Orthogonalization of Mixture Coordinates Using a Generalized Linear Transform

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ABSTRACT

Orthogonal and rotatable experimental designs have many well known advantages for experimental investigation. Such advantages include low computational cost, independently estimable effects, and a well known error structure and associated statistics. Well known examples of orthogonal and rotatable designs include factorial designs, central composites, and Box-Behnken designs. By way of contrast, mixture designs are generally non-orthogonal and non-rotatable. It is possible to orthogonalize a portion of the mixture space; however, for gaseous fuel blends much or all of the mixture space is relevant. Subsequently, it may be possible to apply a non-linear transformation to larger portions of the mixture, but non-linear transformations distort the error structure and complicate statistical tests. Therefore, a linear transformation that maps the entire mixture region to an orthogonal and rotatable design space (and vice-versa) is desired. This paper presents details of such a transformation applicable to any number of factors.

Introduction

Combustion experiments with gaseous fuels often result in mixture-amount or mixture-process-variable (MPV) experiments where most or all of the mixture space is relevant. We first overview essential aspects of mixture and factorial designs and then present a transformation matrix that maps one to the other. We demonstrate the method with application to a four-component mixture. We shall find that for any q-component mixture there are $(q - 1)!$ linear transformations that may be applied piecewise to collectively map the entire mixture space to an orthogonal factor space. All such piecewise maps may be generated via permutation matrices from a single matrix. Even better, a single transformation matrix may be used to map the entire mixture or factor space in conjunction with a straightforward reordering of the components or factors. As illustrations, we use the reordering method to map four-component mixture designs from various three-factor orthogonal and rotatable designs. The mapped designs comprise a factorial design, a central composite design, and a Box-Behnken design. We also show some results for a MPV design having two independent factors and three mixture components.

Background

Combustion equipment designed for refineries must operate on a wide variety of gaseous fuel compositions (termed *refinery fuel gas* or RFG). RFG comprises whatever gaseous byproducts the refinery cannot process to more profitable ends; this RFG is then used to displace other fuels and fire combustion equipment used to refine crude or create petrochemicals. Since different refineries process a wide variety of crudes for many end-use markets, there are as many RFG compositions as there are refineries; indeed, a single refinery often hosts several different RFGs. RFG comprises mostly saturated hydrocarbons (typically those having one to four carbons), hydrogen, minor amounts of diluents, and trace amounts of other species. A 20-component blend is not unusual.

The combustion reaction may be thought of as occurring in two broad steps. In the first step – *pyrolysis* – heat from the combustion reaction dissociates fuel into hydrogen and hydrocarbon fragments. In the second step – *oxidation* – these fuel fragments are then completely burned with air to form carbon dioxide and water vapor and trace unwanted emissions of NOx (10 to 100 ppm) and CO (~0 ppm, but higher under start-up conditions). Because oxidation chemistry is similar for all hydrogen/hydrocarbon blends, it is possible to simulate a complex RFG using three or four components so long as one matches the number and kinds of bonds; i.e., H–H, C–H, and C–C bonds – the so-called *equivalent bond method* (Colannino 2006). Such reduced-component mixtures do a good job of simultaneously matching salient fuel properties including heating value, stoichiometric oxygen requirement, specific gravity, pressure-flow characteristics, and emissions potentials.

At a minimum, combustion performance is a function of furnace operating temperature, ultimate oxygen concentration in the furnace – so called *excess oxygen*, and fuel composition. This minimal set represents two independent factors (temperature and oxygen) and fuel mixtures of one to three mixture components (e.g., hydrogen, natural gas, and propane). Indeed, designers of combustion equipment must guarantee such performance, so accurate models are a must.

Combustion reactions are complex: they comprise many more than 100 elemental reactions leading to final products; and such reactions represent a highly non-linear system within a complex turbulent flow field having simultaneous momentum, heat, and mass transfer. This complex pathway simply cannot be modeled with sufficient accuracy. A practical approach for predicting emissions such as NOx is to fit a simplified model with adjustable parameters by performing MPV experiments and then fitting a semi-empirical equation. However, even the simplest applicable MPV experiment can be cumbersome. For example, measuring binary blends and factor interactions for a ternary mixture embedded in a 2^2 factorial (Figure 1) leads to an equation with potentially 28 adjustable parameters (Equation 1).

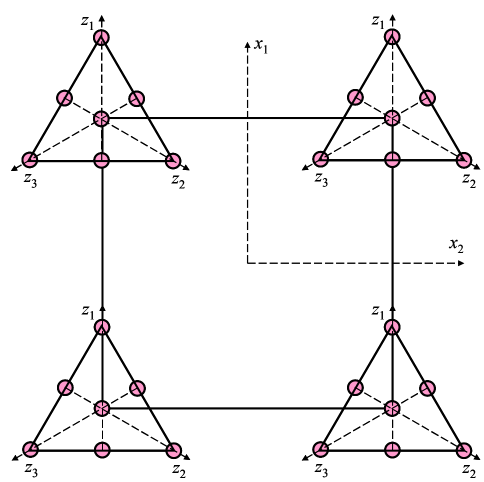

Figure 1, an MPV experimental design. The design represents the simplest mixture-process-variable experiment comprising interactions between two independent factors (x_1 and x_2) and three mixture components (z_1, z_2, and z_3).

