An Introduction to Mixture Designs

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Executive Summary

Mixture experiments present a unique test region that requires special consideration when designing experiments. These designs focus on a blending of components that cannot be altered independently. This results in a constrained design region that differs from more traditional design options. Therefore, a number of design options exist to best sample from this space. This paper will discuss simplex-lattice designs, simplex-centroid designs, optimal mixture designs, and space-filling mixture designs. The different polynomials that can be fit using these designs will be discussed. Additionally, a JMP demonstration on creating these designs and analyzing the data will be shown.

Keywords: Mixture experiments, Constrained, Simplex, Optimal, Space Filling, JMP

Introduction

A common, but often overlooked, experiment is one that involves a mixture of ingredients to form a solution. Mixture experiments are special types of response surface experiments, and in these experiments, the goal is to determine if there is a blend of ingredients that produces a more optimal response (whether this is to maximize or minimize some property). In *Experiments with Mixtures, Designs, Models, and the Analysis of Mixture Data*, Cornell states that the goals of these experiments is to "try to model the dependence of the response variable … on the relative proportions of the components with some form of mathematical equation so that:

1) The influence on the response of each component singly and in combination with the other components can be measured. If this is done successfully, those components having the least effect or felt to be less active might be "screened" out, leaving us with only those components having the greatest effect on the response.
2) Predictions of the response to any mixture or combination of the components proportions can be made
3) Mixtures or blends of the components that yield desirable values of the response can be identified"

One such example of this would be in a mixture of gunpowder. Gunpowder is made up of three ingredients; sulfur, charcoal, and potassium nitrate. An experimenter might be interested in what combination of these ingredients produces the most force upon ignition. The amount of gunpowder in a given bullet is predetermined, but the proportion of the ingredients to be used can be adjusted. This created a constrained region as changing the amount of one factor level automatically affects the values of the other factors in the design. The constraint that the proportions all of the ingredients must add up to 100% creates a unique design region that differs from classical design settings. The additional mixture constraint is as follows:

$$\sum_{i=1}^{q} x_i = x_1 + \cdots + x_q = 1,$$

where q represents the number of ingredients in the system under study and the proportion of the i^{th} ingredient in the mixture is denoted by x_i. In order to understand the relationship between ingredients, there are several techniques that can be used. Some techniques, such as trial and error or procedures using a large number of combinations, lack rigor and can be quite costly. More efficient designs can be utilized in order to understand the overall system performance as ingredient amounts change. This paper will explore the effects of a constrained design region and several techniques that provide useful results.

Background

In order to understand the basic concept of mixture designs and their analysis, we use the original mixture problem example from Cornell (2002) regarding the effect of blending fuels on mileage. Consider only two gasoline stocks, which we label fuels A and B. Assume that the response of interest is the mileage obtained by driving a test car with the fuel, where the mileage is recorded in units of the average number of miles traveled per gallon. Suppose we know that fuel A normally yields 13 miles per gallon (MPG) and fuel B normally yields 7 miles per gallon. These are examples of pure mixtures, blends containing 100% of a single ingredient. We might assume that with a gallon of each, we would average 13+7=20 miles on 2 gallons, which is 10 miles per gallon. This is an example of a binary mixture, a blend made up of fractions of only two ingredients. However, it is possible that a blended mixture might produce a value higher or lower than expected based off the assumption of the two fuel types contributing equally. The question we wish to test is, "If we combine or blend the two fuels and drive the same test car, is there a blend of A and B such as 50:50 blend or a 33:67 blend of A:B that yields a higher average number of miles per gallon than the 10 miles per gallon of averaging the two?" The data from an experiment using a mixture of 50% of each fuel type can be seen in Table 1.

Table 1: "Average Mileage for Each of Five Trials" from Cornell, 2002

Trial	Mileage from Two Gallons of 50%:50% Blended Fuel	Average Mileage per Gallon
1	**24.6**	**12.30**
2	**23.3**	**11.65**
3	**24.3**	**12.15**
4	**23.1**	**11.55**
5	**24.7**	**12.35**
	Overall Average	**12.00**

If the blend is strictly additive, then we would expect an average of 10 miles per gallon, but we are interested in determining if this assumption holds true or not. We can then plot the expected results given our prior knowledge (that 100% of A is 13 MPG, 100% of B is 7 MPG, and a 50%:50% blend is 10 MPG). The assumption we are making is a linear relationship. Additionally, we can use the information from our experiment to update the line and see what our new predictions would be. Perhaps the relationship is not linear and we can observe this in our updated line. For this data, we would then plot 100% of A is 13 MPG, 100% of B is 7 MPG, and a 50%:50% blend is 12 MPG (as seen in the data). This graph can be seen in Figure 1.

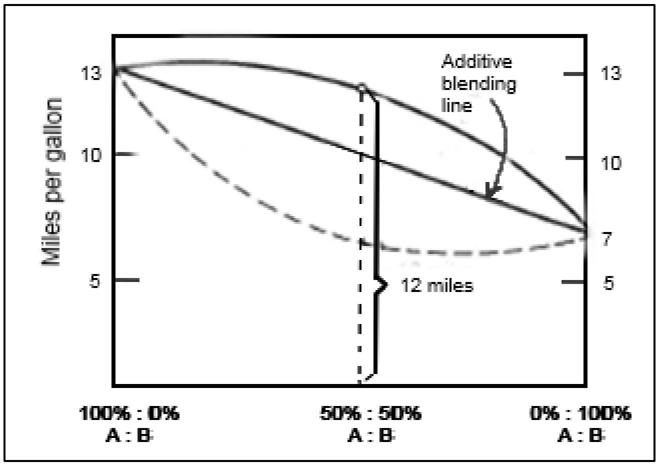


Figure 1: "Plotting the mileage of the 50%:50% blend of fuels A and B. The formula for the additive blending line is mileage = [(13 miles * A) + (7 miles * B)] / 100%" recreated from Cornell, 2002

Since the blend produces a higher MPG than the expected 10 MPG, A and B are complementary to each other when blended. That is, the two fuel types cause a higher than expected resulted and combining the two results in more favorable results. This example shows the overall process of how to conduct a mixture experiment. However, a key step in planning a mixture experiment is determining what design to use to ensure the right data is collected.

