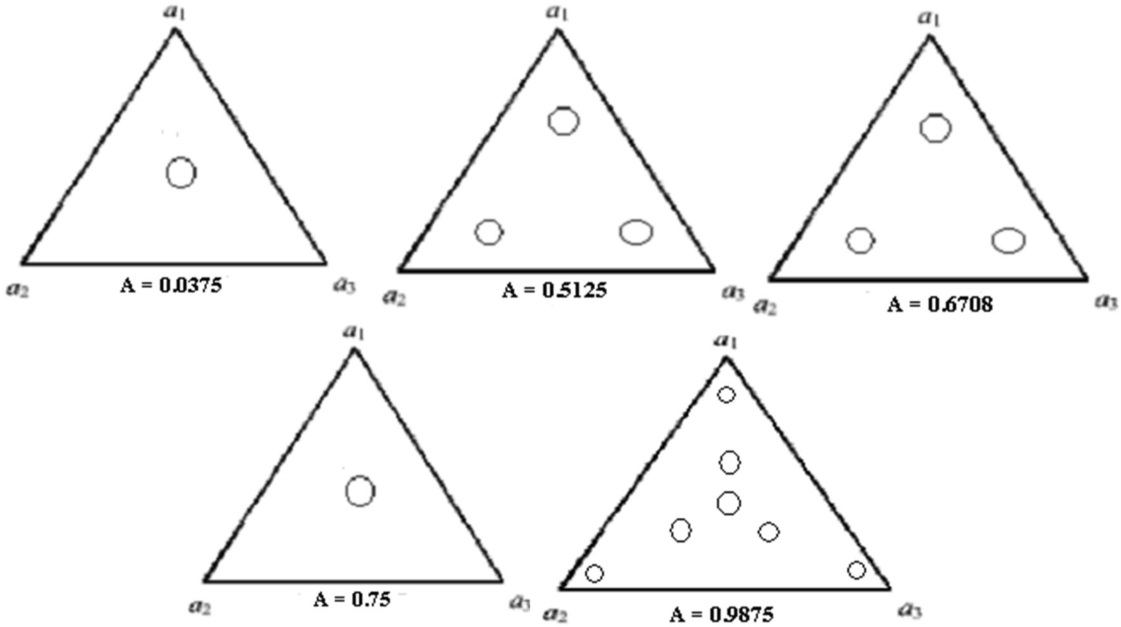

Fig. 4 Structure for the component-amount design in a_1, a_2, and a_3 simplex, formed by the projection of shrinkage simplex-centroid design with the shrinkage parameter $s = 0.05$

component-amount design has the *D*-efficiency 91.2% and the *G*-efficiency design 68.2%. On comparison of this design with the design in Table 9, it can be noted that the constructed design is composed of full mixture blends, although it has less efficiency (Fig. 4).

Example: diazepam solubility experiment

We take an example, given in Smith (Smith 2005, p. 54). Belloto et al. (1985) were interested in studying the solubility of drug diazepam in mixtures of ethanol, propylene glycol, and water. It was desired to develop a mixture model that would predict the diazepam in any mixture of solvents in a three-dimensional simplex. We first choose simplex-centroid design given in Table 1 and augment it with two replicates at each of the pure blends and with three axial check blends.

The *G*-efficiency of the design is 63.14%. We shrink augmented simplex-centroid design, given in Table 11, using the shrinkage parameter $s = 0.1$. The *G*-efficiency of the design is again 63.14%, while its *D*-efficiency is 74.4%. Therefore, mixture design with full mixture blends can be used to develop a mixture model with the stable *G*-efficiency that would predict the diazepam in any mixture of solvents in a three-dimensional simplex (Fig. 5).

Table 11 Augmented simplex-centroid design for three components

Run	x_1 Ethanol	x_2 Glycol	x_3 Water	y Solubility (mg/ml)
1	1	0	0	27.8
2	0	1	0	7.42
3	0	0	1	0.048
4	1/2	1/2	0	27.0
5	1/2	0	1/2	6.02
6	0	1/2	1/2	0.61
7	4/6	1/6	1/6	28.0
8	1/6	4/6	1/6	13.0
9	1/6	1/6	4/6	0.408
10	1/3	1/3	1/3	9.52
11	0	1	0	7.42
12	1	0	0	27.8
13	0	0	1	0.048

Conclusion

Simplex-centroid design is a widely used mixture design for quadratic mixture model. This classical design does not contain full mixture blends except centroid. In real-life situation, a mixture always has a least proportion of every component. In the current study, shrinkage designs are

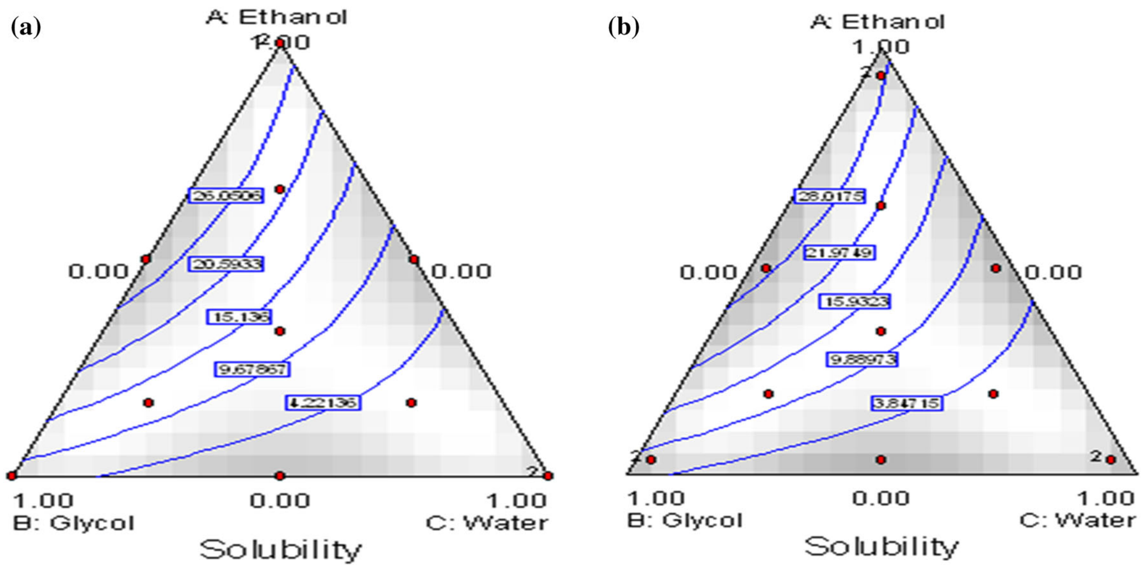

Fig. 5 **a** Contour plot of predicted solubility using the design in Table 11 and **b** contour plot of predicted solubility using shrinkage design with parameter $s = 0.1$

constructed from simplex-centroid designs. The proposed shrinkage simplex-centroid designs are the real-life mixture designs, although they have less efficiency as compared to the simplex-centroid designs available in the literature. This stability in *G*-efficiency of these designs shows that the prediction capability of the model does not change by shrinking conventional simplex-centroid design towards the centroid. Therefore, compromising on the loss in D-optimality of simplex-centroid design, mixture designs with full mixture blends and with the stable G-optimality can be constructed.

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