$$\ln[\mathrm{NOx}] = \begin{array}{l} a_0 + b_1 z_1 + b_2 z_2 + b_3 z_3 + b_{12} z_1 z_2 + b_{13} z_1 z_3 + b_{23} z_2 z_3 + \\ a_1 x_1 + c_{1,1} x_1 z_1 + c_{1,2} x_1 z_2 + c_{1,3} x_1 z_3 + c_{1,12} x_1 z_1 z_2 + c_{1,13} x_1 z_1 z_3 + c_{1,23} x_1 z_2 z_3 + \\ a_2 x_2 + c_{2,1} x_2 z_1 + c_{2,2} x_2 z_2 + c_{2,3} x_2 z_3 + c_{2,12} x_2 z_1 z_2 + c_{2,13} x_2 z_1 z_3 + c_{2,23} x_2 z_2 z_3 + \\ a_{12} x_1 x_2 + c_{12,1} x_1 x_2 z_1 + c_{12,2} x_1 x_2 z_2 + c_{12,3} x_1 x_2 z_3 + c_{12,12} x_1 x_2 z_1 z_2 + c_{12,13} x_1 x_2 z_1 z_3 + c_{12,23} x_1 x_2 z_2 z_3 \end{array} \quad (1)$$

where ln[NOx] represents the natural logarithm of the volume fraction of NOx in the flue gas measured in ppm,

a_j, b_k, and $c_{j,k}$ are adjustable parameters for the j^{th} factor or factor-factor interaction, k^{th} component or component-component interaction, and $j \times k$ factor-component interactions, respectively,

x_j is the j^{th} independent factor transformed to some dimensionless range such as ±1: $-1 < x_k < 1$, (e.g., furnace operating temperature and excess oxygen) and

z_k is the k^{th} mixture fraction, $0 < z_k < 1$ and $\Sigma z_k = 1$, (e.g., hydrogen, natural gas, and propane).

For combustion correlations, Equation (1) is overkill; most of the coefficients will end up pooled as residual error and they are expensive to obtain. It would be best to orthogonally fractionate the design and use fewer points if possible. In non-mixture experiments, response surface and factorial experimental designs have been known for many years (Box and Draper 1989) and have the following desirable properties: they are orthogonal, generating mutually independent and unbiased coefficients; they are rotatable, providing uniform variance and error structure; and they are sequential, capable of being run in orthogonal blocks or fractions. However, preserving these properties in mixture or MPV designs has been challenging.

MPV designs have been used for some time (Hare 1979) and over the years investigators have made various suggestions. One early suggestion was to fractionate the process variables separately from the mixture components (Cornell and Gorman 1984). More recently, orthogonal blocking schemes were given by Nigam (1970), Draper et al (1993), Murthy and Murty (1993), Prescott et al (1993), Lewis et al (1994), Prescott and Draper (1998) and Prescott (2000). It has also been known for some time that orthogonal designs could be run within a portion of the $(q - 1)$-component mixture space. In this way Thompson and Myers (1968) mapped a fully rotatable and orthogonal response surface design. Other transformations map $q - 1$ mixture components to a symmetrical simplex centered at the origin (Claringbold 1955). Even so, the resulting matrix is not orthogonal for the general second-order or greater polynomial, and the resulting coefficients are mutually biased. Orthogonal and rotatable designs in $q - 1$ independent factors can be mapped to larger portions of the simplex using a variety of other transformations including the reciprocal, ratio, and logarithmic transformations (see for example Becker 1969, 1970 and Darroch and Speed 1985). However, such transforms are not used primarily to orthogonalize mixture space, nor can they do so over the entire simplex region. In 2000, Cornell surveyed the field and gave several topics meriting further attention, one of which was the need for robust designs to fractionate mixture and MPV designs. Cornell (2002) also compiled a good survey of mixture designs in a book length treatment; nearly all are characterized by non-orthogonal matrix arrays.

From mixture designs to factorials

Here, we present a piecewise linear transform that can be applied to the entire mixture space and transform mixture and MPV designs to orthogonal spaces. To begin, consider a mixture comprising q components, $q - 1$ being independently determinable according to the following constraints.

$$0 \le z_k \le 1 \text{ and } \sum_{k=1}^{q} z_k = 1 \tag{2}$$

where q is the total number of mixture components

k is an index from 1 to q

z_k is the fraction of the k^{th} mixture component

By way of contrast, factorial designs have no such constraints, although they are usually coded to ±1 or other values of convenience via a linear transform of the original factors.

$$-1 \le x_j \le 1 \qquad (3)$$

where x_j is the j^{th} factor of a factorial or related design.

In the limit, we must account for any number (m) of mixture fractions and transform them to $q-1$ associated factors. In practice, we can represent the entire domain of q mixture fractions with some subset of the infinite matrices **Z**, **X**, and **T**, as defined below.

$$\mathbf{Z}=\begin{pmatrix}1 & 0 & 0 & 0 & \cdots\\ 1 & 1 & 0 & 0 & \cdots\\ 1 & 1/2 & 1/2 & 0 & \cdots\\ 1 & 1/3 & 1/3 & 1/3 & \cdots\\ \vdots & \vdots & \vdots & \vdots & \ddots\end{pmatrix} \mathbf{X}=\begin{pmatrix}+ & - & - & - & \cdots\\ + & + & - & - & \cdots\\ + & + & + & - & \cdots\\ + & + & + & + & \cdots\\ \vdots & \vdots & \vdots & \vdots & \ddots\end{pmatrix} \mathbf{T}=\begin{pmatrix}1 & -1 & -1 & -1 & \cdots\\ 0 & 2 & 0 & 0 & \cdots\\ 0 & 2 & 4 & 0 & \cdots\\ 0 & 2 & 2 & 6 & \cdots\\ \vdots & \vdots & \vdots & \vdots & \ddots\end{pmatrix} \qquad (4)$$

The above matrices have the following regularities.