Method

Mixture experiments present several unique challenges for designing an appropriate test. The test region is highly constrained, and it is not possible to change the levels of one factor independently of the others. A classical design such as a full factorial design will not yield the results that are needed to analyze these mixtures. Instead, several designs that will be discussed in the paper are simplex-lattice designs, simplex-centroid designs, optimal mixture designs, and space filling mixture designs, which if used appropriately, can overcome the challenges.

Simplex-lattice designs

These designs were introduced by Scheffé (1958-1965) and are credited with being the foundation for designed experiments for mixtures. The simplex-lattice design selects points spread evenly over the factor space. They are defined to support a polynomial model of degree m in q components over the lattice. This is denoted as a {q,m} simplex-lattice. Thus, q=2 is a line, q=3 is an equilateral triangle, and q=4 is a tetrahedron. The coordinate system is called the simplex coordinate system. We will have m+1 equally spaced values from 0 to 1 on each axis such that:

$$x_i = 0, \frac{1}{m}, \frac{2}{m}, \ldots, 1 \; where \; x_i \; are \; the \; sampled \; points \; of \; each \; factor$$

These designs sample all possible combinations of the components where the proportions for each are used. For example, for a {3,2} design:

$$(x_1, x_2, x_3) = (1,0,0), (0,1,0), (0,0,1), \left(\frac{1}{2}, \frac{1}{2}, 0\right), \left(\frac{1}{2}, 0, \frac{1}{2}\right), \left(0, \frac{1}{2}, \frac{1}{2}\right)$$

Note, there are no complete mixtures, blends made up of all the ingredients, in this design. For this reason, axial points can be augmented onto these designs to have a complete mixture represented, a design that will be discussed later. Several other examples of these designs can be seen in Figure 2.

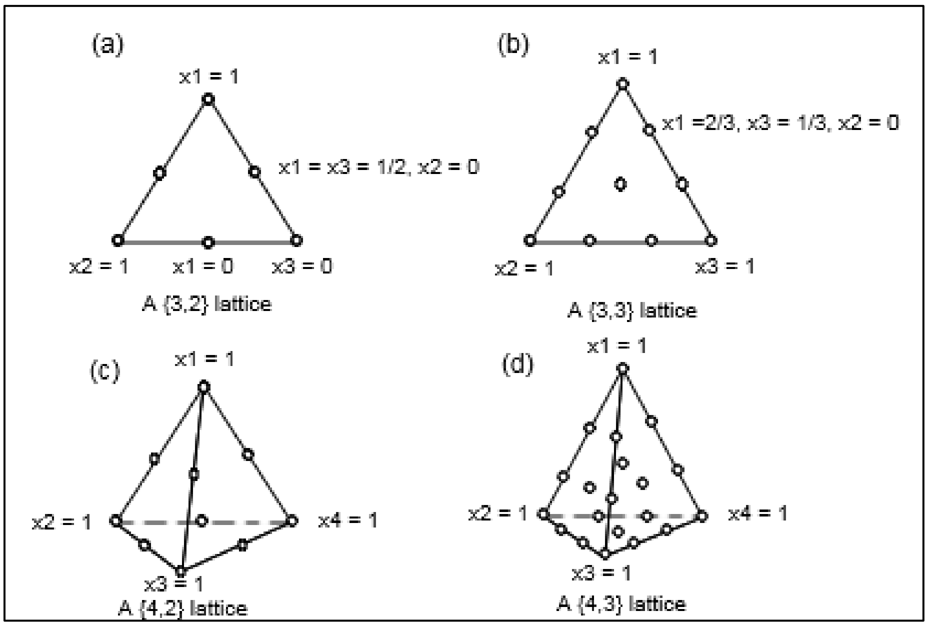


Figure 2: "Simplex lattice designs for q=3 and q=4 components" recreated from Cornell, 2002.

If we look at the difference between the {3,2} and {3,3} lattice, we notice that there are an increased number of points along the axis. There are only 3 points on the {3,2} lattice where there are 4 points on the {3,3} lattice. Additionally, the {3,3} lattice includes and axial point in the center of the region.

Polynomial Model

In mixture experiments, the factors all sum to a constant (an added dependence) so a traditional full model will not be estimable. In other words, since we added a constraint to our design, we will no longer be able to fit the models traditionally used. Therefore, the Scheffé polynomial models, also called canonical polynomials, are used. This model estimates one less term by removing the intercept from the model. The model includes all linear main-effect terms and the two-factor interaction terms, but does not include squared terms. The models can be seen in Figure 3.

$$\boldsymbol{Linear}: \boldsymbol{E}[\boldsymbol{y}] = \sum_{\boldsymbol{i}=1}^{\boldsymbol{q}} \boldsymbol{\beta}_{\boldsymbol{i}} \boldsymbol{x}_{\boldsymbol{i}}$$

$$Quadratic: E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}\sum_{j=2}^{q} \beta_{ij} x_i x_j$$

$$Full\ Cubic: E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}\sum_{j=2}^{q} \beta_{ij} x_i x_j + \sum_{i<j}\sum_{j=2}^{q} \delta_{ij} x_i x_j (x_i - x_j) + \sum_{i<j}\sum_{j<k}\sum_{k=3}^{q} \beta_{ijk} x_i x_j x_k$$

$$Special\ Cubic: E[y] = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}\sum_{j=2}^{q} \beta_{ij} x_i x_j + \sum_{i<j}\sum_{j<k}\sum_{k=3}^{q} \beta_{ijk} x_i x_j x_k$$

