Table 12A.1 (*continued*)

Number of Factors	*Experimental Units*	*Fraction*	*Design Resolution*	*Design Generator**
9	16	$\frac{1}{32}$	III	$E = ABC$
				$F = BCD$
				$G = ACD$
				$H = ABD$
				$J = ABCD$
	64	$\frac{1}{8}$	IV	$G = ABCD$
				$H = ACEF$
				$J = CDEF$
	32	$\frac{1}{16}$	IV	$F = BCDE$
				$G = ACDE$
				$H = ABDE$
				$J = ABCE$
10	16	$\frac{1}{64}$	III	$E = ABC$
				$F = BCD$
				$G = ACD$
				$H = ABD$
				$J = ABCD$
				$K = AB$
	32	$\frac{1}{32}$	IV	$F = ABCD$
				$G = ABCE$
				$H = ABDE$
				$J = ACDE$
				$K = BCDE$
	64	$\frac{1}{16}$	V	$G = BCDF$
				$H = AGDB$
				$J = ABDE$
				$K = ABCE$
	128	$\frac{1}{8}$	V	$H = ABCG$
				$J = BCDE$
				$K = ACDF$

* Either the positive or negative half of the design generators may be used to construct the fractional design.

13 Response Surface Designs

The central topic in this chapter is constructing designs for efficiently estimating response surfaces from factorial treatment designs with quantitative factors. The nature of linear and quadratic response surfaces is discussed, and designs developed specifically for response surface experiments are described. The discussions include estimating response surface equations and methods to explore the surfaces. Special designs are presented for experiments with factors that are ingredients of mixtures.

13.1 Describe Responses with Equations and Graphs

The objective of all experiments includes describing the response to treatment factors. Throughout this text when treatment factors had quantitative levels we have characterized the response (y) to the factor levels (x) with the polynomial regression equation. For example, in Chapter 3 a polynomial equation was used to estimate the relationship between seed production of plants, y, and density of plants in the plot, x. The estimated quadratic regression equation was graphed as a curve, and we were able to visualize the response of seed production to plant density throughout the range of plant densities included in the experiment. One of the main advantages of the response curve includes the ability to visualize the responses throughout the range of factor levels included in the experiment.

Response Surface Graphs for Two Treatment Factors

The response equation can be displayed as a surface when experiments investigate the effect of two quantitative factors such as the effect of temperature and pressure on the rate of a chemical reaction. In Chapter 6 a quadratic polynomial for two quantitative factors, salinity of media and number of days, was estimated to characterize plant response.

The quadratic response equation is represented as a solid surface in the three-dimensional display of Figure 13.1a. The equation is displayed in Figure 13.1b as a contour plot with lines of equal response values similar to contours of equal elevation levels shown on topographic maps.

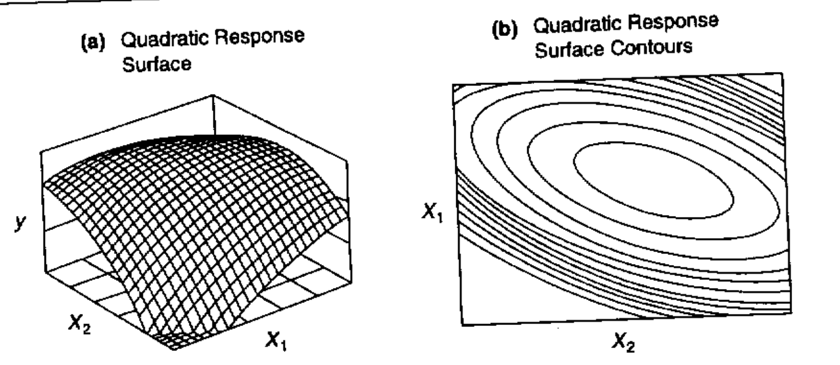

Figure 13.1 The response equation for two factors displayed as (a) a response surface and (b) contours of equal response

The response surface enables the investigator to visually inspect the response over a region of interesting factor levels and to evaluate the sensitivity of the response to the treatment factors. In certain industrial applications the response surfaces are explored to determine the combination of factor levels that provides an optimum operating condition, such as the combination of temperature and time to maximize the yield of chemical production. In other applications the surfaces are explored to find factor-level combinations that economically improve the responses over current operating conditions if it is too expensive to attain optimum conditions.

Response surfaces also can be used for analytical studies of fundamental processes. For example, they are used frequently in biological sciences to investigate the interplay of factors on the response variable, such as the interaction between nitrogen and phosphorus on the growth of plants.

Polynomial Models Approximate the True Response

Response surface design and analysis strategy assumes the mean of the response variable μ_y is a function of the quantitative factor levels represented by the variables $x_1, x_2, \ldots, x_k$. Polynomial models are used as practical approximations to the true response function. The true function commonly is unknown, and the polynomial functions most often provide good approximations in relatively small regions of the quantitative factor levels.

The most common polynomial models used for response surface analysis are the linear, or *first-order,* model and the quadratic, or *second-order,* model. The first-order model for two factors is

$$\mu_y = \beta_o + \beta_1 x_1 + \beta_2 x_2 \qquad \textbf{(13.1)}$$

and the second-order model is

$$\mu_y = \beta_o + \beta_1 x_1 + \beta_2 x_2 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{12} x_1 x_2 \qquad \textbf{(13.2)}$$

The contour plots for first-order models have series of parallel lines representing coordinates of the factor levels that produce equal response values. The contour plots for quadratic models are more complex with a variety of possible contour patterns. One such pattern shown in Figure 13.1b is a symmetrical mound shape contour with a maximum response occurring within the central contour. Four other patterns are shown in Figure 13.2. Figure 13.2a demonstrates a contour plot with a minimum response occurring within the center contour, indicating a symmetrical surface with a depression in the center. Figure 13.2b depicts a rising ridge with the maximum occurring outside the experimental region. Figure 13.2c shows a stationary ridge in the center of the plot with a decreasing response to the right or left of the center line of maximum response. A saddle contour plot or minimax is portrayed in Figure 13.2d in which the response can increase or decrease from the center of the region, depending on the direction of movement from the center.

Sequential Experiments for Response Surface Analysis

Box and Wilson (1951) accelerated the promotion of response surface analysis for industrial applications. Their primary theme was the use of sequential experimentation with the purpose of determining the optimum operating conditions for an industrial process.

The general approach begins with 2^n factorial treatment designs to identify factors that influence the process. Subsequent experiments use factor treatment combinations to locate an area in the factor space that most likely produces optimum responses. Ultimately, a 2^n factorial arrangement in this region is augmented with treatment combinations to characterize the response surface with quadratic polynomials.

Time Scales Can Prevent Effective Sequential Experiments

In some fields of application the time scale for completion of experiments prohibits sequential experimentation. Many biological studies may require months to complete a single experiment. Often accumulated information from previous biological studies enables the investigator to identify the regions of optimum response, and experiments can be designed to explore the response surface in those regions.

The objective of this chapter is to present some of the basic designs and methods of analysis used for identifying optimum conditions in sequential

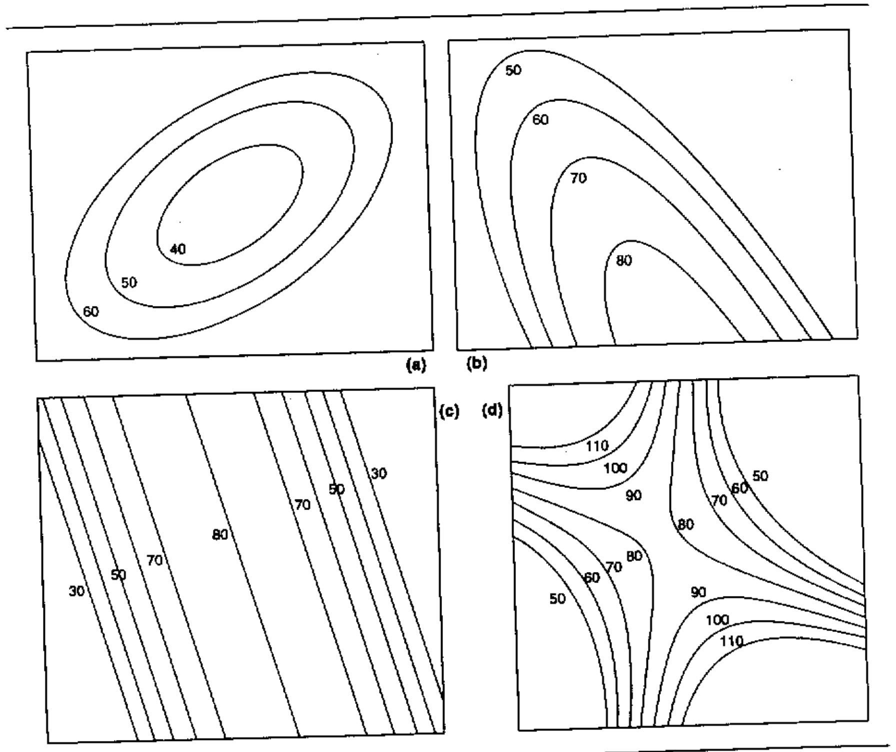

Figure 13.2 Contour plots for a (a) minimum surface, (b) rising ridge, (c) stationary ridge, and (d) saddle

experimentation and to present designs for efficiently estimating the response surface equations when the region of optimum response is identified.

13.2 Identify Important Factors with 2^n Factorials

Complete or fractional factorial experiments are used for the initial experiments conducted in the study of response surfaces. When the region of optimum response is unknown, the 2^n factorials or fractions of the 2^n factorials are used to identify factors that affect the response variable. (These designs were discussed in Chapters 11 and 12.)

Estimate Linear Responses to Factors

The 2^n factorials are suitable designs to estimate the mean responses for the *linear*, or *first-order*, model in Equation (13.1). The inclusion of two or more observations at the middle level of all factors is the usual recommended procedure to estimate

experimental error and to provide a means to evaluate the adequacy of the linear response surface model.

An extensive coverage of methods to identify important factors and factor levels in the region of optimum response conditions can be found in Box and Draper (1987) and in Myers and Montgomery (1995). A brief introduction to the identification of important factors is illustrated with an industrial chemistry experiment to evaluate the factors that affect the vinylation of methyl glucoside.

Example 13.1 Vinylation of Methyl Glucoside

Vinylation of methyl glucoside occurs when it is added to acetylene under high pressure and high temperature in the presence of a base to produce monovinyl ethers. The monovinyl ether products are useful for various industrial synthesis processes. The results of a study on vinylation of methyl glucoside by Marvel et al. (1969) are used to illustrate the methods to identify and evaluate important factors for response surface characterization. The ultimate goal of the project was to determine which conditions produced maximum conversion of methyl glucoside to each of several monovinyl isomers.

Some methods to identify important factors with first-order response surfaces are illustrated with two factors, pressure and temperature. The treatment design was a 2^2 factorial with "temperature" at 130° and 160° C and "pressure" at 325 and 475 psi as factors. Four replications were conducted in the center of the experimental region at a temperature of 145° C and a pressure of 400 psi to provide an estimate of experimental error variance and to evaluate the adequacy of the linear response model. The treatment combinations and percent conversion of methyl glucoside are shown in Table 13.1.

Table 13.1 Vinylation of methyl glucoside in a 2^2 factorial plus four replications at the design center with Temperature and Pressure as factors

Original Factors		*Coded Factors*		
Temperature	*Pressure*	x_1	x_2	*%Conversion*
130	325	−1	−1	8
160	325	+1	−1	24
130	475	−1	+1	16
160	475	+1	+1	32
145	400	0	0	21
145	400	0	0	23
145	400	0	0	20
145	400	0	0	24

Coded Factor Levels for Convenience

Coded factor levels provide a uniform framework to investigate factor effects in any experimental context since the actual values of factor levels depend on the particular factors in the study. Coded levels for the 2^n factorial design factors are

$$x_i = \frac{(A_i - \overline{A})}{D} \quad \textbf{(13.3)}$$

where A_i is the ith level of factor A, $\overline{A}$ is the average level for factor A; and D is $\frac{1}{2}(A_2 - A_1)$. Coded levels of temperature (T) and pressure (P) in Table 13.1 are

$$x_1 = \frac{T - 145}{15} \quad \text{and} \quad x_2 = \frac{P - 400}{75} \quad \textbf{(13.4)}$$

Estimates of the Linear Responses

The estimates of the coefficients for the first-order model in Equation (13.1) a

$$\widehat{\beta}_0 = \overline{y} = \frac{1}{4}(8 + 24 + 16 + 32) = 20$$

$$\widehat{\beta}_1 = \frac{1}{2}T = \frac{1}{4}(-8 + 24 - 16 + 32) = 8$$

$$\widehat{\beta}_2 = \frac{1}{2}P = \frac{1}{4}(-8 - 24 + 16 + 32) = 4$$

The estimates of the linear coefficients, β_1 and β_2, are one-half of the factorial treatment effect estimates for a 2^2 factorial (see Chapter 11).