- **Z** comprises
  - a first column comprising 1s,
  - 0s for all entries above the principal diagonal, and
  - an infinite principal diagonal of reciprocal counting numbers, the first entry being repeated (1, 1, 1/2, 1/3, 1/4, 1/5, …) and
  - the diagonal entries are repeated to the left.
- **X** comprises
  - an infinite principal diagonal and lower triangular region of 1s (in accordance with convention, we represent ±1 factor values by their sign) and
  - an upper triangular region of −1s.
  - **X** may also be derived from **Z** by mapping all zero elements to −1 and all non-zero elements to +1.
- **T** comprises
  - an infinite principal diagonal starting with 1 and followed by the even numbers in natural order: 1, 2, 4, 6, 8, 10, …,
  - a first row of −1s to the right of the first element,
  - a first column of 0s below the first element,
  - 2s for all remaining elements below the principal diagonal.

The General Transformations

For actual (finite) mixture fractions, we shall transform some subset of the mixture map **Z** to a subset of **X** using a subset of the transformation matrix, **T,** (or vice-versa) according to the following relations (which hold for the infinite matrices or finite subsets thereof).

$$\mathbf{X} = \mathbf{Z}\cdot\mathbf{T} \qquad (5)$$
$$\mathbf{Z} = \mathbf{X}\cdot\mathbf{T}^{-1} \qquad (6)$$
$$\mathbf{T} = \mathbf{Z}^{-1}\mathbf{X} \qquad (7)$$

In general, the mixture space will be subdivided into $(q - 1)!$ regions with all regions being identical except for reflections across axes (corresponding to permutation of the given matrices).

Examples with Quaternary Mixtures

For example, consider the quaternary mixture space comprising all possible mixture fractions of z_1, z_2, z_3, and z_4. We shall select the first $q - 1 = 3$ of them, denoted $\mathbf{Z}_{0123}$. Without any loss in generality, the fourth is tacit and may be recovered via Equation (2). $\mathbf{Z}_{0123}$ is equivalent to the first $q \times q$ entries of $\mathbf{Z}$ in Equation (4).

$$\mathbf{Z}_{0123} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 1 & 1/2 & 1/2 & 0 \\ 1 & 1/3 & 1/3 & 1/3 \end{pmatrix} \tag{8}$$

From top row to bottom, $\mathbf{Z}_{0123}$ holds the respective coordinates for the vertices labeled *h*, *e*, *b*, and *a*, in Figure 2, and a leftmost column of 1s; this always results in an invertible square matrix.

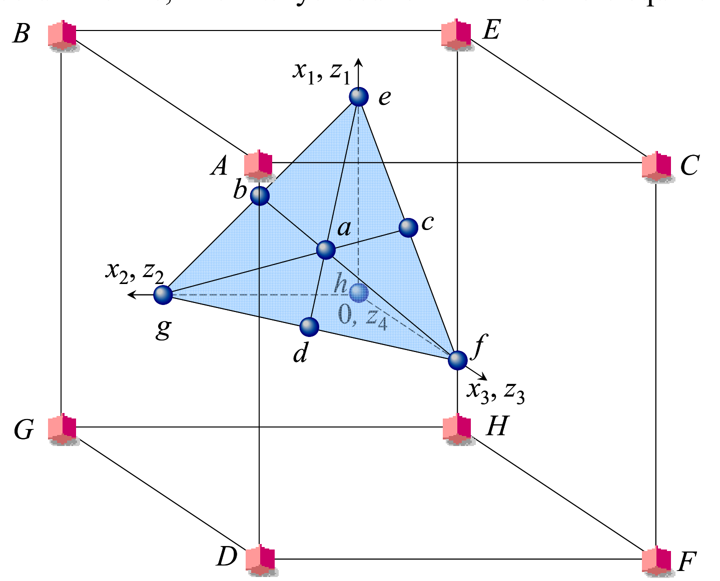

Figure 2, correspondence of mixture and factorial space for a four-component mixture. The four-component mixture may be mapped to the orthogonal three-factor (2^3) factorial design space using linear transformations. Here lower case letters (spherical point markers) map to uppercase ones (cubic point markers) and vice-versa.

The right-tetrahedral region coded by Z_{0123} (shaded Region *efgh* in Figure 2) comprises the four-component mixture space with z_4 tacit and coincident with the origin. We now desire to map points *a – h* in mixture space $(-1 \le z_1, z_2, z_3 \le 1)$ to points *A–H*, respectively, in factor space $(-1 \le x_1, x_2, x_3 \le 1)$ using only linear transformations. This can be done piecewise. However, any transformation will require an asymmetric stretch because the two regions are not geometrically similar; as a result, the resolution will be poorer along the *ah* axis (corresponding to the z_4 axis in the original mixture space). Accordingly, we should select z_1, z_2, and z_3 to be the components needing the highest resolution.

We now turn our attention to a portion of the shaded region in Figure 2: the surface *abe* and Point *h* subtend a tetrahedral volume *abeh*. Region *abeh* represents all mixtures for which $z_1 \ge z_2 \ge z_3$. We shall symbolize this with the mixture subscripts denoted 123 (or preferably 0123 to remind us that it is a four-component mixture corresponding to three independent factors). A total of six such regions exist corresponding to $(q - 1)!$; that is, all possible permutations of $q - 1$ components. Table 1 gives the correspondences.

Table 1, Relation Between Permutation, Region, and Constraint

Permutation Order	Region	Constraint
0123	*abeh*	$z_1 \geq z_2 \geq z_3$
0132	*aceh*	$z_1 \geq z_3 \geq z_2$
0213	*abgh*	$z_2 \geq z_1 \geq z_3$
0231	*adgh*	$z_2 \geq z_3 \geq z_1$
0312	*acfh*	$z_3 \geq z_1 \geq z_2$
0321	*adfh*	$z_3 \geq z_2 \geq z_1$

Note that all regions share the common axis *ah* and are reflections of one another; therefore each region contains *a* and *h* (coded as the first and last points in each matrix – see Table 2); Table 2 gives the explicit coordinates for the mixture and factorial spaces referenced in Table 1.