Figure 3: Canonical Polynomials, notation from Cornell 2002

Each $\boldsymbol{\beta_i}$ is the expected response for the pure mixture $\boldsymbol{x_i}$=1, $\boldsymbol{x_j}$=0, j≠I and is the height of the mixture surface at the vertex $\boldsymbol{x_i = 1}$. The amount of each polynomial given by $\sum_{i=1}^{q} \boldsymbol{\beta_i x_i}$ is called the linear blending portion. The linear canonical polynomial is appropriate when the blending is strictly additive. The quadratic blending, also referred to as synergism as the blending is non-linear, is often necessary when the linear relationship is not sufficient. Positive values of $\boldsymbol{\beta_{ij}}$ represent synergistic blending where negative values represent antagonistic blending. The cubic model can be used to account for both synergistic and antagonistic blending across a single edge. This is represented by the $\boldsymbol{\delta_{ij}}$ terms. The $\boldsymbol{\delta}$ terms represent the quadratic effects and are incredibly useful when the blends are not strictly complementary to one another. These two effects can be seen in figure 4. The $\boldsymbol{\beta_{ijk}}$ terms account for ternary blending among three separate components in the interior of the design.

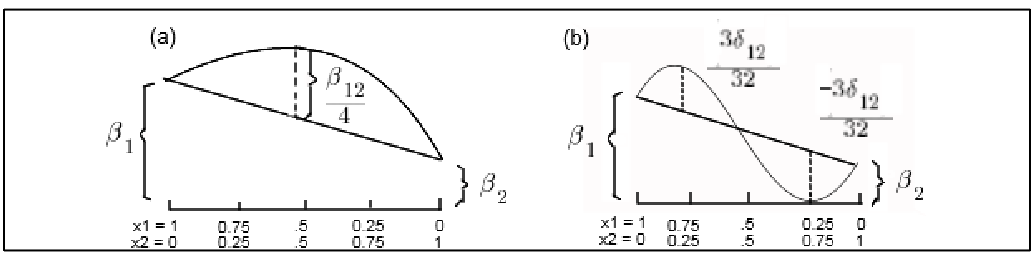


Figure 4: "Nonlinear blending. (a) Quadratic blending with $\boldsymbol{\beta_{ij} > 0}$. (b) Cubic blending with $\boldsymbol{\delta_{12} > 0}$" recreated from Myers et al., 2016.

Simplex Centroid Design

Prediction in the middle of the model would be difficult or risky using a simplex-lattice design since those designs do not contain test points in the center of the design region. The simplex centroid design attempts to correct for this by introducing a center point to the design. A simplex centroid design of q components is composed of pure mixture runs, all combinations of 2 to the k factors at equal levels, and a center point run with equal amounts of all ingredients. Therefore, the design will consist of $2^q - 1$ distinct design points. Figure 5 shows an example for q=3 and q=4.

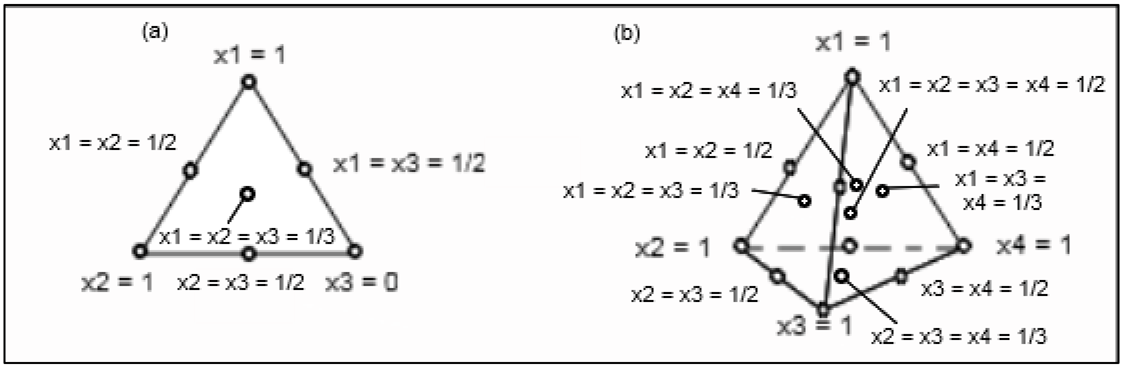


Figure 5: "Simplex-centroid designs with three and four components. (a) q=3. (b) q=4" recreated from Myers, et al 2016.

This design allows for a polynomial of the form:

$$E[y] = \sum_{i=1}^{q} \boldsymbol{\beta_i x_i} + \sum_{i<j}\sum_{j=2}^{q} \boldsymbol{\beta_{ij} x_i x_j} + \sum_{i<j}\sum_{j<k}\sum_{k=3}^{q} \boldsymbol{\beta_{ijk} x_i x_j x_k} + \cdots \beta_{12\ldots q} x_1 x_2 \ldots x_q$$

Note that when q =3, this is the same as the special cubic polynomial (as seen in figure 3). Since the cubic model is the natural result of these designs, they are incredibly powerful if the experimenter suspects there are cubic terms in the final model.

Augmenting Simplex Designs

The simplex designs that have been discussed place all of the design points, with the exception of the centroid, on the boundary of the simplex. This means that almost all of the points represent either a pure blend or a binary blend. We only have a single point (if a centroid is included) that gives information about a complete mixture. Since there is often a desire to understand the performance of complete mixtures, augmenting the simplex designs with axial runs and the overall centroid can be a powerful tool.

The axis of component i is defined as the line or ray extending from the base point $x_i = 0, x_j = \frac{1}{q-1}$ for all j≠i to the opposite vertex where $x_i = 1, x_j = 0$ for all j≠i. That is, this line connects the corner points to the midpoints of the opposite plane. This can be seen in Figure 6.