The variance of the four observations at the design center is $s^2 = 3.33$, and an estimate of the standard error for the coefficient estimates is

$$s_{\widehat{\beta}} = \sqrt{\frac{4}{16}(3.33)} = 0.91$$

Whether the experimental error variance is adequately estimated with replication only at the center of the design factor levels can matter. If the variance of the response in any way depends on the factor level, then replication of the design at the high- and low-factor-level combinations is recommended to detect any heterogeneous variability among the treatment combinations.

The estimates of the regression coefficients indicate that increases in Temperature or Pressure will increase the vinylation of methyl glucoside. The estimated first-order model equation is

$$\widehat{y} = 20 + 8x_1 + 4x_2$$

The temperature and pressure interaction TP measures lack of fit to the linear model and is represented by the term $\beta_{12}x_1x_2$ in the quadratic model in Equation (13.2). The estimate of the coefficient β_{12} is one-half of the TP interaction, or

$$\widehat{\beta}_{12} = \frac{1}{2}TP = \frac{1}{4}(8 - 24 - 16 + 32) = 0$$

The standard error of $\widehat{\beta}_{12}$ is 0.91, the same as that for the linear coefficients. The estimated interaction component of 0 indicates that Temperature and Pressure are acting independently on the conversion.

Center Design Points to Evaluate Surface Curvature

Replicate observations at the design center not only provide an estimate of experimental error, but they also provide a means to measure the degree of curvature in the experimental region. Let $\overline{y}_f$ be the mean of the four treatment combinations for the 2^2 factorial and $\overline{y}_c$ be the mean of the center points. There is some evidence for curvature on the response surface if the average response in the center of the design coordinates, $\overline{y}_c$, is larger or smaller than the average response at the extreme levels of the factors, $\overline{y}_f$. The difference $(\overline{y}_f - \overline{y}_c)$ is an estimate of $\beta_{11} + \beta_{22}$, where β_{11} and β_{22} are the quadratic regression coefficients in Equation (13.2). The observed means are $\overline{y}_f = 20$ and $\overline{y}_c = 22$, with a difference of $\overline{y}_f - \overline{y}_c = -2$. The standard error of the difference is estimated as $\sqrt{3.33(\frac{1}{4} + \frac{1}{4})} = 1.29$; the linear response appears to adequately describe the surface in this region.

The contour plot for the estimated linear response equation is shown in Figure 13.3. The values of the contours ascend as the levels of Temperature and Pressure increase. Ascending contours indicate that a combination of Temperature and Pressure to maximize conversion may exist in a direction perpendicular to the contours.

Path of Steepest Ascent to an Optimum Response

Ultimately, the investigator will want to characterize the region of optimum response. To do so requires the investigator to locate the region of factor levels that produces optimum conditions. The **method of steepest ascent** is a procedure developed to move the experimental region in the response variable in a direction of maximum change toward the optimum.

Based on the estimated linear equation $\widehat{y} = 20 + 8x_1 + 4x_2$, the path of steepest ascent perpendicular to the contours of equal response moves 4 units in the x_2 direction for every 8 units in the x_1 direction. Equivalently, the path has a movement of 4/8 = 0.5 unit in x_2 for every 1 unit movement in x_1.

The path of steepest ascent is started at the center of the design with $(x_1, x_2) = (0, 0)$. The center of the design for values of temperature and pressure is $(T, P) = (145, 400)$ in Figure 13.3. A change of $\Delta x_1 = 1$ unit in the x_1 direction is a 15°C change in Temperature and a $\Delta x_2 = 0.5$ unit in the x_2 direction is a 37.5 psi change in Pressure.

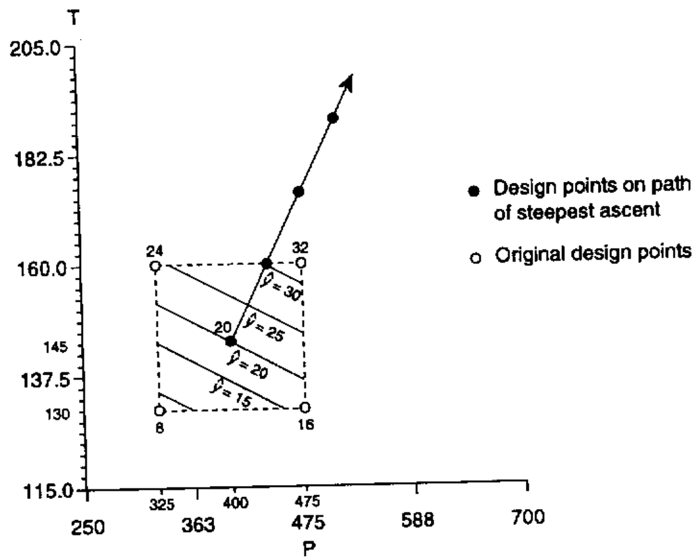

Figure 13.3 Contour plot for the linear response of methyl glucoside vinylation, % conversion, to temperature (T) and pressure (P)

The objective is to move along the path of steepest ascent until a maximum response is observed. The chemist will perform experiments at combinations of Temperature and Pressure along the path of steepest ascent. Suppose the chemist wants to make changes relative to a 1-unit change in x_1. The levels of Temperature and Pressure along the path beginning at $(T, P) = (145, 400)$, the design center, with 1-unit changes in x_1 and one-half unit changes in x_2 are shown in Table 13.2.

Table 13.2 Path of steepest ascent to search for region of maximum response in vinylation of methyl glucoside

Step	x_1	x_2	T	P
0	0	0	145	400.0
1	1	0.5	160	437.5
2	2	1.0	175	475.0
3	3	1.5	190	512.5
4	4	2.0	205	550.0
⋮	⋮	⋮	⋮	⋮

Eventually, as the chemist advances along the path of steepest ascent the increases in the response become smaller until an actual decrease is observed in the response. The decrease in response should indicate that the region of maximum response is in the neighborhood of the current temperature and pressure conditions.

At that point in the process an experiment can be designed to estimate a quadratic polynomial equation that approximates the response surface.

13.3 Designs to Estimate Second-Order Response Surfaces

A new experiment has to be designed to characterize the response surface once the region of optimum response is identified. The surface usually is approximated by a quadratic equation to characterize any curvature in the surface.

The 2^n factorials or fractions thereof are useful designs to identify the important factors and regions of optimum response. However, in the region of optimum response, these designs provide insufficient information to estimate quadratic response equations. At least three levels are required for each factor, and the design must have $1 + 2n + n(n-1)/2$ distinct design points to estimate the parameters in a quadratic regression model for approximation to the curved surface.

Desirable properties for experimental designs for response surface estimation include the ability to estimate experimental error variance and allow for a test of lack of fit to the model. Designs should also efficiently estimate the model coefficients and predict responses.

Several classes of designs with these desirable properties that have been developed for second-order response surface approximation are discussed in this section.

3^n Factorials for Quadratic Surface Estimation

The 3^n factorials can be used to estimate the quadratic polynomial equations. However, the number of treatment combinations required by the 3^n factorials leads to an impractical experiment size. While a 3^n design with two factors only requires 9 treatment combinations, a design with three factors requires 27 and one with four factors requires 81 treatment combinations.

Central Composite Designs Are an Alternative to 3^n Factorials

Box and Wilson (1951) introduced **central composite designs** requiring fewer treatment combinations than 3^n factorials to estimate quadratic response surface equations. The central composite designs are 2^n factorial treatment designs with $2n$ additional treatment combinations called *axial points* along the coordinate axes of the coded factor levels. The coordinates for the axial points on the coded factor axes are $(\pm\alpha, 0, 0, \ldots, 0)$, $(0, \pm\alpha, 0, \ldots, 0)$, $\ldots$, $(0, 0, 0, \ldots, \pm\alpha)$. Generally, m replications are added to the center of the design at coordinate $(0, 0, \ldots, 0)$.

The central composite designs are used to advantage in sequential experimentation. The first step of the sequence consists of a series of tests conducted along a path of steepest ascent, such as that illustrated in Table 13.2. Eventually, the tests lead to a set of factor levels that provide an apparent maximum on the path. For example, suppose the responses on the path of steepest ascent are those shown in Figure 13.4, with a maximum response of 36 observed at $T = 190°C$ and $P = 512.5$ psi on the path.

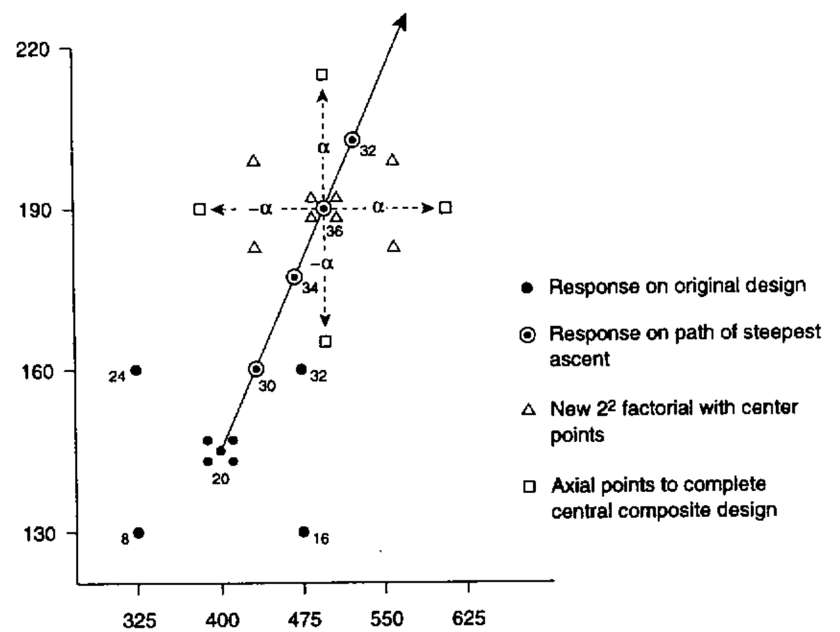

Figure 13.4 Path of steepest ascent and a central composite design

As a second step in the sequence the chemist can conduct a new 2^2 factorial experiment with several replications at the design center of $(T, P) = (190, 512.5)$.

Suppose the difference $(\bar{y}_f - \bar{y}_c)$ computed from the new experiment indicates a high degree of curvature on the surface. The third step in the sequential experiment consists of additional runs of the experiment at the axial points $(\pm\alpha, 0)$ and $(0, \pm\alpha)$ shown in boxes in Figure 13.4. This last set of treatment combinations on the axes, along with the 2^2 factorial and center points, constitutes a central composite design as a result of the sequential experimentation.

One replication of a central composite design consists of $N_f = 2^n$ treatment combinations from the 2^n factorial, $N_a = 2n$ treatment combinations at the axial points in the design, and m replications at the center for a total of $N = N_f + N_a + m$ design observations.

The coordinates on the coded x_1 and x_2 axes for the central composite design with two factors are shown in Display 13.1. A graphic display of the coordinate locations for the coded factor levels of two- and three-factor central composite designs are depicted in Figure 13.5. A quadratic equation can be estimated from this design because each factor has five levels. In addition, as we shall see in the

Display 13.1 Central Composite Design Coordinates

2^2 Design		*Axial*		*Center**	
x_1	x_2	x_1	x_2	x_1	x_2
-1	-1	$-\alpha$	0	0	0
$+1$	-1	$+\alpha$	0		
-1	$+1$	0	$-\alpha$		
$+1$	$+1$	0	$+\alpha$		

*m replications

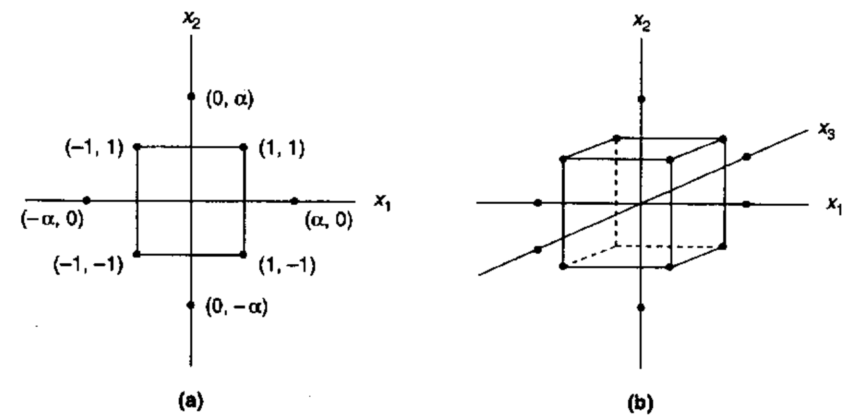

Figure 13.5 Central composite designs for (a) two factors and (b) three factors

next section, any significant deviations from the quadratic approximation can be evaluated.