Table 2, Explicit Correspondence Between Mixture and Factorial Points

$\mathbf{Z}_{0123} \leftrightarrow \mathbf{X}_{0123}$

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 1 & 1/2 & 1/2 & 0 \\ 1 & 1/3 & 1/3 & 1/3 \end{pmatrix} \leftrightarrow \begin{pmatrix} + & - & - & - \\ + & + & - & - \\ + & + & + & - \\ + & + & + & + \end{pmatrix}$$

$\mathbf{Z}_{0132} \leftrightarrow \mathbf{X}_{0132}$

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 1 & 1/2 & 0 & 1/2 \\ 1 & 1/3 & 1/3 & 1/3 \end{pmatrix} \leftrightarrow \begin{pmatrix} + & - & - & - \\ + & + & - & - \\ + & + & - & + \\ + & + & + & + \end{pmatrix}$$

$\mathbf{Z}_{0213} \leftrightarrow \mathbf{X}_{0123}$

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 1 & 0 & 1 & 0 \\ 1 & 1/2 & 1/2 & 0 \\ 1 & 1/3 & 1/3 & 1/3 \end{pmatrix} \leftrightarrow \begin{pmatrix} + & - & - & - \\ + & - & + & - \\ + & + & + & - \\ + & + & + & + \end{pmatrix}$$

$\mathbf{Z}_{0231} \leftrightarrow \mathbf{X}_{0231}$

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 1 & 0 & 1 & 0 \\ 1 & 0 & 1/2 & 1/2 \\ 1 & 1/3 & 1/3 & 1/3 \end{pmatrix} \leftrightarrow \begin{pmatrix} + & - & - & - \\ + & + & - & - \\ + & - & + & + \\ + & + & + & + \end{pmatrix}$$

$\mathbf{Z}_{0312} \leftrightarrow \mathbf{X}_{0312}$

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 1 \\ 1 & 1/2 & 0 & 1/2 \\ 1 & 1/3 & 1/3 & 1/3 \end{pmatrix} \leftrightarrow \begin{pmatrix} + & - & - & - \\ + & - & - & + \\ + & + & - & + \\ + & + & + & + \end{pmatrix}$$

$\mathbf{Z}_{0321} \leftrightarrow \mathbf{X}_{0321}$

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 1 \\ 1 & 0 & 1/2 & 1/2 \\ 1 & 1/3 & 1/3 & 1/3 \end{pmatrix} \leftrightarrow \begin{pmatrix} + & - & - & - \\ + & - & - & + \\ + & - & + & + \\ + & + & + & + \end{pmatrix}$$

Table 3 mathematically expresses the six transforms and the regions they superintend.

Table 3, Linear Piecewise Transformations for the Four-component Mixture Space

Linear Piecewise Transformations	Region	Constraint
$\mathbf{X}_{0123} = \mathbf{Z}_{0123}\mathbf{T}_{0123}$; $\mathbf{Z}_{0123} = \mathbf{X}_{0123}\mathbf{T}_{0123}^{-1}$	*abeh*	$z_1 \geq z_2 \geq z_3$
$\mathbf{X}_{0132} = \mathbf{Z}_{0132}\mathbf{T}_{0132}$; $\mathbf{Z}_{0132} = \mathbf{X}_{0132}\mathbf{T}_{0132}^{-1}$	*aceh*	$z_1 \geq z_3 \geq z_2$
$\mathbf{X}_{0213} = \mathbf{Z}_{0213}\mathbf{T}_{0213}$; $\mathbf{Z}_{0213} = \mathbf{X}_{0213}\mathbf{T}_{0213}^{-1}$	*abgh*	$z_2 \geq z_1 \geq z_3$
$\mathbf{X}_{0231} = \mathbf{Z}_{0231}\mathbf{T}_{0231}$; $\mathbf{Z}_{0231} = \mathbf{X}_{0231}\mathbf{T}_{0231}^{-1}$	*adgh*	$z_2 \geq z_3 \geq z_1$
$\mathbf{X}_{0312} = \mathbf{Z}_{0312}\mathbf{T}_{0312}$; $\mathbf{Z}_{0312} = \mathbf{X}_{0312}\mathbf{T}_{0312}^{-1}$	*acfh*	$z_3 \geq z_1 \geq z_2$
$\mathbf{X}_{0321} = \mathbf{Z}_{0321}\mathbf{T}_{0321}$; $\mathbf{Z}_{0321} = \mathbf{X}_{0321}\mathbf{T}_{0321}^{-1}$	*adfh*	$z_3 \geq z_2 \geq z_1$

The Use of Permutation Matrices

The particular reflections may be derived from **Z** using a permutation matrix **P**; e.g.,

$$\mathbf{Z}_{0123}\mathbf{P}_{0312} = \mathbf{Z}_{0312} \tag{9}$$

where $\mathbf{P}_{0312} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \end{pmatrix}$ and $\mathbf{Z}_{0312} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 1 \\ 1 & 1/2 & 0 & 1/2 \\ 1 & 1/3 & 1/3 & 1/3 \end{pmatrix}$.

The desired permutation matrix is derived straightforwardly from the permutation order (referenced in the subscript order) according to the following rules.