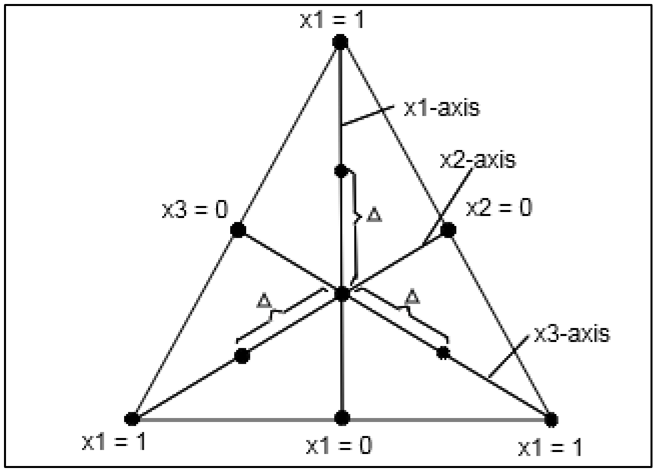


Figure 6: "The axes for components $x_1, x_2, and\ x_3$" recreated from Myers, et al 2016.

In the image above, the axial points are positioned along the component axes a distance of Δ from the centroid. Myers, et al. (2016) recommend that axial runs be placed midway between the centroid of the simplex and each vertex such that $\Delta = \frac{q-1}{2q}$. These points are also referred to as axial check blends as axial runs are often used to check the adequacy of the fit of an initial model. Figure 7 shows an example of a {3,2} simplex-lattice design augmented with axial runs (total of 10 runs in the design).

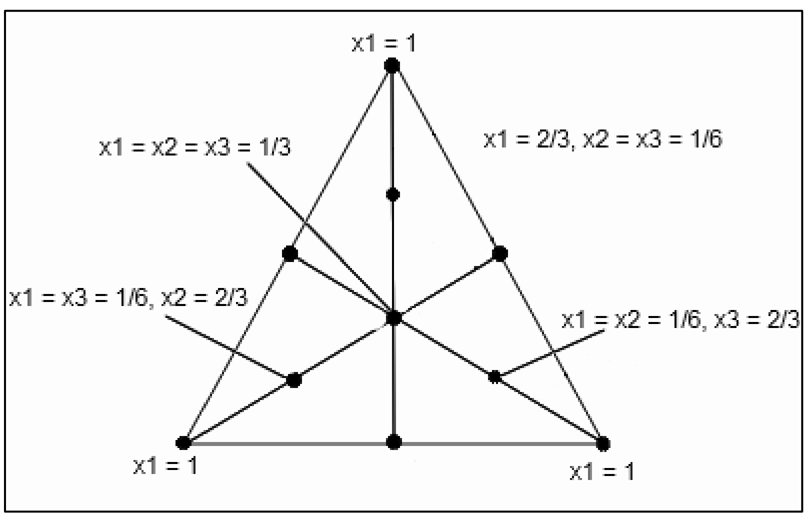


Figure 7: "An augmented simplex-lattice design" recreated from Myers, et al 2016.

This design is comparable to Figure 2(b) that contains the {3,3} simplex lattice. Both of these designs have 10 points, but this augmented simplex-lattice design has 4 complete mixture points while trading off 3 binary points. This tradeoff is something to consider when assessing the objectives of a test. These designs are commonly used for screening experiments or when component effects are of interest.

Optimal designs

Optimal designs for mixture models can use the same criterion as traditional optimal designs, the most common being D and I optimal. For more information on these criteria see Myers, et al. (2016). Note that the optimal design for a first order model is a pure mixture only design, for a second order model it is a simplex-lattice design, and for a cubic model it is a simplex-centroid design. Optimal designs however allow us to specify exactly what model we wish to build and construct a design most optimal given the number of runs allotted. This is also useful when we have an unusual number of runs and cannot use one of the traditional designs. Additionally, Goos and Jones (2011) note that ingredients used in mixture experiments are often mixtures themselves. This introduces additional constraints on the design as the lower and upper bound of the proportion of an ingredient is no longer 0 to 1. The example presented by Goos and Jones has three ingredients in a mixture. However, the proportions (a_1, a_2, a_3) do not all have the same ranges. Instead:

$$0.2 \leq a_1 \leq 0.8$$

$$0.2 \leq a_2 \leq 0.8$$

$$0 \leq a_3 \leq 0.6$$

Figure 8 shows this constrained design region.

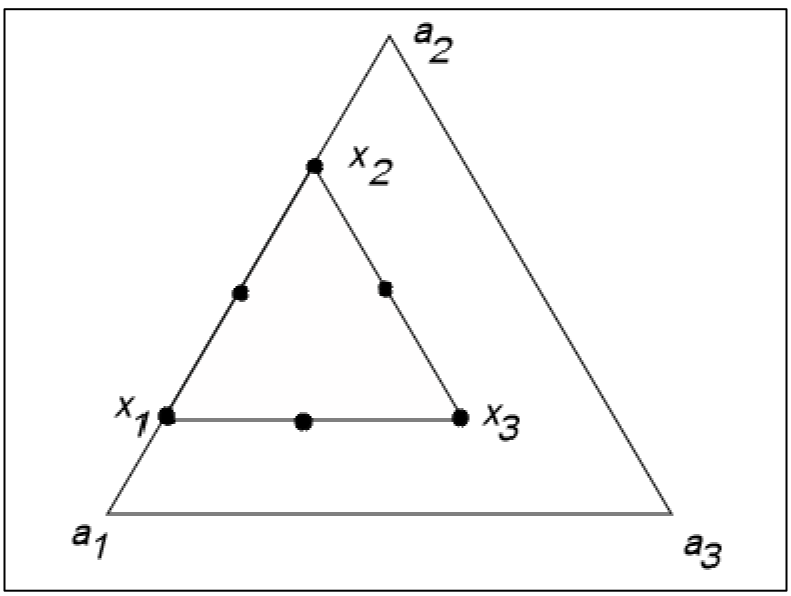


Figure 8: "Optimal design for a second-order Scheffé model in three ingredients in the presence of lower bounds on the ingredient proportions" recreated from Goos and Jones 2011.