The $N = 2^n + 2n + m$ experimental units required for the central composite design with n factors are fewer than those required by 3^n factorials with three or more factors. Thus, the central composite designs are more economical in the use of experimental resources and provide the ability to estimate quadratic response equations. Fractions of the 2^n designs with high-order interactions aliased can be used as the 2^n design base when there are many factors in the study.

Rotatable Designs to Improve Response Surface Explorations

Equal precision for all estimates of means is a desirable property in any experimental setting. However, the precision of the estimated values on the response surface based on the estimated regression equation will not be constant over the entire experimental region. A property of *rotatability* developed for central composite designs requires that the variance of estimated values be constant at points equally distant from the center of the design which is coded coordinates $(0, 0, \ldots, 0)$.

Rotatability of a design becomes important in the exploration of a response surface because the precision of the estimated surface does not depend on the orientation of the design with respect to the true response surface or the direction of the search for optimum conditions. The 2^n factorials used as first-order designs to implement the method of steepest ascent searches for regions of optimal responses are rotatable designs. Thus, the orientation of the design does not hinder the method of steepest ascent search because some responses are estimated with less precision than others.

The central composite design can be made rotatable by setting the axial point values as $\alpha = (2^n)^{1/4}$. The value of α for a two-factor design is $\alpha = (4)^{1/4} = \sqrt{2} = 1.414$, and for a three-factor design $\alpha = (8)^{1/4} = 1.682$. If there are r_f replications of the 2^n factorial and r_a replications of the axial treatment combinations a more general form for α is $\alpha = (r_f 2^n / r_a)^{1/4}$. If a 2^{n-p} fractional factorial is used as the basis for the central composite design, then $\alpha = (r_f 2^{n-p}/r_a)^{1/4}$.

Example 13. 2 Rotatable Design for Vinylation of Methyl Glucoside

Suppose the path of steepest ascent for the methyl glucoside study in Table 13.2 provided a maximum response at $T = 190°C$ and $P = 512.5$ psi and a rotatable central composite design is to be constructed with the design center at $(T, P) = (190, 512.5)$. Also, the relationship between the design coordinates (x_1, x_2) and temperature and pressure levels (T, P) remain as before where a change of one unit in x_1 is 15°C and a change of one unit in x_2 is 75 psi. With $\alpha = \sqrt{2}$, the design coordinates and the required temperature and pressure settings will be

	Axial				Center	2^n Design			
x_1	$-\sqrt{2}$	$+\sqrt{2}$	0	0	0	-1	$+1$	-1	$+1$
x_2	0	0	$-\sqrt{2}$	$+\sqrt{2}$	0	-1	-1	$+1$	$+1$
T	169*	211	190	190	190	175	205	175	205
P	512.5	512.5	406.4	618.6	512.5	437.5	437.5	587.5	587.5

*Example calculation, $169° = 190° - \sqrt{2}(15°)$

Designs for Uniform Precision in the Center of the Design

As previously stated, the variance of the estimated surface is not constant over the entire surface. Box and Hunter (1957) showed that the number of center points in the rotatable central composite designs could be chosen to provide a design with uniform precision for the estimated surface within one unit of the design center coordinates on the coded scale. They reasoned that the investigator is most interested in the response surface near the center of the design when a stationary point of the surface is located near the center of the design. The stationary point is a point of maximum or minimum response or a saddle point as shown in Figure 13.2d.

Some central composite rotatable designs with uniform precision are shown in Table 13.3.

Table 13.3 Uniform precision rotatable central composite designs

Number of Factors	2	3	4	5	5	6	6
Fraction of 2^n	1	1	1	1	$\frac{1}{2}$	1	$\frac{1}{2}$
α	1.414	1.682	2.000	2.378	2.000	2.828	2.378
N_f	4	8	16	32	16	64	32
N_a	4	6	8	10	10	12	12
m	5	6	7	10	6	15	9
N	13	20	31	52	32	91	53

The central composite designs require five levels of each factor coded as $-\alpha, -1, 0, 1, \alpha$. In some instances the preparation of five levels for some factors may be too difficult, expensive, or time-consuming. The *face-centered cube design* is a variation of the central composite design with $\alpha = 1$ that requires only three levels of each factor. Substituting $\alpha = 1$ into Display 13.1, the design for two factors becomes a 3^2 factorial. The design is most attractive when the region of interest is a cuboidal region produced by the design rather than the spherical region produced by the central composite design.

The design is not rotatable but the absence of this desirable property may be offset by the desire to have a cuboidal inference region and also by the savings in experimental resources. The face-centered cube design requires fewer runs at the center point of the design than does the central composite design to achieve a stable variance of estimated values throughout the design region. However, it should be remembered that replicate runs are needed at some design point or points to estimate experimental error variance. A face-centered cube design for three or more factors requires fewer treatment combinations than the 3^n factorials; thus, it is another alternative to the 3^n design, requiring fewer experimental units.

Box–Behnken Designs Another Alternative to 3^n Factorials

A class of three-level designs to estimate second-order response surfaces was proposed by Box and Behnken (1960). The designs are rotatable, or nearly so, with a reduction in the number of experimental units compared to the 3^n designs. The designs are formed by combining 2^n designs with incomplete block designs. Details for construction can be found in Box and Draper (1987). The coded factor levels for the treatment combinations required in a design for three factors are shown in Display 13.2. A complete set of treatment combinations for a 2^2 factorial occurs for each pair of factors accompanied by the 0 level of the remaining factors. Several replications of the design center $(0, 0, \ldots, 0)$ are included.

Display 13.2 Box–Behnken Design Coordinates for a Three-Factor Design

Factor	*A*	*B*	*C*
Coded Level	x_1	x_2	x_3
	− 1	− 1	0
2^2 factorial	+ 1	− 1	0
for *A* and *B*	− 1	+ 1	0
	+ 1	+ 1	0
	− 1	0	− 1
2^2 factorial	+ 1	0	− 1
for *A* and *C*	− 1	0	+ 1
	+ 1	0	+ 1
	0	− 1	− 1
2^2 factorial	0	+ 1	− 1
for *B* and *C*	0	− 1	+ 1
	0	+ 1	+ 1
	0	0	0
Design center	0	0	0
	0	0	0

These designs are spherical rather than cuboidal since the design points fall on the edges of a cube rather than on the corners like those of the face-centered cube design. The Box–Behnken design should only be used if one is *not* interested in predicting responses at the corners of the cuboidal region.

Incomplete Block Designs to Increase Precision

Incomplete block designs are useful to reduce experimental error variance when the number of treatments is large or when the experimental conditions preclude the conduct of complete replications at one time or under the same conditions.

Box and Hunter (1957) gave the conditions for blocking second-order response surface designs so that the block effects do not affect the estimates of the parameters for the response surface equation. They showed that two conditions must be satisfied for the blocks to be orthogonal to the parameter estimates of the response surface equation. Let n_b be the number of treatments in the *b*th block. The two conditions necessary are

1. Each block must be a first-order orthogonal design. For each block the following relationship must hold for each pair of design variables x_i and x_j:

$$\sum_{k=1}^{n_b} x_{ik}x_{jk} = 0 \qquad i \neq j = 0, 1, 2, \ldots, n \tag{13.5}$$

2. The fraction of the total sum of squares for each design variable contributed by every block must be equal to the fraction of the total observations placed in the block. Thus, the following relationship must hold between the design variables and the number of observations for every block:

$$\frac{\sum_{k=1}^{n_b} x_{ik}^2}{\sum_{k=1}^{N} x_{ik}^2} = \frac{n_b}{N} \qquad i = 1, 2, \ldots, n \tag{13.6}$$

A suggested strategy for blocking the central composite design places the N_f treatments for the 2^n design and m_f design center points in one block and the N_a axial treatments with m_a design center points in a second block. This blocking arrangement satisfies the first condition, Equation (13.5).

The central composite rotatable design for two factors arranged into two blocks is shown in Display 13.3. The first block is composed of $N_f = 4$ treatment combinations of the 2^2 factorial plus $m_f = 2$ design center points, and the second block consists of $N_a = 4$ axial treatment combinations plus $m_a = 2$ design center points. The computations required to evaluate the first condition for an orthogonal block design are the sums of crossproducts between x_1 and x_2 in each block. It is easy to verify that $\Sigma\, x_1x_2 = 0$ in both blocks.

Display 13.3 Central Composite Rotatable Design for Two Factors in Two Incomplete Blocks

Factor	*A*	*B*
Coded Level	x_1	x_2
	− 1	− 1
	+ 1	− 1
Block 1	− 1	+ 1
	+ 1	+ 1
	0	0
	0	0
	1.414	0
	− 1.414	0
Block 2	0	1.414
	0	− 1.414
	0	0
	0	0

For the complete design

$$\sum_{k=1}^{12} x_{1k}^2 = \sum_{k=1}^{12} x_{2k}^2 = 8$$

and for both block 1 and block 2

$$\sum_{k=1}^{6} x_{1k}^2 = \sum_{k=1}^{6} x_{2k}^2 = 4$$

The number of treatment observations in blocks 1 and 2 are $n_1 = n_2 = 6$, and the total number of observations is $N = 12$ with a ratio $n_i/N = 6/12 = 1/2$. The second condition, Equation (13.6), requires that the ratio of the sums of squares of x_1 and x_2 in every block to that for the entire experiment be equal to n_i/N. For both block 1 and block 2 the sum of squares ratio is $4/8 = 1/2$, which is equivalent to the ratio for n_i/N; the design is orthogonal.

For the second condition to be satisfied the following relationship must hold:

$$\alpha^2 = n\left[\frac{1+p_a}{1+p_f}\right] \tag{13.7}$$

where $p_a = m_a/N_a$ and $p_f = m_f/N_f$. For the design to satisfy the two conditions and be rotatable $\alpha = (2^n r_f/r_a)^{1/4}$. It is not always possible to find a design that exactly satisfies Equation (13.7) with $\alpha = (2^n r_f/r_a)^{1/4}$, but in practice values of the design observation numbers can be determined to provide designs with near orthogonal blocking and rotatability. Box and Draper (1987) provide relative proportions of r_f and r_a required for rotatability and orthogonal blocking when $p_a = p_f$.

For the design in Display 13.3 the fraction of design center observations in each block is $p_a = p_f = 1/2$ and $\alpha = \sqrt{2}$.

Evaluating the condition for rotatability and orthogonality in Equation (13.7) we have

$$\alpha^2 = n\left[\frac{1+p_a}{1+p_f}\right] = 2\left[\frac{1+1/2}{1+1/2}\right] = 2$$

and $\alpha = \sqrt{2}$ as required for rotatability.

The central composite rotatable designs listed in Table 13.3 can be placed in useful incomplete block designs for near rotatable and orthogonal central composite designs. The 2^n factorial or 2^{n-p} fractional factorial is placed in one or more incomplete blocks and the axial treatment combinations are placed in one separate block. For the designs listed in Table 13.3 the number of blocks for the 2^n factorial or fractional factorial, and a suggested number of design center points in each block are shown in Table 13.4.

Table 13.4 Incomplete block designs for near rotatable and orthogonal central composite designs

Number of Factors	2	3	4	5	5	6	6
Fraction of 2^n	1	1	1	1	$\frac{1}{2}$	1	$\frac{1}{2}$
N_f	4	8	16	32	16	64	32
m_f	2	2	2	4	2	2	2
*Number of Blocks**	1	2	2	4	1	8	2
α	1.414	1.682	2.000	2.378	2.000	2.828	2.378
N_a	4	6	8	10	10	12	12
m_a	2	2	1	1	4	1	4

*See Appendix 11A for defining contrasts to block the 2^n factorial or fractional factorial design portion.

Reducing the Number of Design Points

The expense, difficulty, or time consumption with certain types of experiments may necessitate reducing the experiment size. The amount of reduction is limited by the statistical model to estimate the response surface. The second-order response surface equation for n factors has a constant term, n linear terms, n quadratic terms, and $n(n-1)/2$ interaction terms for a total of $(n+1)(n+2)/2$ terms. Thus, the minimum number of points a design could have to estimate the second-order response surface is $(n+1)(n+2)/2$.

Designs have been developed to have as close to the minimum number of points as possible to estimate the second-order response surface. Tables of these designs or methods to construct them can be found in Box and Draper (1974), Roquemore (1976), Notz (1982), Draper (1985), Draper and Lin (1990), and Myers and Montgomery (1995).

Most of the designs are based on 2^{n-p} fractional factorials augmented with design points to estimate second-order response surface models. In most cases the designs are saturated with few or no replicated design points. An independent estimate of experimental error is required to test the efficacy of the response surface model, unless the design is replicated. In addition, the saturated designs do not allow a test for lack of fit of the hypothesized second-order response surface model.