- The first row and column comprise a 1 followed by 0s.
- The first row and column are indexed from zero and incremented by unity. That makes the second diagonal element the start of Row 1 and Column 1.
- The remaining elements are filled from Row 1 to Row $q - 1$ by placing a 1 in the column corresponding to the subscripted element and 0s elsewhere.
- The permuted $\mathbf{Z}_{0abc\ldots}$ matrix may also be built directly from the subscript order, row by row, in a top-down cumulative fashion with
  - 1 placed in column *a*,
  - 1/2 placed in each of columns *a* and *b*,
  - 1/3 placed in each of columns *a*, *b*, *c*, etc.

In general, the permutation matrices have the following properties.

- Inverse equal to the transpose
  - $\mathbf{P}^{-1} = \mathbf{P}^{\mathrm{T}}$
- Permutation of **Z** and **X** by post-multiplication
  - $\mathbf{Z}_{0123\ldots}\mathbf{P}_{0abc\ldots} = \mathbf{Z}_{0abc\ldots}$
  - $\mathbf{X}_{0123\ldots}\mathbf{P}_{0abc\ldots} = \mathbf{X}_{0abc\ldots}$
- Permutation of **T** by pre- and post-multiplication
  - $\mathbf{P}^{\mathrm{T}}_{0abc\ldots}\mathbf{T}_{0123\ldots}\mathbf{P}_{0abc\ldots} = \mathbf{T}_{0abc\ldots}$
- Correct transformation preserved by post-multiplication with the appropriate **T** matrix
  - $\mathbf{Z}_{0abc\ldots}\mathbf{T}_{0abc\ldots} = \mathbf{X}_{0abc\ldots}$

Mapping of a 2^3 Factorial Design to 4-Component Mixture Space

For illustrative purposes, we continue with our four-component example; any point satisfying the constraint $z_1 \geq z_2 \geq z_3$ will be transformed from mixture space to factorial space via $\mathbf{X}_{0123} = \mathbf{Z}_{0123}\mathbf{T}_{0123}$. Using this method, experimental designs in factor space may be mapped to mixture fractions and vice-versa. For example, Figure 3 shows how the 2^3 factorial design maps to its corresponding mixture design (incomplete simplex-centroid). The figure also shows the matrix for the mixture fraction using traditional coordinates [i.e., letting $z_4 = 1-(z_1+z_2+z_3)$ and omitting the 1s column].

$$
\begin{array}{c}
\underline{\text{Pt}} \\ 1 \\ 2 \\ 3 \\ 4 \\ 5 \\ 6 \\ 7 \\ 8
\end{array}
\quad
\begin{pmatrix}
1 & x_1 & x_2 & x_3 \\
\hline
+ & - & - & - \\
+ & - & - & + \\
+ & - & + & - \\
+ & - & + & + \\
+ & + & - & - \\
+ & + & - & + \\
+ & + & + & - \\
+ & + & + & +
\end{pmatrix}
=
\begin{pmatrix}
1 & z_1 & z_2 & z_3 \\
\hline
1 & 0 & 0 & 0 \\
1 & 0 & 0 & 1 \\
1 & 0 & 1 & 0 \\
1 & 0 & 1/2 & 1/2 \\
1 & 1 & 0 & 0 \\
1 & 1/2 & 0 & 1/2 \\
1 & 1/2 & 1/2 & 0 \\
1 & 1/3 & 1/3 & 1/3
\end{pmatrix}
=
\begin{pmatrix}
z_1 & z_2 & z_3 & z_4 \\
\hline
0 & 0 & 0 & 1 \\
0 & 0 & 1 & 0 \\
0 & 1 & 0 & 0 \\
0 & 1/2 & 1/2 & 0 \\
1 & 0 & 0 & 0 \\
1/2 & 0 & 1/2 & 0 \\
1/2 & 1/2 & 0 & 0 \\
1/3 & 1/3 & 1/3 & 0
\end{pmatrix}
$$

Figure 3, correspondence between full factorial design and mixture design. For convenience, the mixture design is also expressed in traditional mixture coordinates at right.

As an example of the method, consider Point 3 in Figure 3 where $x_2 > x_1 = x_3$. This point is borders two regions of factor space: $x_2 \geq x_1 \geq x_3$ and $x_2 \geq x_3 \geq x_1$. Therefore, we may use either $\mathbf{Z}_{0213} = \mathbf{X}_{0213}\mathbf{T}_{0213}^{-1}$ or $\mathbf{Z}_{0231} = \mathbf{X}_{0231}\mathbf{T}_{0231}^{-1}$ to map $x_3^{\mathsf{T}} = (+ \quad - \quad + \quad -) \rightarrow z_3^{\mathsf{T}} = (1 \quad 0 \quad 1 \quad 0)$. Arbitrarily selecting the first, we have the following.

$$
\mathbf{T}_{0231} = \mathbf{P}_{0231}^{\mathsf{T}}\mathbf{T}_{0123}\mathbf{P}_{0231} =
\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \end{pmatrix}
\begin{pmatrix} 1 & -1 & -1 & -1 \\ 0 & 2 & 0 & 0 \\ 0 & 2 & 4 & 0 \\ 0 & 2 & 2 & 6 \end{pmatrix}
\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 \end{pmatrix}
=
\begin{pmatrix} 1 & -1 & -1 & -1 \\ 0 & 6 & 2 & 2 \\ 0 & 0 & 2 & 0 \\ 0 & 0 & 2 & 4 \end{pmatrix}
$$

$$
z_3^{\mathsf{T}} = x_3^{\mathsf{T}}\mathbf{T}_{0213}^{-1} = (+ \quad - \quad + \quad -)
\begin{pmatrix} 1 & 1/6 & 1/6 & 1/6 \\ 0 & 1/6 & -1/12 & -1/12 \\ 0 & 0 & 1/2 & 0 \\ 0 & 0 & -1/4 & 1/4 \end{pmatrix}
= (1 \quad 0 \quad 1 \quad 0)
$$

Using a Single Linear Transform with Reordering

Rather than employ permutation matrices and transform all possible piecewise linear regions, a simpler and computationally less expensive method for mapping *m* components is as follows.