The design choice is still the same, however this is in an increasingly constrained region. We must now define pseudo-components, the more specific components that make up the a_i components, as:

$$x_i = \frac{a_i - L_i}{1 - \sum_{i=1}^{q} L_i}$$

where L_i is the lower bound for proportion a_i. Now, the design is more realistic as the pseudo-components will take on values from 0 to 1 but the a_i components will not. For example, a run in which $x_1 = 1 \; and \; x_2 = x_3 = 0$, then $a_1 = .8, a_2 = .2, and \; a_3 = 0$

Space filling designs

Space filling designs attempt to do as their name suggests; fill the entire space. These designs spread the points throughout the design region. Within JMP statistical software, there are two different optimality criteria that can be used. These two methods are MaxPro and Centroid, which are both Fast Flexible Filling design criteria. Let p be the number of factors and n be the specified sample size. The MaxPro criterion attempts to find points in clusters that minimize the following:

$$C_{MaxPro} = \sum_{i}^{n-1} \sum_{j=i+1}^{n} {}^{1}\Big/ \prod_{k=1}^{p} (x_{ik} - x_{jk})^2$$

“The MaxPro criterion maximizes the product of the distances between potential design points in a way that involves all factors” (JMP). These designs maximize the space-filling properties on projections to all possible subsets of factors. The centroid method differs by placing a design point at the centroid of each cluster. “It has the property that the average distance from an arbitrary point in the design space to its closest neighboring design point is smaller than for other designs” (JMP). For more information see Hanson (2008).

Conclusion

Mixture experiments are a common test that presents a unique statistical problem. The constrained design region and the inability to change a single factor at a time requires a special type of design. Several designs discussed are the simplex-lattice designs, simplex-centroid designs, optimal mixture designs, and space filling mixture designs. Each of these designs can be used depending on the objective of the test. Additionally, the polynomial that we wish to fit will help determine what type of design is best. Mixture designs can be a very powerful solution to a common experimental need and are applicable within the Department of Defense. Many research labs work with complex chemical solutions and these designs can be used to optimize the levels of each ingredient in the solution. Additionally, any textile that is a combination of several different materials can utilize these designs for testing different properties such as breaking strength. Mixture designs allow for a more efficient and effective test.

JMP Demo with analysis

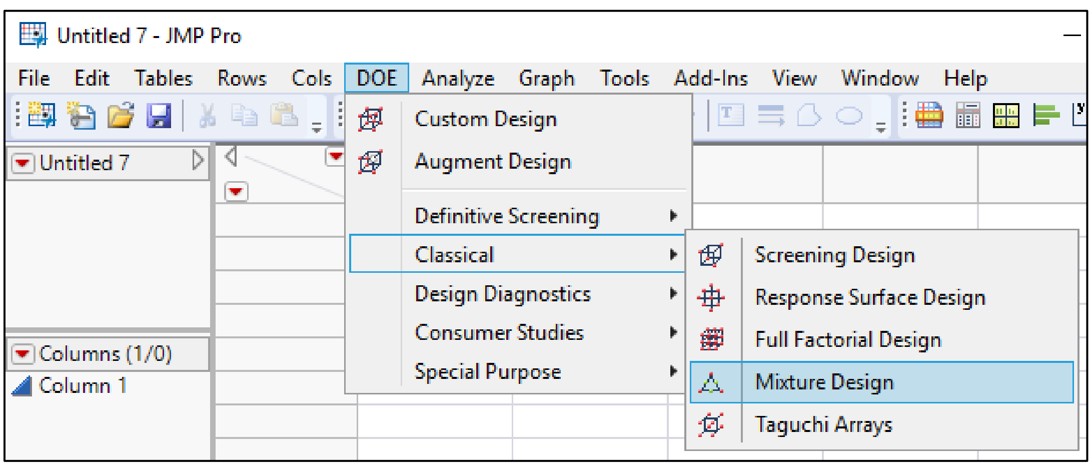
This example comes from Cornell, 2002. Three constituents- polyethylene (x_1), polystyrene (x_2), and polypropylene (x_3) were blended together and the resulting fiber material was spun to form yarn for draperies. A {3,2} simplex lattice design was used. The values of thread elongation in kilograms of force are measured and shown in the table below.