An Evaluation of Response Surface Designs

Myers et al. (1992) used the prediction variance of the second-order response surface equation to evaluate many of the popular second-order response surface designs. A design was considered superior if the variance of predicted values was smaller than that of other designs.

The central composite designs were found superior in general over spherical regions covered by the design points (see Figure 13.5). When designs were restricted to the cuboidal regions ($\alpha = 1$ in Figure 13.5), the resulting face-centered cube design formed by the central composite design was found generally superior to the Box–Behnken design in the cuboidal region.

Among the saturated designs, the designs by Roquemore (1976), Notz (1982), and Box and Draper (1974) were found quite efficient relative to others that were evaluated.

Myers and Montgomery (1995) presented design efficiencies for estimating model coefficients and for prediction variances in a spherical region. Their general conclusions were that central composite and Box–Behnken designs were quite efficient as were some of the saturated designs from Roquemore (1976).

13.4 Quadratic Response Surface Estimation

When the supposed region of optimum response has been identified by the method of steepest ascent or other methods of experimentation it is often necessary to characterize the response surface in that region of the factors. Using the designs described in the previous section, experiments can be conducted to obtain data for estimating a quadratic approximation to the response surface.

The estimated response equation will enable the researcher to locate a stationary response point that could be a maximum, a minimum, or a saddle point on the surface. An examination of the contour plot will indicate how sensitive the response variable is to each of the factors and to what degree the factors interplay as they affect the response variable.

Example 13.3 Tool Life Response to Lathe Velocity and Cutting Depth

A new cutting tool available from a vendor was going to be used by a company. The vendor claimed the new model tool would reduce production costs because it would last longer than the old model; thus, tool replacement cost would be reduced. The life of a metal cutting tool is dependent on several operating conditions, including the speed of the lathe and the depth of the cut made by the tool.

The plant engineer had determined from previous studies that maximum tool life was achieved for the current tool with a lathe velocity setting of 400 and a cutting depth setting of 0.075. The engineer wanted to determine the optimum settings required for the new tool. A central composite design was used for an experiment to characterize the life of the new tool under varying lathe speeds and cutting depths within the region of current optimum operating conditions for maximum tool life. The data from the experiment are shown in Table 13.5.

Table 13.5 Observed tool life from a factorial experiment with lathe speed and cutting depth treatment factors in a central composite design

Original Factors		*Coded Factors*		
Lathe Speed	*Cutting Depth*	x_1	x_2	*Tool Life*
600	0.100	+ 1	+ 1	154
600	0.050	+ 1	− 1	132
200	0.100	− 1	+ 1	166
200	0.050	− 1	− 1	83
683	0.075	$\sqrt{2}$	0	156
117	0.075	$-\sqrt{2}$	0	144
400	0.110	0	$\sqrt{2}$	166
400	0.040	0	$-\sqrt{2}$	91
400	0.075	0	0	167
400	0.075	0	0	175
400	0.075	0	0	170
400	0.075	0	0	176
400	0.075	0	0	156
400	0.075	0	0	170

The Estimated Response Surface Equation

The second-order response surface model of Equation (13.2) is fit to the data by least squares regression procedures. The equation can be estimated by any computer program for regression analysis. A brief account of least squares estimation for regression models is given in Appendix 13A.1. A detailed presentation of regression analysis can be found in Rawlings (1988). The estimated second-order response surface equation for tool life from the data in Table 13.5 is

$$\hat{y} = 169 + 6.747x_1 + 26.385x_2 - 10.875x_1^2 - 21.625x_2^2 - 15.250x_1x_2$$

Sum of Squares Partitions for the Regression Analysis

The sum of squares partitions in the analysis of variance for the regression model are shown in Table 13.6. The sum of squares for the full second-order model is

$$SSR(x_1, x_2, x_1^2, x_2^2, x_1x_2) = 10{,}946$$

The regression sum of squares is partitioned into reductions for linear and quadratic components of the model using the principle of reduced model and full model sum of squares partitions.

The partition for the linear components of the model, x_1 and x_2, or

$$SSR(x_1, x_2) = 5{,}933$$

Table 13.6 Analysis of variance for quadratic response surface model

Source of Variation	*Degrees of Freedom*		*Sum of Squares*	*Mean Square*
Total	13		11,317	
Regression	5		10,946	2,189.2
Linear (x_1, x_2)		2	5,933	2,966.5
Quadratic (x_1^2, x_2^2, x_1x_2)		3	5,013	1,671.0
Error	8		371	46.4
Lack of Fit		3	111	37.0
Pure Error		5	260	52.0

is the regression sum of squares for the reduced first-order model $y = \beta_o + \beta_1 x_1 + \beta_2 x_2 + e$. The partition for the quadratic components is the difference between the regression sum of squares for the full model and the reduced model, or

$$SSR(x_1^2, x_2^2, x_1x_2 \mid x_1, x_2) = 10{,}946 - 5{,}933 = 5{,}013$$

The sum of squares for error, $SSE = 371$, is partitioned into two parts. The sum of squares for pure experimental error, $SSE(\text{pure error}) = 260$, with 5 degrees of freedom is computed from the six replicate observations at the center of the design with factor coordinates $(V, D) = (400, 0.075)$. The remaining 3 degrees of freedom sum of squares for error, $SSE(\text{Lack of fit}) = 111$, can be attributed to error in specification of the response surface model, referred to as lack of fit, or it can be attributed to experimental error. Since the six center points of the design provide an estimate of pure experimental error, the sum of squares designated as lack of fit can be used to test the significance of lack of fit to the quadratic model.

Tests of Hypotheses About the Second-Order Model

The hypotheses of interest in the analysis are

- Significance of the complete second-order model:

$$H_0\colon\ \beta_1 = \beta_2 = \beta_{11} = \beta_{22} = \beta_{12} = 0$$

$$F_0 = \frac{2{,}189.2}{52} = 42.1 \qquad \text{Reject } H_0 \text{ since } F_0 > F_{.05,5,5} = 5.05$$

- Significance of linear components for the model:

$$H_0\colon\ \beta_1 = \beta_2 = 0$$

$$F_0 = \frac{2{,}966.5}{52} = 57.0 \qquad \text{Reject } H_0 \text{ since } F_0 > F_{.05,2,5} = 5.79$$

- Significance of quadratic deviations from the linear model:

$$H_0\colon\ \beta_{11} = \beta_{22} = \beta_{12} = 0$$

$$F_0 = \frac{1{,}671}{52} = 32.1 \qquad \text{Reject } H_0 \text{ since } F_0 > F_{.05,3,5} = 5.41$$

- Significance of lack of fit to the quadratic model:

$$F_0 = \frac{37}{52} = 0.71 \qquad \text{Do not reject } H_0 \text{ since } F_0 < F_{.05,3,5} = 5.41$$

The complete quadratic regression model is significant, and the lack of fit to the quadratic model is not significant; thus, we can conclude the second-order model is an adequate approximation to the true response surface. A contour plot of the quadratic response surface model depicted in Figure 13.6 shows a maximum surface with a maximum tool life in the middle of the center contour.

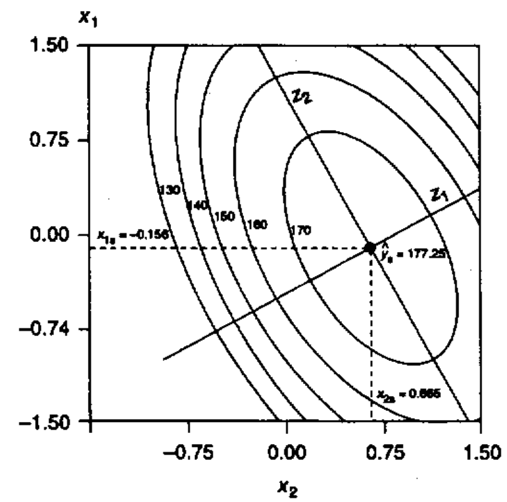

Figure 13.6 Response surface contour plot for the tool life experiment response equation $\hat{y} = 169 + 6.747x_1 + 26.385x_2 - 10.875x_1^2 - 21.625x_2^2 - 15.250x_1x_2$

The coordinates of the contour plot are displayed for the coded values of the two factors. The orientation of the contours indicates some interaction between lathe speed x_1 and cutting depth x_2. For example, a constant cutting tool life of 150 can be maintained for faster lathe speeds, increasing x_1, by decreasing the cutting depth, decreasing x_2.

The contours also indicate the relative sensitivity of tool life to the coded factor levels x_1 and x_2. The tool life contours increase more rapidly toward the maximum on the coded cutting depth axis x_2 than they do on the coded lathe speed axis x_1.

13.5 Response Surface Exploration

The significant quadratic equation and the contour plot of the equation have given us a general picture of the relationship between tool life and the two design factors, lathe speed and cutting depth.

Estimates of the coordinates for the stationary point on the surface and an estimate of the response at the stationary point provide a more specific characterization of the response surface. Sometimes, it is useful to know the direction and amount of change to make in one or more of the design factor levels to achieve the maximum change in response.

A more specific characterization of the sensitivity of response to the design factors can be achieved with the canonical form of the response equation. The location of coordinates for the stationary point and derivation of the canonical form of the response equation requires some knowledge of calculus and matrix algebra. However, the results of the computations are understandable when they are displayed in graphic form on the contour plot of Figure 13.6.

Locating Coordinates for the Stationary Point of the Response Surface

The x_1 and x_2 coordinates for the stationary point are obtained from the partial derivatives of the estimated response function with respect to x_1 and x_2. The estimated response for tool life is

$$\widehat{y} = 169 + 6.747x_1 + 26.385x_2 - 10.875x_1^2 - 21.625x_2^2 - 15.250x_1x_2 \quad (13.8)$$

The partial derivatives are set equal to 0:

$$\frac{\partial \widehat{y}}{\partial x_1} = 0 \qquad \frac{\partial \widehat{y}}{\partial x_2} = 0$$

to produce the equations

$$2(-10.875)x_1 + (-15.250)x_2 = -6.747$$

$$(-15.250)x_1 + 2(-21.625)x_2 = -26.385$$

The solutions of the equations for x_1 and x_2 are $\widehat{x}_{1s} = -0.156$ and $\widehat{x}_{2s} = 0.665$. These values are the coordinates for the maximum response on the surface at the stationary point indicated on Figure 13.6.

The estimated response at the stationary point is found by substituting $\widehat{x}_{1s} = -0.156$ and $\widehat{x}_{2s} = 0.665$ into Equation (13.8); the estimated stationary point response is

$$\widehat{y}_s = 169 + 6.747(-0.156) + 26.385(0.665) - 10.875(-0.156)^2$$

$$- 21.625(0.665)^2 - 15.250(-0.156)(0.665) = 177.25$$

Given $x_1 = (V - 400)/200$ and $x_2 = (D - 0.075)/0.025$, the values of lathe speed (V) and cutting depth (D) at the stationary point are

$$V = -0.156(200) + 400 = 368.8$$

and

$$D = 0.665(0.025) + 0.075 = 0.092$$

The general solution for a stationary point with any number of x_i variables in the response equation is given in Appendix 13A.2.

The Canonical Analysis to Simplify the Quadratic Equation

The canonical form of a quadratic equation is an effective aid to visualize the surface and to determine the relative sensitivity of the response variable to each of the factors. It is difficult to visualize the surface by examining the estimated coefficients for the normal form of the quadratic response equation. Likewise, it is difficult to determine the changes in factor levels necessary to produce a specified change in the response.

The canonical analysis rotates the axes of the x_i variables to a new coordinate system, and the center of the new coordinate system is placed at the stationary response point of the surface. The canonical form of the equation for two variables is

$$\widehat{y} = \widehat{y}_s + \lambda_1 Z_1^2 + \lambda_2 Z_2^2 \quad (13.9)$$

where Z_1 and Z_2 are the rotated axes' variables. Notice only the quadratic terms of the canonical variables Z_1 and Z_2 are included in the canonical form of the response equation. An outline of the computations required to obtain the canonical form for the tool life response equation is given in Appendix 13A.3.

The canonical form for the tool life response equation is

$$\widehat{y} = 177.25 - 25.58Z_1^2 - 6.92Z_2^2 \quad (13.10)$$

where the center of the new coordinate system is located at $x_1 = -0.156$ and $x_2 = 0.665$ in the original coordinate system shown in Figure 13.6. The relationship between the two coordinate systems was determined (Appendix 13A.3) to be

$$Z_1 = 0.4603x_1 + 0.8877x_2 - 0.5185$$
$$Z_2 = 0.8877x_1 - 0.4603x_2 + 0.4446 \quad \textbf{(13.11)}$$

Notice the Z_1 and Z_2 canonical axes are oriented with the contours of the surface. The sizes and signs of the λ_i indicate the type of quadratic response surface that has been estimated.