- Truncate the **X** or **Z** matrix given in Equation (4) to an $q \times q$ matrix.
- Reorder the source components or factors from greatest to lowest value.
- Perform the transformation **X** = **ZT** or **Z** = **XT**$^{-1}$.
- Reorder the transformed coordinates to correspond with the original source order.

This method requires only one set of truncated transformation matrices. Mixture coordinates are then orthogonalized using a single generalized linear transform preceded and followed by reordering the inputs and results, as necessary. The procedure is applicable to any number of mixture components or factor dimensions. Using it, typical experimental designs (e.g., full or fractional factorials, inscribed central-composites, Box-Behnken designs, etc.) can be expressed in mixture components and vice versa: Table 4 shows the results of mapping a three-factor central composite design to traditional mixture fractions. Table 5 does the same for a three-factor Box-Behnken design. As an example of this latter method, consider Point + − + in Table 4, that is $x_3^{\mathrm{T}} = \begin{pmatrix}1 & 0.595 & -0.595 & 0.595\end{pmatrix}$. In order to use the

Table 4, Mixture Coordinate Equivalents for a Three-Factor Inscribed Central Composite Design

Pt	Factorial Coordinates			Mixture Design Coordinates			
	x_1	x_2	x_3	z_1	z_2	z_3	z_4
− − −	−0.595	−0.595	−0.595	0.068	0.068	0.068	0.797
− − +	−0.595	−0.595	0.595	0.068	0.068	0.365	0.500
− + −	−0.595	0.595	−0.595	0.068	0.365	0.068	0.500
− + +	−0.595	0.595	0.595	0.068	0.365	0.365	0.203
+ − −	0.595	−0.595	−0.595	0.662	0.068	0.068	0.203
+ − +	0.595	−0.595	0.595	0.365	0.068	0.365	0.203
+ + −	0.595	0.595	−0.595	0.365	0.365	0.068	0.203
+ + +	0.595	0.595	0.595	0.266	0.266	0.266	0.203
0 0 0	0	0	0	1/6	1/6	1/6	1/2
0 0 0	0	0	0	1/6	1/6	1/6	1/2
= 0 0	–	0	0	0	1/4	1/4	1/2
‡ 0 0	+	0	0	2/3	1/6	1/6	0
0 = 0	0	–	0	1/4	0	1/4	1/2
0 ‡ 0	0	+	0	1/6	2/3	1/6	0
0 0 =	0	0	–	1/4	1/4	0	1/2
0 0 ‡	0	0	+	1/6	1/6	2/3	0

Table 5, Mixture Coordinate Equivalents for a Three-Factor Box Behnken Design

	Factorial Coordinates			Mixture Design Coordinates			
Pt	x_1	x_2	x_3	z_1	z_2	z_3	z_4
− − 0	−	−	0	0	0	1/2	1/2
− 0 −	−	0	−	0	1/2	0	1/2
0 − −	0	−	−	1/2	0	0	1/2
0 + +	0	+	+	1/6	5/12	5/12	0
+ 0 +	+	0	+	5/12	1/6	5/12	0
+ + 0	+	+	0	5/12	5/12	1/6	0
0 0 0	0	0	0	1/6	1/6	1/6	1/2
0 0 0	0	0	0	1/6	1/6	1/6	1/2
0 0 0	0	0	0	1/6	1/6	1/6	1/2
− 0 +	−	0	+	0	1/4	3/4	0
− + 0	−	+	0	0	3/4	1/4	0
0 − +	0	−	+	1/4	0	0	0
0 + −	0	+	−	1/4	3/4	0	0
+ − 0	+	−	0	3/4	0	1/4	0
+ 0 −	+	0	−	3/4	1/4	0	0

$\mathbf{T}_{0123}$ transform – i.e., $z_3^{\mathsf{T}} = x_3^{\mathsf{T}}\mathbf{T}_{0123}^{-1}$ – we must reorder x_3^{T} as $^{*}x_3^{\mathsf{T}} = (1 \quad 0.595 \quad 0.595 \quad -0.595)$ where the asterisk (*) denotes the reordered vector. Then we apply $*z_3^{\mathsf{T}} = *x_3^{\mathsf{T}}\mathbf{T}_{0231}^{-1}$ giving $*z_3^{\mathsf{T}} = *x_3^{\mathsf{T}}\mathbf{T}_{0231}^{-1}$ and the following result.

$$*z_3^{\mathsf{T}} = *x_3^{\mathsf{T}}\mathbf{T}_{0213}^{-1} = (1 \quad 0.595 \quad 0595 \quad -0.595)\begin{pmatrix} 1 & 1/6 & 1/6 & 1/6 \\ 0 & 1/2 & 0 & 0 \\ 0 & -1/4 & 1/4 & 0 \\ 0 & -1/12 & -1/12 & 1/6 \end{pmatrix} = (1 \quad 0.365 \quad 0.365 \quad 0.068)$$

The row vector is then reordered to coincide with the original factor order of the original source vector: $x_3^{\mathsf{T}} = (1 \quad 0.595 \quad -0.595 \quad 0.595) \rightarrow z_3^{\mathsf{T}} = (1 \quad 0.365 \quad 0.068 \quad 0.365)$.

Mixture-Process Variable (MPV) Experiments

It is possible to fractionate any MPV experimental design. For example, Figure 4 shows the component and factor disposition that transform to a $2^2 \times 3^2$ factorial design (like markers denoting orthogonal fractions). The design may also be fractionated via standard techniques.