Observed Yarn Elongation Values

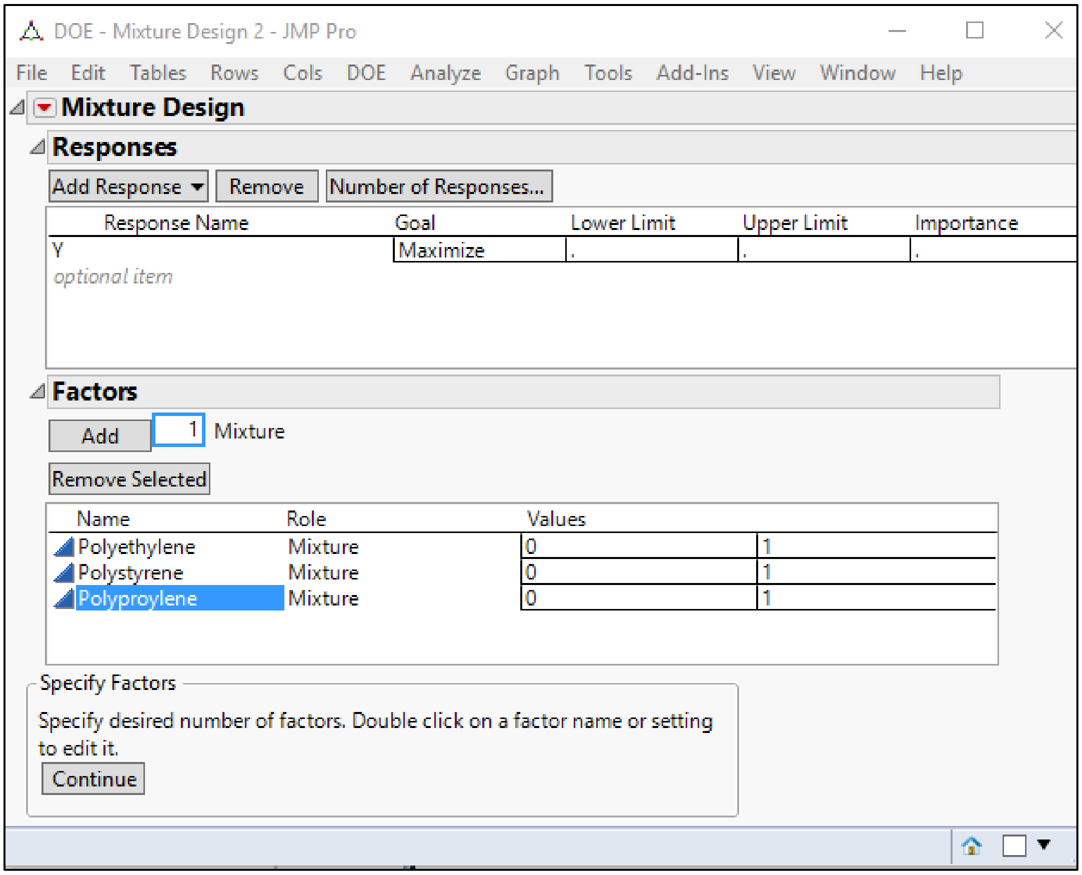
Design Point	Component Proportions x_1	x_2	x_3	Observed Elongation Values (y_n)	Average Elongation Value ($\bar{y}$)
1	1	0	0	11.0, 12.4	11.7
2	1/2	1/2	0	15.0, 14.8, 16.1	15.3
3	0	1	0	8.8, 10.0	9.4
4	0	1/2	1/2	10.0, 9.7, 11.8	10.5
5	0	0	1	16.8, 16.0	16.4
6	0	0	1/2	17.7, 16.4, 16.6	16.9

To create this design in JMP:

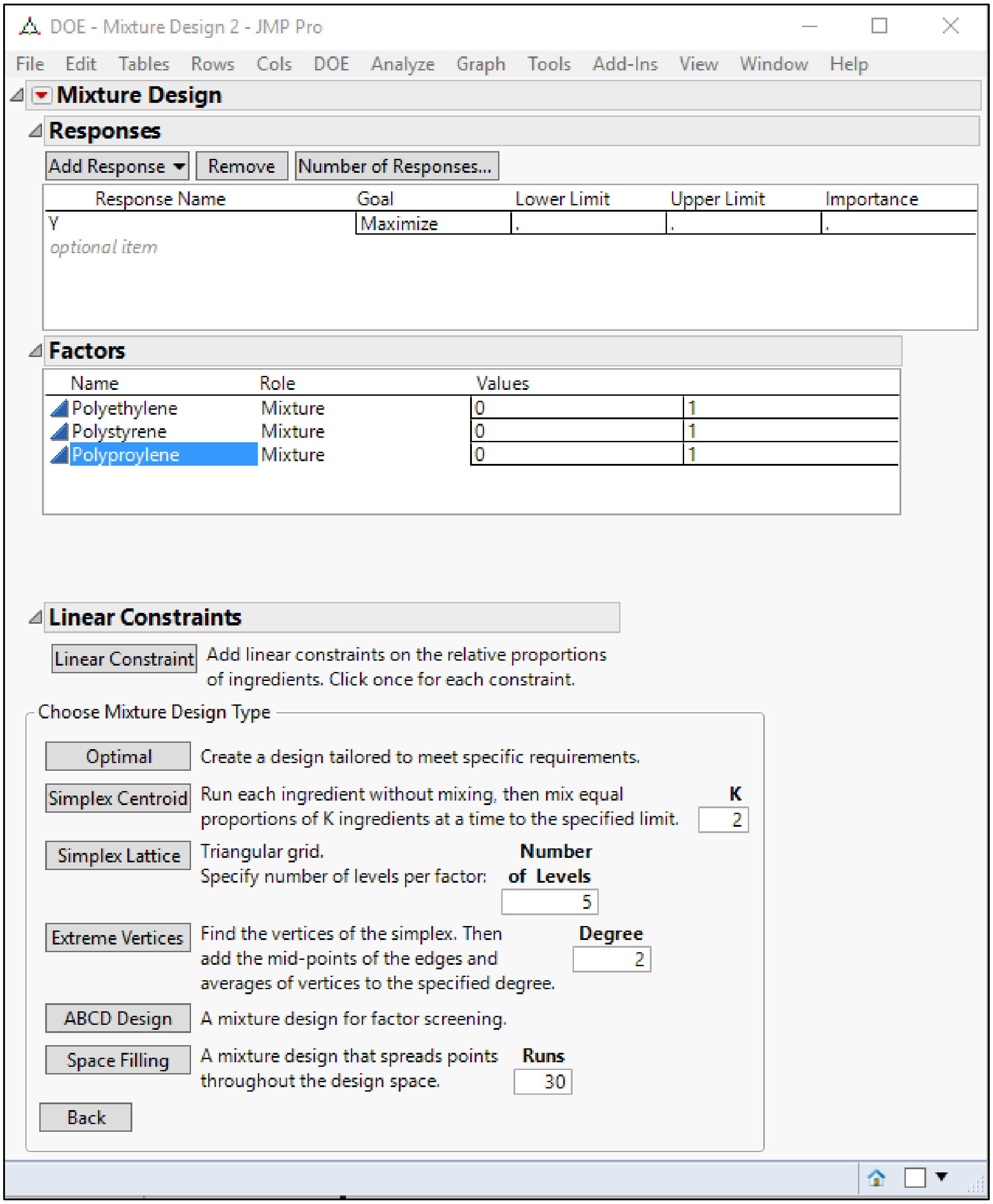
1. Open a New Data table
2. Select "DOE -> Classical -> Mixture Design"