The λ_i coefficients for the tool life surface are $\lambda_1 = -25.58$ and $\lambda_2 = -6.92$. Examination of the surface in Figure 13.6 reveals that any movement away from the center of the Z_1, Z_2 coordinate system results in a response decrease. Thus, when all λ_i coefficients are negative the surface is a maximum surface such as that for the tool life surface in Figure 13.6.

If the λ_i coefficients are positive, then any movement from the center of the Z_1, Z_2 coordinate system results in a response increase and the surface is a minimum surface as shown in Figure 13.2a. If one coefficient is positive and the other negative, say $\lambda_1 > 0$ and $\lambda_2 < 0$, then movement away from (0, 0) along the Z_1 axis results in an increased response and movement along the Z_2 axis results in a decrease. Thus, the surface is a saddle or minimax at the stationary point as shown in Figure 13.2d. If one of the $\lambda_i = 0$, the surface is a stationary ridge (Figure 13.2c) because the response will not change along the Z_i axis.

The lengths of the principal axes of the ellipses formed by the contours are proportional to $|\lambda_i|^{-1/2}$. For the tool life surface $|-25.58|^{-1/2} = 0.20$ and $|-6.92|^{-1/2} = 0.38$, and the fitted surface is attenuated along the Z_2 axis as seen in Figure 13.6.

Suppose, for illustration, that lathe speed and cutting depth for maximum tool life at coordinates $x_1 = -0.156$ and $x_2 = 0.665$ were impractical. The least change in tool life as lathe speed and cutting depth change is exhibited on the surface along the Z_2 axis direction when $Z_1 = 0$. The x_1 and x_2 coordinates along the Z_2 axis when $Z_1 = 0$ can be obtained from the first equation in Equations (13.11). The least loss in tool life can be found on settings corresponding to values of x_1 and x_2 that satisfy $0.4603x_1 + 0.8877x_2 - 0.5185 = 0$.

The coefficients of the x_i in Equations (13.11) can provide information about the relationships of lathe speed and cutting depth to tool life. Consider the coefficients for the second equation relating Z_2 to x_1 and x_2, $Z_2 = 0.8877x_1 - 0.4603x_2 + 0.4446$. The pair of coefficients (0.8877, −0.4603) indicate a compensation between lathe speed and cutting depth on tool life. An increase in lathe speed, to some extent, can be compensated for by a decrease in cutting depth along the elongated Z_2 axis.

The estimated response equation in its original form or in the canonical form is only valid for the region of factor levels included in the experiment. Any attempt to estimate the tool life outside of the limits bounded by lathe speeds of 117 and 683 and cutting depths of 0.04 and 0.11 could be quite misleading. An entirely different

response model may be necessary to describe tool life outside of the region used by the current study.

13.6 Designs for Mixtures of Ingredients

Some treatment designs involve two or more factors that are ingredients of a mixture in which the percentages of the ingredients must sum to 100% of the mixture. Therefore, the levels of one factor are not independent of other factor levels.

Many food products, construction materials, and other commercial products are formed from mixtures of two or more ingredients in a recipe. Some examples are

- fabrics with a blend of cotton and polyester fiber
- fruit juice blends of orange, pineapple, and apple juices with water
- concrete formed from water, aggregate, and cement
- fertilizer formulations of nitrogen, phosphorus, and potassium

This section includes a brief introduction to selecting designs and estimating response surface equations for mixture experiments. Cornell (1990) provides an in-depth coverage of design and analysis of mixture experiments.

Factor Levels Are Proportions of Ingredients

Variation in the proportions of the ingredients in mixtures can affect the properties of the end product. Investigations with mixture experiments concentrate on the relationship of the measured response variable to the relative proportions of the separate ingredients present in the product rather than on the total amounts of the factors.

If $x_1, x_2, \ldots, x_k$ are the variables representing the proportions of the k ingredients or components of the mixture, the values of the x_i are constrained such that

$$0 \le x_i \le 1 \quad i = 1, 2, \ldots, k \quad \textbf{(13.12)}$$

and the proportions of the k ingredients in the mixture sum to unity, or

$$\sum_{i=1}^{k} x_i = x_1 + x_2 + \cdots + x_k = 1 \quad \textbf{(13.13)}$$

If the proportion of one ingredient is $x_i = 1$, then the other ingredients are absent from the mixture and the product is a pure or single-component mixture. For example, a two-component mixture experiment with cotton and polyester fabric blends represented by the proportions x_1 and x_2 may have pure cotton fabric, in which case $x_1 = 1$ and $x_2 = 0$, or a pure polyester fabric, where $x_1 = 0$ and

$x_2 = 1$. The allowable values of x_1 and x_2 for a two-component mixture design are coordinate values along the line $x_1 + x_2 = 1$ (shown in Figure 13.7).

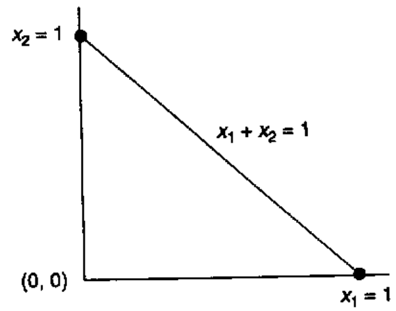

Figure 13.7 Factor space for a two-component mixture, $x_1 + x_2 = 1$

Depict the Factor Space with a Simplex Coordinate System

The coordinate values for a three-component mixture design are the coordinate values found on the plane defined by $x_1 + x_2 + x_3 = 1$ in Figure 13.8a. The geometric description of the factor space for k components is that of a simplex in $(k-1)$ dimensions. The two-dimensional simplex coordinate system for the three-component mixture design is shown in Figure 13.8b as an equilateral triangle.

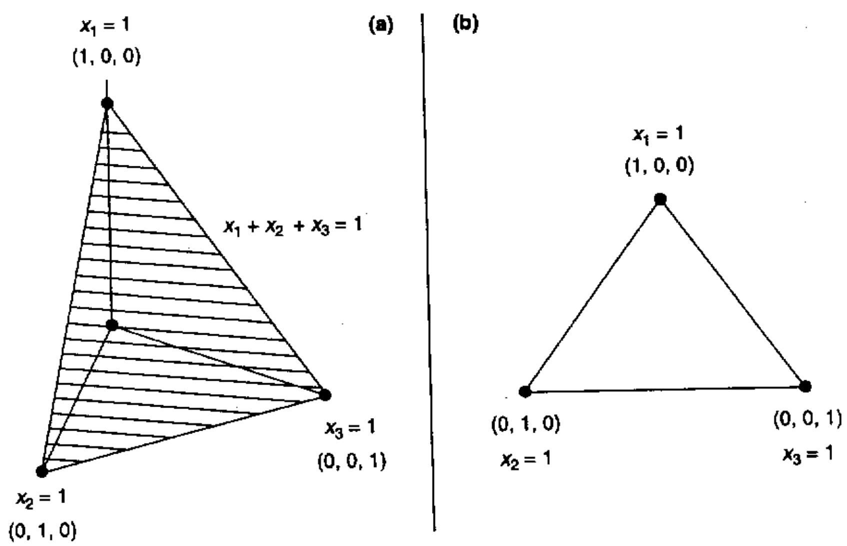

Figure 13.8 Factor space for (a) a three-component mixture, $x_1 + x_2 + x_3 = 1$ and (b) a three-component simplex coordinate system

The vertices of the triangle represent single-component mixtures with one $x_i = 1$ and all others equal to 0. The sides of the triangle represent design coordinates for two-component mixtures with one $x_i = 0$. Design coordinates in the triangle interior represent three-component mixtures with $x_1 > 0$, $x_2 > 0$, and $x_3 > 0$. Any combination of component proportions for a mixture experiment must be on the boundaries or inside the triangle of coordinates in Figure 13.8b.

The axes of the x_i variables in the three-variable simplex coordinate system are shown in Figure 13.9. The axis for component i is the line from the base point $x_i = 0$ and $x_j = 1/(k-1)$ for all other components $j \neq i$ to the vertex, where $x_i = 1$ and $x_j = 0$ for $j \neq i$. For example, with the three-component design in Figure 13.9, the x_1 axis extends from the base coordinate $(0, \frac{1}{2}, \frac{1}{2})$ to the vertex coordinate $(1, 0, 0)$.

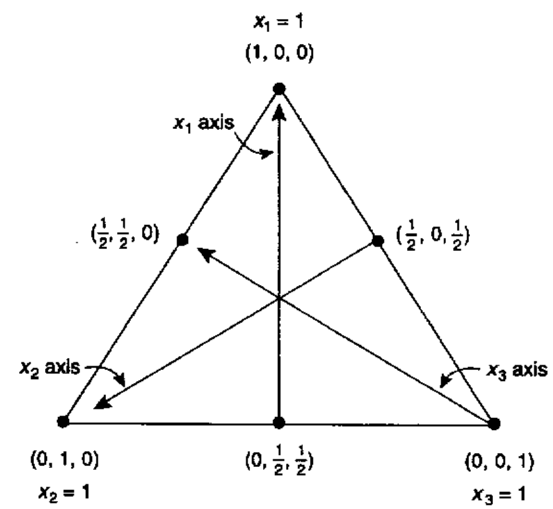

Figure 13.9 Simplex coordinate axes for a three-component mixture with design coordinates for a $\{3, 2\}$ simplex-lattice design

Treatment Designs for Mixtures

Simplex-Lattice Designs

The array made up of a uniform distribution of design coordinates on the simplex coordinate system is known as a *lattice*; see Figure 13.9. The simplex-lattice designs consist of a lattice of design coordinates constructed to enable the estimation of polynomial response surface equations.

The designation $\{k, m\}$ is used for a simplex-lattice design with k components to estimate a polynomial response surface equation of degree m. For example, a

$\{3,2\}$ simplex-lattice design has three components in the mixture design to estimate a quadratic response surface equation.

The proportions of each component included in a $\{k, m\}$ simplex-lattice design are

$$x_i = 0, \frac{1}{m}, \frac{2}{m}, \cdots, 1 \tag{13.14}$$

The design consists of all possible combinations of those levels of the x_i, where $\Sigma\, x_i = 1$ for any combination of proportions.

The combinations of mixture proportions shown at each • in Figure 13.9 are the coordinate values for a $\{3,2\}$ simplex-lattice. The proportions for each x_i with $m = 2$ are $x_i = 0$, $\frac{1}{2}$, and 1. It can be seen in Figure 13.9 that $\Sigma\, x_i = 1$ for each design point.

The $\{k, 2\}$ simplex-lattice design to estimate quadratic response surface equations only has mixtures on the boundaries of the coordinate system with one or more of the components absent from the mixture. The general $\{k, m\}$ simplex-lattice will consist of single-component mixtures, two-component mixtures, and so forth up to mixtures consisting of at most m components. If $m = k$, there will be one mixture at the centroid of the coordinate system in the experiment that contains all mixture components. For example, the $\{3,3\}$ simplex-lattice would include the mixture with component proportions $(\frac{1}{3}, \frac{1}{3}, \frac{1}{3})$ as well as the single-component mixtures and the two-component mixtures with proportions $\frac{1}{3}$ and $\frac{2}{3}$ for the two components.

Simplex-Centroid Designs

The simplex-centroid design is a design on the simplex coordinate system that consists of mixtures containing $1, 2, 3, \ldots,$ or k components, each in equal proportions. Consequently, there are k single-component mixtures, all possible two-component mixtures with proportion $\frac{1}{2}$ for each component, all possible three-component mixtures with proportion $\frac{1}{3}$ for each component, and so forth up to one k-component mixture with proportion $\frac{1}{k}$ for each component. The mixtures for the simplex-centroid design are contrasted with the mixtures for the $\{3,2\}$ and $\{3,3\}$ simplex-lattice designs in Table 13.7.

Augmented Simplex-Centroid Designs

The mixture combinations for the simplex-lattice and simplex-centroid designs lie on the edges of the simplex factor space with the exception of one centroid point that contains all mixture components. More complete mixtures are possible by augmenting the simplex-centroid design with mixtures on the axes of the simplex factor space.

The design points are positioned on each axis equidistant from the centroid toward the vertices. A k-component design will have k additional design points with coordinates

x_1	x_2	$\cdots$	x_k
$\frac{(k+1)}{2k}$	$\frac{1}{2k}$	$\cdots$	$\frac{1}{2k}$
$\frac{1}{2k}$	$\frac{(k+1)}{2k}$	$\cdots$	$\frac{1}{2k}$
$\vdots$	$\vdots$	$\vdots$	$\vdots$
$\frac{1}{2k}$	$\frac{1}{2k}$	$\cdots$	$\frac{(k+1)}{2k}$

The addition of the axial points will provide a better distribution of information throughout the experimental region. The three additional design points required by augmenting the simplex-centroid design for three components are $(\frac{4}{6}, \frac{1}{6}, \frac{1}{6})$, $(\frac{1}{6}, \frac{4}{6}, \frac{1}{6})$, and $(\frac{1}{6}, \frac{1}{6}, \frac{4}{6})$. The complete design is depicted in Figure 13.10.