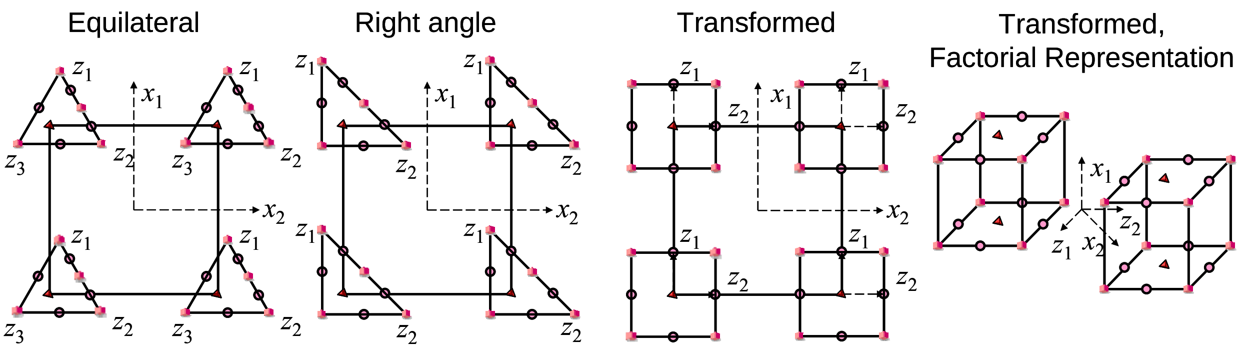

Figure 4, illustration of a mixture-process-variable design that maps to a 2^2 x 3^2 factorial design. Point markers are segregated in three orthogonal groups. The design contains a non-uniform point distribution in the equilateral or right-angular coordinates. The mixture design is centered at $z_1 = z_2 = 1/4$, $z_3 = 1/2$ rather than $z_1 = z_2 = z_3 = 1/3$. However, the design is symmetrical and orthogonal in the transformed coordinates.

Any of a variety of orthogonal and/or rotatable designs could be used as well. Notwithstanding, it should be noted that the MPV design is not uniform: it provides higher resolution for z_1 x z_2 blends than for z_1 x z_3 or z_2 x z_3 blends. Because the ternary component space cannot be mapped to a square factor space in a radially symmetric way, the simplex does not (and cannot) map to factor space without some kind of radial asymmetry. Of note, the design center maps from (1/4, 1/4, 1/2) rather than the original ternary design center (1/3, 1/3, 1/3). This lack of "center-preservedness" is a general feature for this class of transforms. This radial asymmetry is perhaps most easily apprehended by reference to Figure 5 where otherwise identical figures are transformed to and from right-angle, equilateral, and rectangular systems.

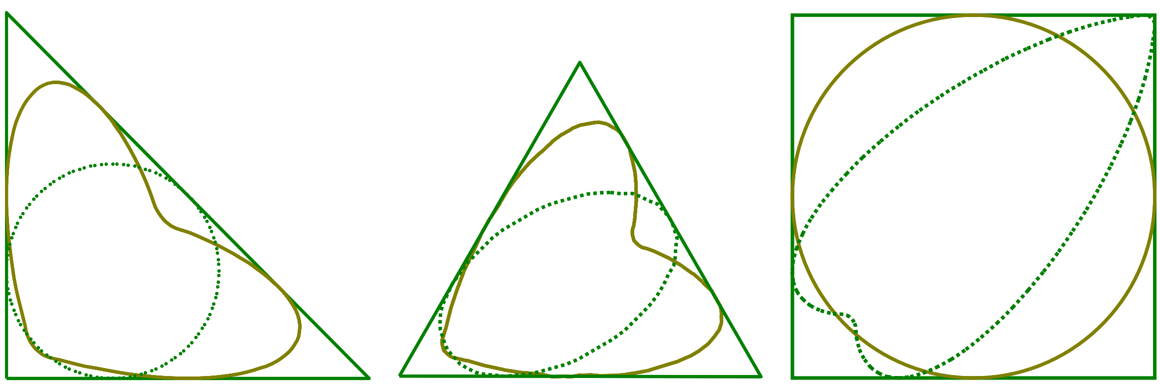

Figure 5, mapping from right angle to equilateral to rectangular coordinates and vice versa. Note that the associated stretches are radially asymmetric. For example, the dotted circle in right-angled coordinates (left) becomes an ellipse in equilateral coordinates (center) and an inverted cardioid in rectangular coordinates (right). Likewise, the solid circle in rectangular coordinates transforms to respective cardioids in equilateral and right-angular coordinates.

Being that this is the case, one should match the asymmetry of the design to associated asymmetries in the response, to the extent possible. This was done in the case of the particular ternary fuel blend used as an example. That is, H_2 and C_3H_8 (z_1 and z_2 respectively) are the primary causative agents for NOx emissions: NOx from RFG is formed via two mechanisms – the so-called *thermal* and *prompt* NOx mechanisms. Thermal NOx formation is responsible for ~80% of the total NOx; the mechanism becomes more facile with increasing flame temperature and such is disproportionately elevated by hydrogen. Prompt NOx accounts for ~20% of the total NOx formed and is facilitated by hydrocarbon fragments in the fuel (primarily via dissociation of C_3H_8). Natural gas, the third component, is mostly

CH_4 – a refractory molecule that contributes much less significantly to thermal or prompt NOx than H_2 and C_3H_8. Therefore, H_2 and C_3H_8 were assigned to the vertical and horizontal directions in the right-angular coordinates.

Center-Preserving Transformations

It is possible to map right-angled coordinates to other systems that are center-preserving; this is obtained at the expense of requiring more transforms [$q!$ rather than $(q - 1)!$ transforms which comprise $q - 1$ families of reflected shapes]. Figure 6 illustrates several center-preserving transformations (e.g., 1/3, 1/3, 1/3 → 0, 0) and associated regions, each of which may be transformed to or from one another.