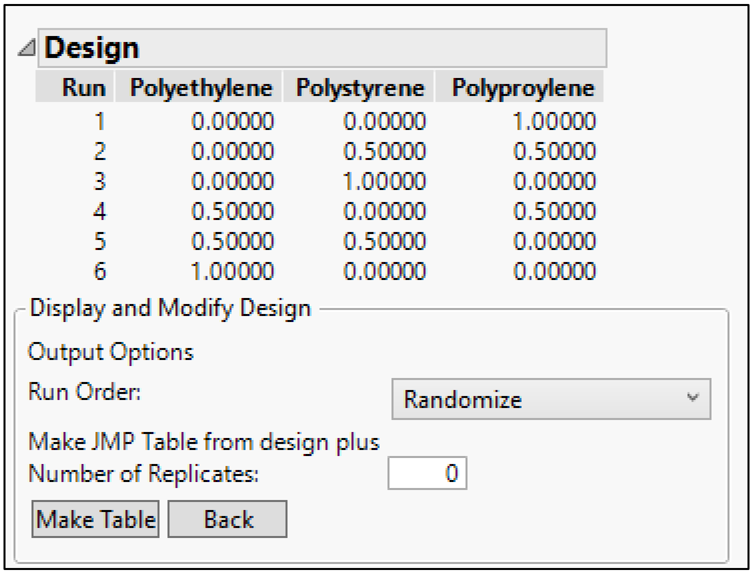
3. Enter the factors into the factor window. Select the role of the variable and the number of levels and enter in the appropriate values



4. Select “Continue”
5. JMP will provide a number of design options at this point. The optimal design, simplex centroid, simplex lattice, extreme vertices, ABCD Design, and space filling design. This paper did not discuss extreme vertices designs, but this design is used when not all of the ingredients can take values from 0-1 (creating a unique design space). The ABCD designs are designs that include axial runs. Inputs are required for several of the design choices. For this design, we are creating a simplex lattice design with the number of levels being 2.



6. Enter “2” into number of levels. This is the box where the image above currently has a “5”.
7. Select “Simplex Lattice”
8. JMP will output a Design. Select “Make Table”

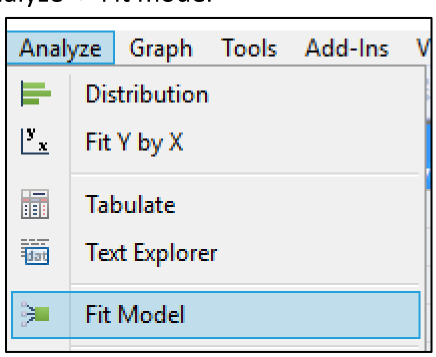


Run	Polyethylene	Polystyrene	Polyproylene
1	0.00000	0.00000	1.00000
2	0.00000	0.50000	0.50000
3	0.00000	1.00000	0.00000
4	0.50000	0.00000	0.50000
5	0.50000	0.50000	0.00000
6	1.00000	0.00000	0.00000

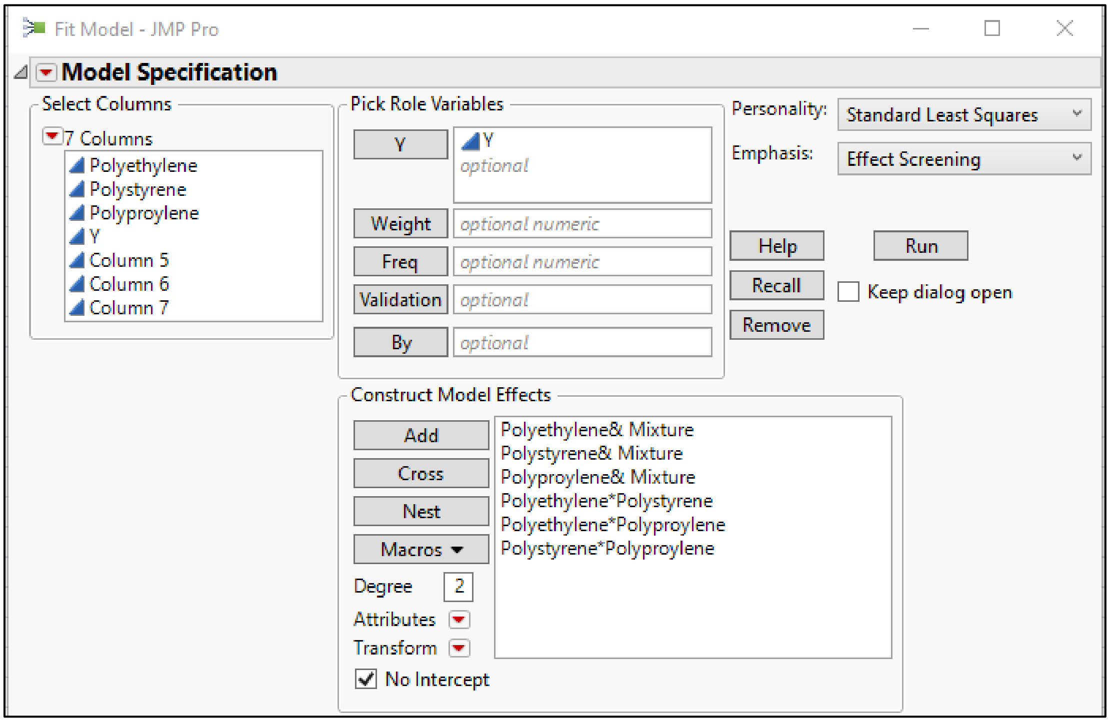
9. This will produce a single-run simplex lattice design. For this example, we will need to add the additional runs for the replicates. Not all runs have the same number of replications so this will need to be done manually. The final table should look as follows:

	Polyethylene	Polystyrene	Polyproylene	Y
1	0	0.5	0.5	10
2	0	1	0	8.8
3	0.5	0	0.5	17.7
4	0.5	0.5	0	15
5	1	0	0	11
6	0	0	1	16.8
7	0	0.5	0.5	9.7
8	0	1	0	10
9	0.5	0	0.5	16.4
10	0.5	0.5	0	14.8
11	1	0	0	12.4
12	0	0	1	16
13	0	0.5	0.5	11.8
14	0.5	0	0.5	16.6
15	0.5	0.5	0	16.1

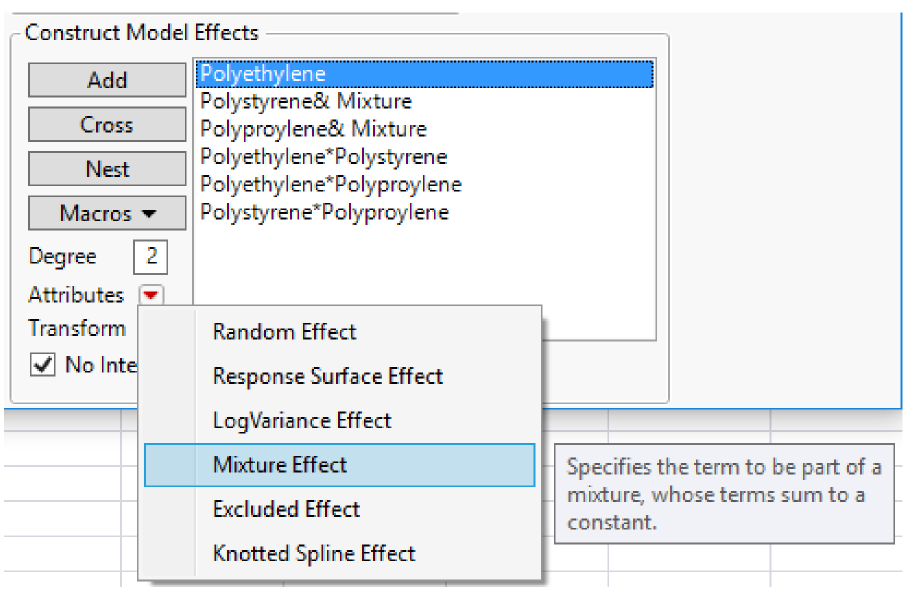
10. To analyze the data select "Analyze -> Fit model"



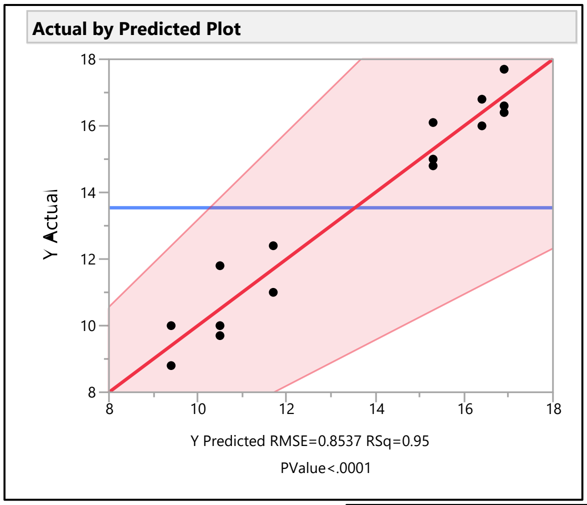
11. Select the "Y" variable and insert it into the "Y" window. Add the necessary factors that we wish to estimate into the "Construct Model Effects" window. For this example, we wish to estimate the main effects and the two factor interactions. Since we created this design using the Mixture models window in JMP, the main effects are already included with the "Mixture" attribute.



12. If the main effects were not already given the "Mixture" attribute, we could include this by selecting the main effect -> selecting the red drop down next to attribute -> Selecting "Mixture Effect". This is necessary so that we fit the appropriate polynomial (discussed previously) for a mixture design.



13. Select "Run"
14. The analysis will be completed. Several items that are of particular interest to us are the actual by predicted plot and the parameter estimates. The actual by predicted plot helps us visualize the accuracy of our model and displays an R-squared value that measures the percent of variability captured by the model. The parameter estimates tell us important information about each factor effect such as the estimate, the standard error, and the p-value.



Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Polyethylene(Mixture)	11.7	0.603692	19.38	<.0001 *
Polystyrene(Mixture)	9.4	0.603692	15.57	<.0001 *
Polyproylene(Mixture)	16.4	0.603692	27.17	<.0001 *
Polyethylene*Polystyrene	19	2.608249	7.28	<.0001 *
Polyethylene*Polyproylene	11.4	2.608249	4.37	0.0018 *
Polystyrene*Polyproylene	-9.6	2.608249	-3.68	0.0051 *

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	5	128.29600	25.6592	35.2032
Error	9	6.56000	0.7289	**Prob > F**
C. Total	14	134.85600		<.0001 *

Tested against reduced model: Y=mean

The final model built is:

$$\hat{y}(x) = 11.7x_{Polyethylene} + 9.4x_{Polystyrene} + 16.4x_{Polyproylene} + 19x_{Polyethylene}x_{Polystyrene} + 11.4x_{Polyethylene}x_{Polyproylene} - 9.6x_{Polystyrene}x_{Polyproylene}$$

If the goal of this experiment is to produce yarn with high elongation, the single-component blend that produces the most optimal results is component 3. Components 1 and 2 and 1 and 3 have binary synergistic effects. Component 2 and 3 result in the yarn having a lower average elongation value than would be expected by averaging the elongation values of the yarn produced by a single-component blend. If a binary blend is desired, we would conclude that we should combine component 1 with either of the other components.

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