Table 13.7 Simplex-lattice designs and a simplex-centroid design for a three-component mixture

$\{3,2\}$ *Lattice*			$\{3,3\}$ *Lattice*			*Centroid*		
x_1	x_2	x_3	x_1	x_2	x_3	x_1	x_2	x_3
1	0	0	1	0	0	1	0	0
0	1	0	0	1	0	0	1	0
0	0	1	0	0	1	0	0	1
$\frac{1}{2}$	$\frac{1}{2}$	0	$\frac{2}{3}$	$\frac{1}{3}$	0	$\frac{1}{2}$	$\frac{1}{2}$	0
$\frac{1}{2}$	0	$\frac{1}{2}$	$\frac{2}{3}$	0	$\frac{1}{3}$	$\frac{1}{2}$	0	$\frac{1}{2}$
0	$\frac{1}{2}$	$\frac{1}{2}$	0	$\frac{2}{3}$	$\frac{1}{3}$	0	$\frac{1}{2}$	$\frac{1}{2}$
			$\frac{1}{3}$	$\frac{2}{3}$	0	$\frac{1}{3}$	$\frac{1}{3}$	$\frac{1}{3}$
			$\frac{1}{3}$	0	$\frac{2}{3}$			
			0	$\frac{1}{3}$	$\frac{2}{3}$			
			$\frac{1}{3}$	$\frac{1}{3}$	$\frac{1}{3}$			

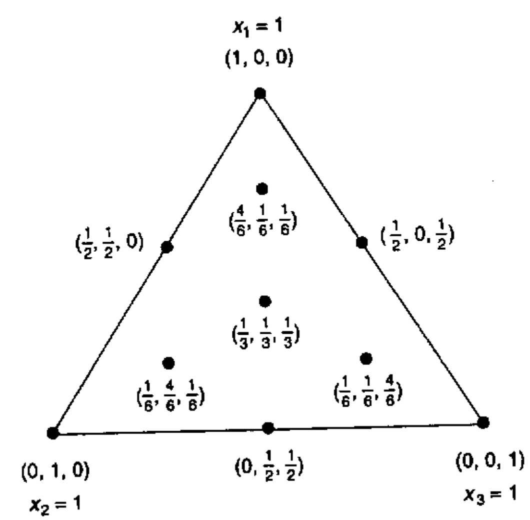

Figure 13.10 Augmented simplex-centroid design for a three-component mixture experiment

Pseudocomponents for Ingredients with Lower Bounds

Many mixtures require all components to be present in at least some minimum proportions. Clearly, concrete requires some minimum proportions of water, cement, and aggregate. Lower bounds, L_i, on component proportions impose the constraint

$$0 \le L_i \le x_i \le 1 \tag{13.15}$$

on the component proportions. Suppose the lower bounds for cement (x_1), water (x_2), and aggregate (x_3) are

$$0.10 \le x_1 \qquad 0.20 \le x_2 \qquad 0.30 \le x_3$$

and a {3,2} simplex-lattice design is going to be used for the experiment. The lower bounds on the component proportions limit the design to a subregion of the original factor space on the simplex shown in Figure 13.9 or Table 13.7.

To simplify the construction of the design coordinates a set of pseudocomponents are constructed by coding the original component variables to a simplex coordinate system for the pseudocomponent variables $\tilde{x}_i$ with constraints $0 \le \tilde{x}_i \le 1$. If the lower bound for component i is L_i and $L = \Sigma L_i$, then the pseudocomponent $\tilde{x}_i$ is computed as

$$\tilde{x}_i = \frac{x_i - L_i}{1 - L}$$

A design in the original components can be constructed on the basis of coordinates for the pseudocomponents that are set up in a regular simplex with $\Sigma \tilde{x}_i = 1$. The proportions of the original components required for mixtures in the experiment can be derived by the reverse transformation

$$x_i = L_i + \tilde{x}_i(1 - L)$$

For the concrete example the lower bounds were $L_1 = 0.10$, $L_2 = 0.20$, and $L_3 = 0.30$ with sum $L = 0.10 + 0.20 + 0.30 = 0.60$. The pseudocomponents are

$$\tilde{x}_1 = \frac{x_1 - 0.10}{0.40} \qquad \tilde{x}_2 = \frac{x_2 - 0.20}{0.40} \qquad \tilde{x}_3 = \frac{x_3 - 0.30}{0.40}$$

and the transformations back to original component proportions from the pseudocomponents are

$$x_1 = 0.10 + 0.40\tilde{x}_1 \qquad x_2 = 0.20 + 0.40\tilde{x}_2 \qquad x_3 = 0.30 + 0.40\tilde{x}_3$$

The complete design for the concrete mixture experiment is shown in Table 13.8 with values for the pseudocomponent coordinates and the original components on the subregion of the original simplex.

Table 13.8 Pseudocomponent and original component coordinates in a {3,2} simplex-lattice design for the concrete mixture experiment

Pseudocomponents			*Original Components*		
$\tilde{x}_1$	$\tilde{x}_2$	$\tilde{x}_3$	*Cement*	*Water*	*Aggregate*
1	0	0	0.50	0.20	0.30
0	1	0	0.10	0.60	0.30
0	0	1	0.10	0.20	0.70
$\frac{1}{2}$	$\frac{1}{2}$	0	0.30	0.40	0.30
$\frac{1}{2}$	0	$\frac{1}{2}$	0.30	0.20	0.50
0	$\frac{1}{2}$	$\frac{1}{2}$	0.10	0.40	0.50

13.7 Analysis of Mixture Experiments

Canonical Polynomials to Approximate Surfaces

The general form of the polynomial function used to approximate linear response surfaces is

$$\mu_y = \beta_0 + \beta_1 x_1 + \cdots + \beta_k x_k \tag{13.16}$$

The restriction on mixture components, $x_1 + x_2 + \cdots + x_k = 1$, creates a dependency among the x_i in the linear function. Multiplying β_0 by $(x_1 + x_2 + \cdots + x_k) = 1$ provides a reexpression of the model as

$$\mu_y = \beta_0 \left(\sum_{i=1}^{k} x_i \right) + \sum_{i=1}^{k} \beta_i x_i = \beta_1^* x_1 + \beta_2^* x_2 + \cdots + \beta_k^* x_k \qquad (13.17)$$

where $\beta_i^* = \beta_0 + \beta_i$, $i = 1, 2, \ldots, k$. The reexpressed equation with parameters β_i^* is known as a **canonical polynomial**. The canonical polynomial and the original polynomial are equivalent because one is derived from the other and the degree of the polynomial and the number of components are unchanged upon reexpression.

The quadratic polynomial function used to approximate response surfaces is

$$\mu_y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i} \sum_{<\, j} \beta_{ij} x_i x_j \qquad (13.18)$$

The quadratic canonical polynomial produced by enacting the restriction $\Sigma x_i = 1$ is

$$\mu_y = \sum_{i=1}^{k} \beta_i^* x_i + \sum_{i} \sum_{<\, j} \beta_{ij}^* x_i x_j \qquad (13.19)$$

where $\beta_i^* = \beta_0 + \beta_i + \beta_{ii}$ and $\beta_{ij}^* = \beta_{ij} - \beta_{ii} - \beta_{jj}$. The new parameters of the quadratic canonical polynomial for three-mixture components expressed in terms of the original polynomial parameters are

$$\beta_1^* = \beta_0 + \beta_1 + \beta_{11} \qquad \beta_2^* = \beta_0 + \beta_2 + \beta_{22} \qquad \beta_3^* = \beta_0 + \beta_3 + \beta_{33}$$

$$\beta_{12}^* = \beta_{12} - \beta_{11} - \beta_{22} \qquad \beta_{13}^* = \beta_{13} - \beta_{11} - \beta_{33} \qquad \beta_{23}^* = \beta_{23} - \beta_{22} - \beta_{33}$$

The interpretation of the canonical polynomials is illustrated with a mixture experiment on gasoline component blends.

Example 13.4 A Gasoline-Blending Mixture Experiment

The octane of a gasoline blend depends upon the proportions of the various petroleum components blended to produce the fuel. The objective of most gasoline-blending studies is to develop a linear blending model to determine the most profitable blend of gasoline components. Coefficients in the linear blending model, referred to as *blending values*, describe the blending behavior of a given fuel component. However, the blending composition depends on a number of factors including the quality of the components. Thus, the linearity of the blending components is lost, and a more complex quadratic or blending component interaction model must be considered.

The analysis of a mixture experiment is illustrated with a gasoline-blending mixture experiment. To evaluate the need for an interaction blending model the experiment was designed to estimate the quadratic canonical polynomial.

A mixture experiment was set up to evaluate the effect of three components on the octane ratings of gasoline. The components alkylate (A), light straight run (B), and reformate (C), were used in a simplex-centroid design with seven mixtures. The octane ratings were determined for two replicate runs of each mixture. The octane ratings for each of the mixtures are shown in Table 13.9.

Table 13.9 Octane ratings from a mixture experiment on gasoline blends

Components*				
x_1	x_2	x_3	y_{ij}	$\bar{y}_{i.}$
1	0	0	106.6, 105.0	105.80
0	1	0	83.3, 81.4	82.35
0	0	1	99.4, 91.4	95.40
$\frac{1}{2}$	$\frac{1}{2}$	0	94.1, 91.4	92.75
$\frac{1}{2}$	0	$\frac{1}{2}$	101.9, 98.0	99.95
0	$\frac{1}{2}$	$\frac{1}{2}$	92.3, 86.5	89.40
$\frac{1}{3}$	$\frac{1}{3}$	$\frac{1}{3}$	96.3, 91.7	94.00

*x_1 = alkylate, x_2 = light straight run, x_3 = reformate

Source: R. D. Snee (1981), Developing blending models for gasoline and other mixtures. *Technometrics* 23, 119–130.

Estimating the Quadratic Canonical Polynomial Response Surface Model

For convenience the asterisk will be dropped from the coefficients in the canonical polynomial equations. The full quadratic canonical polynomial model for the gasoline-blending experiment is

$$y_{ij} = \beta_1 x_{1j} + \beta_2 x_{2j} + \beta_3 x_{3j} + \beta_{12} x_{1j} x_{2j} + \beta_{13} x_{1j} x_{3j} + \beta_{23} x_{2j} x_{3j} + e_{ij}$$
$$i = 1, 2, \ldots, t \qquad j = 1, 2, \ldots, r \qquad (13.20)$$

where the experimental errors e_{ij} are assumed to be independent and normally distributed with mean 0 and variance σ^2. Also, $t = 7$ mixture treatments and $r = 2$ replications per mixture produce a total of $N = rt = 14$ observations.

The hypothesis of initial interest is whether the response depends on the mixture components according to the quadratic model. When the null hypothesis is true the mean response is adequately described by the reduced model $y_{ij} = \beta_0 + e_{ij}$, where $\beta_1 = \beta_2 = \beta_3 = \beta_0$ and $\beta_{12} = \beta_{13} = \beta_{23} = 0$.

An analysis of variance of the data in Table 13.9 for the mixture treatments in a completely randomized design will provide an estimate of pure experimental error. The sum of squares among the mixtures is

$$SST = r\sum_{i=1}^{t}(\overline{y}_{i.} - \overline{y}_{..})^2 = 669.32$$

with $(t-1) = 6$ degrees of freedom. The sum of squares for experimental error is

$$SSE = \sum_{i=1}^{t}\sum_{j=1}^{r}(y_{ij} - \overline{y}_{i.})^2 = 73.74$$

with $(N-t) = 7$ degrees of freedom. Thus, the mean square for experimental error is $MSE = 10.53$ with 7 degrees of freedom.

The least squares estimates of the parameters for the canonical polynomials require a fit to a regression model without the usual intercept term β_0. Many computer regression programs have the capability to fit the regression model without the intercept term, and they will provide the correct least squares estimates of the parameters for the canonical polynomials.

The estimated full quadratic equation is

$$\widehat{y} = 105.8x_1 + 82.3x_2 + 95.4x_3 - 5.1x_1x_2 - 2.4x_1x_3 + 2.3x_2x_3 \qquad \textbf{(13.21)}$$

with experimental error sum of squares $SSE_f = 73.76$ with $14 - 6 = 8$ degrees of freedom.

Tests of Hypotheses About the Model

A Test for the Complete Model

If the response does not depend on the mixture components, the fully reduced model is $y_{ij} = \beta_0 + e_{ij}$ and the surface has a constant height. The experimental error sum of squares for this reduced model is

$$SSE_r = \sum_{i=1}^{t}\sum_{j=1}^{r}(y_{ij} - \overline{y})^2 = 743.05$$

with $N - 1 = 13$ degrees of freedom.