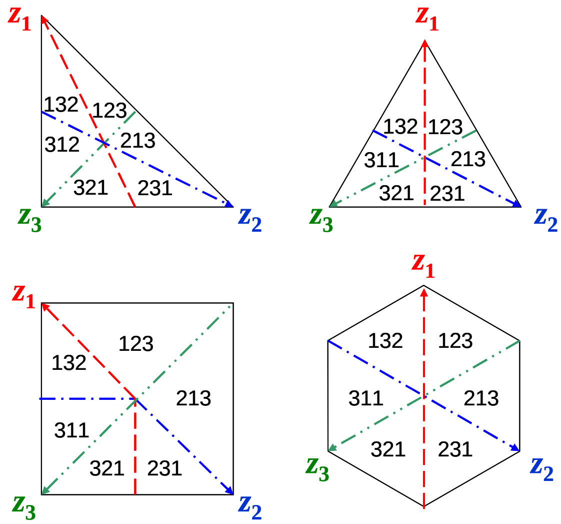

Figure 6, center-preserving transforms. The numbers refer to the $q!$ piecewise continuous regions over which a transform is applicable. For example, the 132 region indicates the mixture region where $z_1 \geq z_3 \geq z_2$.

The disadvantage of center-preserving transformations is that there are more of them. For example, the piecewise linear transformation from right-triangular to rectangular coordinates generates six different regions – two reflections each of three distinctly shaped regions as opposed to reordering of a single piecewise transform. Table 6 gives the transforms to and from right-angled coordinates to equilateral, rectangular, and hexagonal coordinates for a ternary mixture system. Center-preserving transforms do not facilitate mapping factorial and like designs to and from mixture space as conveniently as a single transform (indeed, any region can be transformed to any other – one could transform the equilateral region to a circular one, though the transforms would not be linear); so, we do not treat the topic any further.

Table 6, Transforms to and from Right Angle Coordinates to Other Coordinate Systems

Region	From Right Angle Coordinates to								
	Equilateral			Rectangular			Hexagonal		
123	1	0	1	1	0	1	1	1	–1
$z_1 \geq z_2 \geq z_3$	1	√3/2	−1/2	1	√3	0	1	1	3
	1	−√3/2	−1/2	1	–√3	–1	1	–2	–2
132				1	0	1	1	1	–1
$z_1 \geq z_3 \geq z_2$				1	√3	–1	1	0	2
				1	–√3	0	1	–1	–1
213				1	√3/2	3/2	1	3	1
$z_2 \geq z_1 \geq z_3$				1	√3/2	–1/2	1	–1	1
				1	–√3	–1	1	–2	–2
231	same as above for all regions			1	0	2	1	2	0
$z_2 \geq z_3 \geq z_1$				1	√3/2	–1/2	1	–1	1
				1	–√3/2	–3/2	1	–1	–1
312				1	–√3/2	3/2	1	1	–1
$z_3 \geq z_1 \geq z_2$				1	√3	–1	1	0	2
				1	–√3/2	–1/2	1	–1	–1
321				1	0	2	1	2	0
$z_3 \geq z_2 \geq z_1$				1	√3/2	–3/2	1	–1	1
				1	–√3/2	–1/2	1	–1	–1

Table 6, continued

Region	To Right Angle Coordinates from								
	Equilateral			Rectangular			Hexagonal		
123	1/3	1/3	1/3	1/3	1/3	1/3	1/3	1/3	1/3
$z_1 \geq z_2 \geq z_3$	0	√3/3	−√3/3	−√3/9	2√3/9	−√3/9	5/12	−1/12	−1/3
	2/3	−1/3	−1/3	2/3	−1/3	−1/3	−1/4	1/4	0
132	same as above for all regions			1/3	1/3	1/3	1/3	1/3	1/3
$z_1 \geq z_3 \geq z_2$				√3/9	√3/9	−2√3/9	1/2	0	−1/2
				2/3	−1/3	−1/3	−1/6	1/3	−1/6
213				1/3	1/3	1/3	1/3	1/3	1/3
$z_2 \geq z_1 \geq z_3$				√3/18	5√3/18	2√3/9	1/4	−1/4	0
				1/2	−1/2	0	−1/12	5/12	−1/3
231				1/3	1/3	1/3	1/3	1/3	1/3
$z_2 \geq z_3 \geq z_1$				−√3/9	7√3/18	−5√3/18	1/3	−1/6	−1/6
				2/3	−1/3	−1/3	0	1/2	−1/2
312				1/3	1/3	1/3	1/3	1/3	1/3
$z_3 \geq z_1 \geq z_2$				√3/18	2√3/9	−5√3/18	1/2	0	−1/2
				1/2	0	−1/2	−1/6	1/3	−1/6
321				1/3	1/3	1/3	1/3	1/3	1/3
$z_3 \geq z_2 \geq z_1$				√3/9	5√3/18	−7√3/18	1/3	−1/6	−1/6
				2/3	−1/3	−1/3	0	1/2	−1/2

Conclusions

A method has been given for mapping orthogonal and rotatable factor designs to mixture designs or vice-versa. In its simplest form, the method uses a single linear transformation (with a reordering of the source and target coordinates as necessary so that the transform remains appropriate). The method is applicable to any number of factors and provides a series of linear transforms between mixture and factorial designs, thus mapping orthogonal factorial designs to mixture spaces and vice-versa. The transform allows mixture, mixture-amount, and mixture-process-variable designs to be made orthogonal and rotatable in the transformed space. The mapping is necessarily asymmetrical but results in orthogonal and rotatable designs in the target space.

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