The sum of squares reduction for the full quadratic response surface model is

$$SSR = SSE_r - SSE_f = 743.05 - 73.76 = 669.29$$

with $13 - 8 = 5$ degrees of freedom. The sum of squares for the quadratic model accounts for 5 of the 6 degrees of freedom for treatments with 1 degree of freedom remaining for lack of fit to the quadratic model. The analysis of variance is summarized in Table 13.10.

The null hypothesis for the quadratic response equation is H_0: $\beta_1 = \beta_2 = \beta_3 = \beta_0$ and $\beta_{12} = \beta_{13} = \beta_{23} = 0$. The test statistic $F_0 = MSR/MSE = 133.86/10.53 = 12.71$ exceeds the critical value of $F_{.05,5,7} = 3.97$, and the null hypothesis is rejected.

Table 13.10 Analysis of variance for mixture experiment with gasoline blends

Source of Variation	*Degrees of Freedom*	*Sum of Squares*	*Mean Square*
Treatments	6	669.32	111.55
Regression	5	669.29	133.86
Lack of fit	1	0.03	0.03
Error	7	73.74	10.53

A Test for the Quadratic Terms

The investigator would want to know whether the full quadratic model is necessary to approximate the response surface or whether the linear surface is adequate to explain the relationship between octane rating and the component mixtures. A test of the null hypothesis H_0: $\beta_{12} = \beta_{13} = \beta_{23} = 0$ will determine whether the quadratic components of the model are necessary. The full model and reduced model principle can be used to determine the significance of the quadratic components. The sums of squares for experimental error are required from the full quadratic model and the reduced linear model $y_{ij} = \beta_1 x_{1j} + \beta_2 x_{2j} + \beta_3 x_{3j} + e_{ij}$. The estimated reduced model linear equation is

$$\widehat{y} = 105.1x_1 + 82.1x_2 + 95.5x_3 \qquad \textbf{(13.22)}$$

with experimental error sum of squares $SSE_{lin} = 77.37$ with $14 - 3 = 11$ degrees of freedom. The sum of squares reduction due to the quadratic terms after the linear terms are fit is

$$SSR(\text{quadratic}) = SSE_{lin} - SSE_f = 77.37 - 73.76 = 3.61$$

with $11 - 8 = 3$ degrees of freedom with $MSR(\text{quadratic}) = 3.61/3 = 1.20$. The test statistic $F_0 = MSR(\text{quadratic})/MSE = 1.20/10.53 = 0.11$ with critical region $F_0 > F_{.05,3,7} = 4.35$ is not significant, and the quadratic terms do not improve the approximation of the model to the mixture response surface. Thus, a linear blending model is adequate for this set of blending components.

A Test for the Linear Terms

Since the quadratic components of the model account for very little of the regression sums of squares it is fairly obvious the linear terms account for most of the sum of squares for the regression model. A formal test of the null hypothesis H_0: $\beta_1 = \beta_2 = \beta_3 = \beta_0$ is derived from the sum of squares reduction for the linear model as

$$SSR(\text{linear}) = SSE_r - SSE_{lin} = 743.05 - 77.37 = 665.68$$

with $13 - 11 = 2$ degrees of freedom with $MSR(\text{linear}) = 665.68/2 = 332.84$. The test statistic is $F_0 = MSR(\text{linear})/MSE = 332.84/10.53 = 31.61$ with critical region $F_0 > F_{.05,2,7} = 4.74$, and the null hypothesis is rejected.

Interpretations for the Estimated Response Equation

The estimated linear canonical polynomial, $\widehat{y} = 105.1x_1 + 82.1x_2 + 95.5x_3$ in Equation (13.22), provides a significant and adequate fit to the mixture response surface. The estimated standard error for each of the $\widehat{\beta}_i$ determined from the regression program is $s_{\widehat{\beta}_i} = 1.89$, and the individual coefficient estimates are significant by the Student t test, $t_0 = \widehat{\beta}_i / s_{\widehat{\beta}_i}$, with 7 degrees of freedom.

The coefficient $\widehat{\beta}_i$ is the estimated response at the vertex of the simplex design representing the mixture with 100% of that component or the single-component mixture. Alternatively, it represents the estimated response at the maximum value for that component. The variables for Example 13.4 were the proportions x_1 = alkylate, x_2 = light straight run, and x_3 = reformate. For example, with 100% alkylate and 0% each of light straight run and reformate the estimated octane rating is $\widehat{y} = 105.1$. Likewise, with 100% light straight run the estimated octane is $\widehat{y} = 82.1$, and with 100% reformate the estimated octane is $\widehat{y} = 95.5$. The estimated linear surface is depicted in Figure 13.11.

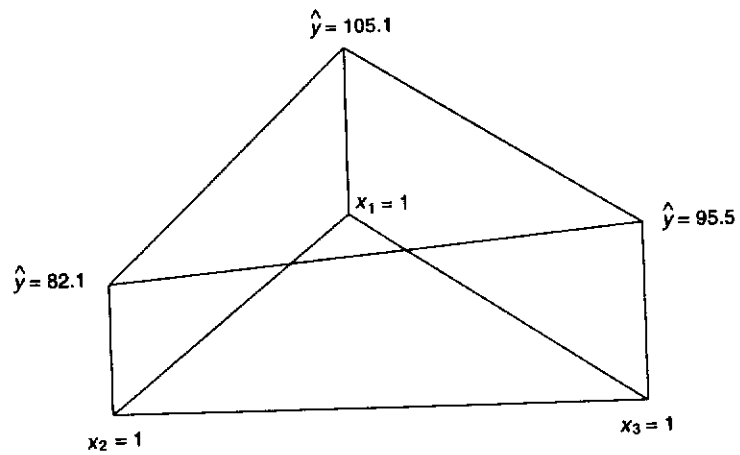

Figure 13.11 Estimated linear surface for the gasoline-blending mixture experiment

The β_{ij} describe the departures from the linear response surface. An illustration of a quadratic response for a two-component system is depicted in Figure 13.12. If the two components are additive, with a linear blending of the two components the mean response is $\mu_l = \beta_1 x_1 + \beta_2 x_2$ (shown as the straight line blending in Figure

(13.12). A nonlinear quadratic blending of the two components with $\beta_{12} > 0$ is shown by the response curve $\mu_q = \beta_1 x_1 + \beta_2 x_2 + \beta_{12} x_1 x_2$. The coefficient β_1 represents the height of the curve when $x_1 = 1$ and $x_2 = 0$, and β_2 represents the height of the curve when $x_1 = 0$ and $x_2 = 1$. The $\beta_{12} x_1 x_2$ term contributes to the response whenever $x_1 > 0$ and $x_2 > 0$. The maximum departure from the linear blending occurs at $x_1 = x_2 = \frac{1}{2}$ when $\beta_{12} x_1 x_2 = \frac{1}{4}\beta_{12}$. The blending of the two components in Figure 13.12 is said to be synergistic because the response for the 1:1 mixture at $x_1 = x_2 = \frac{1}{2}$ exceeds the simple average of the pure mixtures depicted by the linear blending line. If the coefficient β_{12} were negative the nonlinear blending line would fall below that of the linear blending line and the components would be antagonistic to one another.

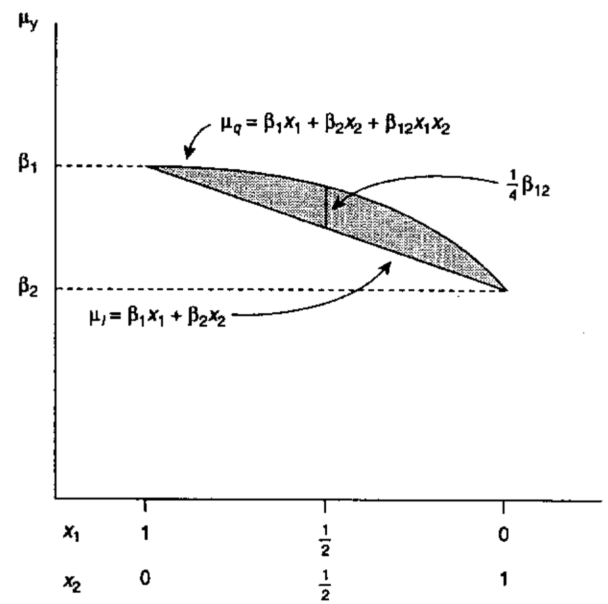

Figure 13.12 Comparison of linear and nonlinear blending for a two-component mixture system

EXERCISES FOR CHAPTER 13

1. A 2^2 factorial experiment was conducted to determine whether the volume of two reagents had an effect on the ability of an assay method to measure levels of a specific drug in serum. Two replications of the treatments were tested in a completely randomized design with two additional

replications at the center of the design. The serum for each test was sampled from a serum pool spiked with a single dose of the drug. The data are shown next with reagent volumes in μl.

Reagent		
A	*B*	*% Drug Recovered*
10	20	32, 35
40	20	44, 47
10	50	51, 53
40	50	68, 72
25	35	48, 53

a. Estimate the experimental error variance.
b. Estimate the linear response equation and standard errors of the coefficient estimates. Are the linear effects of the reagents significant?
c. Estimate β_{12} for AB interaction and standard error of the estimate. Is there significant interaction?
d. Estimate the departure from a linear surface, $\beta_{11} + \beta_{22}$, and standard error of the estimate. Is there a significant departure from a linear surface?
e. Determine the first five steps on the path of steepest ascent from the center of the design with steps of one unit in x_1 for reagent A. Show the levels of both factors at each step.

2. Consider the experiment on vinylation of methyl glucoside in Example 13.1. Suppose the chemist has observed a maximum response at $T = 175$°C and $P = 475.5$ psi via the path of steepest ascent in Table 13.2. Use those levels as the average factor levels in Equation (13.4), and design a central composite rotatable design to estimate the quadratic response surface equation. Show the actual levels of T and P required for each treatment combination in the design.

3. Show the coded design coordinates required for a uniform precision rotatable central composite design with four factors.

4. Describe the response surface for the following canonical forms given for quadratic response surfaces:

a. $\hat{y} = 100 - Z_1^2 - 2Z_2^2$

b. $\hat{y} = 50 + 2Z_1^2$

c. $\hat{y} = 75 + Z_1^2 - 2Z_2^2$

5. An animal scientist studied the relationship between metabolism of methionine, a sulfur amino acid, and carotene, vitamin A, as they affect the growth of chickens. The optimum levels of methionine and carotene were thought to be 0.9% methionine in the diet and 50 micrograms carotene per day. A central composite rotatable design was used for the experiment. Eight chicks were randomly assigned to each of the treatment diets, and their weight gains were recorded after 38 days. The average weight gains for the treatments follow.

Original Factors		*Coded Factors*		
Methionine	*Carotene*	x_1	x_2	*Weight Gain*
1.183	85.36	+1	+1	445
1.183	14.64	+1	−1	331
0.617	85.36	−1	+1	443
0.617	14.64	−1	−1	336
1.183	50.00	$\sqrt{2}$	0	414
0.500	50.00	$-\sqrt{2}$	0	389
0.900	100.00	0	$\sqrt{2}$	435
0.900	0.00	0	$-\sqrt{2}$	225
0.900	50.00	0	0	442
0.900	50.00	0	0	412
0.900	50.00	0	0	418
0.900	50.00	0	0	440
0.900	50.00	0	0	441

a. Estimate the quadratic response surface equation for weight gain, and summarize the sum of squares partitions in an analysis of variance table.
b. Test the significance of the complete quadratic model, the quadratic deviations from the linear model, the significance of the linear components of the model, and the lack of fit to the quadratic model. What are your conclusions?
c. The response surface has a maximum within the design coordinates. Determine the levels of methionine and carotene that produce the maximum response, and estimate the maximum response.
d. Compute the canonical equation (see Appendix 13A.3), and describe the response surface. Based on the canonical equation, what is the relationship between methionine and carotene? Can one be used to compensate for the other in the animal's diet?

6. The experiment on vinylation of methyl glucoside used in Example 13.1 (Marvel et al., 1969) included four factors in a central composite rotatable design placed in an incomplete block design. The percent conversion of methyl glucoside to a vinylation product was the response variable of interest. The actual and coded levels of the four factors used in the experiment were

	Coded Level	−2	−1	0	1	2
x_1	Time, hours	1	3	5	7	9
x_2	Temperature, °C	115	130	145	160	175
x_3	Pressure, psi	250	325	400	475	550
x_4	Solvent ratio (water/dioxane)	95	80	65	50	35

The percent conversion of methyl glucoside to a vinylation product for each treatment combination in the experiment is shown in the table that follows:

Block 1					*Block 2*					*Block 3*				
y	x_1	x_2	x_3	x_4	y	x_1	x_2	x_3	x_4	y	x_1	x_2	x_3	x_4
10	−1	−1	−1	−1	56	−1	1	1	1	13	−2	0	0	0
21	1	1	−1	−1	7	−1	−1	−1	1	43	2	0	0	0
5	−1	−1	1	1	35	1	1	1	−1	2	0	−2	0	0
46	1	1	1	1	27	1	−1	1	1	52	0	2	0	0
6	1	−1	−1	1	19	1	−1	−1	−1	18	0	0	−2	0
16	1	−1	1	−1	52	1	1	−1	1	52	0	0	2	0
22	−1	1	1	−1	17	−1	−1	1	−1	15	0	0	0	−2
18	−1	1	−1	1	24	−1	1	−1	−1	58	0	0	0	2
21	0	0	0	0	33	0	0	0	0	34	0	0	0	0
31	0	0	0	0	30	0	0	0	0	39	0	0	0	0

a. Estimate the quadratic response surface equation for percent conversion, and summarize the sum of squares partitions in an analysis of variance table.
b. Test the significance of the complete quadratic model, the quadratic deviations from the linear model, the significance of the linear components of the model, and the lack of fit to the quadratic model. What are your conclusions?
c. Determine the factor levels that produce the maximum response with the quadratic model. Is the optimum within the current design factor levels?
d. Compute the canonical equation from the quadratic model estimates (see Appendix 13A.3), and describe the response surface. Based on the canonical equation, what type of surface has been estimated?

7. Construct an incomplete block design for a central composite design with three treatment factors such that the conditions in Equations (13.5) and (13.6) are satisfied. What is the value of α required for the relationship to hold in Equation (13.7)?

8. An experiment is planned to evaluate the flavor quality of a fruit juice containing orange, pineapple, lime, and papaya juices. The minimum allowable proportions of the four juices in the mix are orange $> .15$, pineapple $> .10$, lime $> .10$, and papaya $> .20$. Design a mixture experiment with a simplex-lattice design. List the design both as pseudocomponents and as original components.

9. A highway-paving mixture is formed by dispersing liquid sulfur into liquid asphalt to produce a sulfur–asphalt binder. The binder is then mixed with sand to produce the paving mixture. A mixture experiment is to be conducted with a minimum of 5% sulfur and a minimum of 10% asphalt.
   a. Design the mixture experiment with a simplex-centroid design. Show the design as pseudocomponents and as original components.
   b. Construct the design as an augmented simplex-centroid. Show the pseudocomponents and original components.

10. A mixture experiment was conducted with the sulfur–asphalt binder described in Exercise 13.9. The experiment was conducted with a minimum of 10% sulfur, a minimum of 20% asphalt, and a minimum of 50% sand in the mixtures. Two replications of each mixture were prepared, and a specimen from each replication was subjected to a strength test. A simplex-lattice design was used for the experiment. The strength data with mixture proportions for pseudocomponents, where x_1 is the pseudocomponent for sulfur, x_2 for asphalt, and x_3 for sand, follow.

Pseudocomponents			*Strength*
x_1	x_2	x_3	y_{ij}
1	0	0	12.0, 13.7
0	1	0	2.4, 3.6
0	0	1	2.6, 4.3
0.5	0.5	0	18.9, 16.8
0.5	0	0.5	19.4, 17.1
0	0.5	0.5	4.6, 7.3

a. Given the minimum values for sulfur, asphalt, and sand, determine the actual proportions for the three components for each of the treatment mixtures in the experiment.
b. Estimate the experimental error variance.
c. Estimate the linear and quadratic response surface polynomial. Determine the significance of the linear terms and the significance of the quadratic addition to the model.
d. Interpret the coefficients in the model.

13A.1 Appendix: Least Squares Estimation of Regression Models

The least squares estimation of parameters for the regression model follows the procedures illustrated in previous chapters for various experiment designs. The difference to be noted for the regression model is the inclusion of the continuous value x_i variables in the model that were not seen previously in the experiment design models.

Estimation is illustrated for a model with two x_i variables. The multiple linear regression model is

$$y_j = \beta_0 x_{0j} + \beta_1 x_{1j} + \beta_2 x_{2j} + e_j \qquad j = 1, 2, \ldots, n$$

The intercept or constant term for the model written in the general form as $\beta_0 x_{0j}$ is most often identified in the model only as β_0 since the variable x_{0j} takes on a constant value $x_{0j} = 1$ for all observations.

The least squares estimators for the regression coefficients are found by differentiating

$$\sum_{j=1}^{n} e_j^2 = \sum_{j=1}^{n} (y_j - \beta_0 x_{0j} - \beta_1 x_{1j} - \beta_2 x_{2j})^2$$

with respect to each of the β_i and setting the result equal to 0. The partial derivatives set to 0 are

$$\frac{\partial}{\partial \beta_0} = -2\sum_{j=1}^{n} x_{0j}(y_j - \beta_0 x_{0j} - \beta_1 x_{1j} - \beta_2 x_{2j}) = 0$$

$$\frac{\partial}{\partial \beta_1} = -2\sum_{j=1}^{n} x_{1j}(y_j - \beta_0 x_{0j} - \beta_1 x_{1j} - \beta_2 x_{2j}) = 0$$

$$\frac{\partial}{\partial \beta_2} = -2\sum_{j=1}^{n} x_{2j}(y_j - \beta_0 x_{0j} - \beta_1 x_{1j} - \beta_2 x_{2j}) = 0$$

The resulting normal equations are

$$\widehat{\beta}_0 \Sigma x_{0j}^2 + \widehat{\beta}_1 \Sigma x_{0j}x_{1j} + \widehat{\beta}_2 \Sigma x_{0j}x_{2j} = \Sigma x_{0j}y_j$$

$$\widehat{\beta}_0 \Sigma x_{0j}x_{1j} + \widehat{\beta}_1 \Sigma x_{1j}^2 + \widehat{\beta}_2 \Sigma x_{1j}x_{2j} = \Sigma x_{1j}y_j$$

$$\widehat{\beta}_0 \Sigma x_{0j}x_{2j} + \widehat{\beta}_1 \Sigma x_{1j}x_{2j} + \widehat{\beta}_2 \Sigma x_{2j}^2 = \Sigma x_{2j}y_j$$

The simultaneous solutions of equations for the $\widehat{\beta}_i$ result in the least squares estimators for the β_i. The estimates for a given problem can be obtained with any statistical computing package that includes a program for multiple linear regression.

The interested reader can find a detailed account of regression analysis methodology in Rawlings (1988). A brief outline of the model formulation, construction of the normal equations, and solutions to the normal equations are given here assuming some knowledge of matrix notation. The multiple linear regression model can be written in matrix form as

$$\boldsymbol{y} = \boldsymbol{X\beta} + \boldsymbol{e}$$

where

$$\boldsymbol{y} = \begin{bmatrix} y_1 \\ y_2 \\ \vdots \\ y_n \end{bmatrix} \quad \boldsymbol{X} = \begin{bmatrix} 1 & x_{11} & \cdots & x_{k1} \\ 1 & x_{12} & \cdots & x_{k2} \\ \vdots & \vdots & \ddots & \vdots \\ 1 & x_{1n} & \cdots & x_{kn} \end{bmatrix} \quad \boldsymbol{\beta} = \begin{bmatrix} \beta_o \\ \beta_1 \\ \vdots \\ \beta_k \end{bmatrix} \quad \boldsymbol{e} = \begin{bmatrix} e_1 \\ e_2 \\ \vdots \\ e_n \end{bmatrix}$$

Denoting $\boldsymbol{X}'$ as the transpose of the $\boldsymbol{X}$ matrix the normal equations in matrix form are

$$\boldsymbol{X}'\boldsymbol{X}\widehat{\boldsymbol{\beta}} = \boldsymbol{X}'\boldsymbol{y}$$

where

$$\boldsymbol{X}'\boldsymbol{X} = \begin{bmatrix} n & \Sigma x_1 & \Sigma x_2 & \cdots & \Sigma x_k \\ \Sigma x_1 & \Sigma x_1^2 & \Sigma x_1 x_2 & \cdots & \Sigma x_1 x_k \\ \Sigma x_2 & \Sigma x_1 x_2 & \Sigma x_2^2 & \cdots & \Sigma x_2 x_k \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ \Sigma x_k & \Sigma x_1 x_k & \Sigma x_2 x_k & \cdots & \Sigma x_k^2 \end{bmatrix} \qquad \boldsymbol{X}'\boldsymbol{y} = \begin{bmatrix} \Sigma y \\ \Sigma x_1 y \\ \Sigma x_{2y} \\ \vdots \\ \Sigma x_k y \end{bmatrix}$$

The solutions to the normal equations for $\widehat{\boldsymbol{\beta}}$ are found by multiplying both sides of the normal equations by the inverse matrix for $\boldsymbol{X}'\boldsymbol{X}$ denoted $(\boldsymbol{X}'\boldsymbol{X})^{-1}$. The solution is written as

$$\widehat{\boldsymbol{\beta}} = (\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{X}'\boldsymbol{y}$$

The computations are illustrated with data for two independent variables, x_1 and x_2, to estimate the coefficients for the first-order model $y = \beta_o + \beta_1 x_1 + \beta_2 x_2 + e$. The data matrices are

$$\boldsymbol{y}' = [41,\ 52,\ 54,\ 73,\ 66,\ 67,\ 79\ ]$$

$$\boldsymbol{X}' = \begin{bmatrix} 1 & 1 & 1 & 1 & 1 & 1 & 1 \\ 1 & 2 & 3 & 4 & 5 & 6 & 7 \\ 1 & 2 & 1 & 3 & 2 & 2 & 3 \end{bmatrix}$$

$$\boldsymbol{X}'\boldsymbol{X} = \begin{bmatrix} 7 & 28 & 14 \\ 28 & 140 & 63 \\ 14 & 63 & 32 \end{bmatrix} \quad \boldsymbol{X}'\boldsymbol{y} = \begin{bmatrix} 432 \\ 1{,}884 \\ 921 \end{bmatrix}$$

$$(\boldsymbol{X}'\boldsymbol{X})^{-1} = \begin{bmatrix} 1.16 & -0.03 & -0.44 \\ -0.03 & 0.06 & -0.11 \\ -0.44 & -0.11 & 0.44 \end{bmatrix}$$

The solution to the normal equations is

$$\widehat{\boldsymbol{\beta}} = (\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{X}'\boldsymbol{y} = \begin{bmatrix} 1.16 & -0.03 & -0.44 \\ -0.03 & 0.06 & -0.11 \\ -0.44 & -0.11 & 0.44 \end{bmatrix} \begin{bmatrix} 432 \\ 1{,}884 \\ 921 \end{bmatrix} = \begin{bmatrix} 31.43 \\ 3.57 \\ 8.00 \end{bmatrix}$$

The estimated equation is $\widehat{y}_j = \widehat{\beta}_0 x_0 + \widehat{\beta}_1 x_{1j} + \cdots + \widehat{\beta} x_{kj}$, and the sum of squares for experimental error for the full regression model is calculated as

$$SSE_f = \sum_{j=1}^{n} (y_j - \widehat{y}_j)^2 = (\boldsymbol{y} - \boldsymbol{X}\widehat{\boldsymbol{\beta}})'(\boldsymbol{y} - \boldsymbol{X}\widehat{\boldsymbol{\beta}}) = \boldsymbol{y}'\boldsymbol{y} - \widehat{\boldsymbol{\beta}}'\boldsymbol{X}'\boldsymbol{y}$$

$$= \sum_{j=1}^{n} y_j^2 - \widehat{\beta}_o \Sigma y_j - \widehat{\beta}_1 \Sigma x_{1j} y_j - \cdots - \widehat{\beta}_k \Sigma x_{kj} y_j$$

with $n - k - 1$ degrees of freedom.

The reduced model without the variables $x_i, i = 1, 2, \ldots, k$, is $y_j = \beta_0 x_{0j} + e_j = \mu + e_j$. The estimated regression equation with the reduced model is $\widehat{y}_j = \widehat{\beta}_0 x_0$. The sum of squares for experimental error for the reduced model is

$$SSE_r = \sum_{j=1}^{n} (y_j - \widehat{y}_j)^2 = \sum_{j=1}^{n} y_j^2 - \widehat{\beta}_0 \sum_{j=1}^{n} y_j$$

with $n - 1$ degrees of freedom.

For the reduced model $\widehat{\beta}_0 = \overline{y}_.$ and $SSE_r = \sum_{j=1}^{n} (y_j - \overline{y}_.)^2$. The sum of squares for regression due to the inclusion of the independent variables $x_i, i = 1, 2, \ldots, k$, in the model is

$$SSR = SSE_r - SSE_f$$

with k degrees of freedom.

Given $\Sigma y_j^2 = 27{,}716$, the experimental error sum of squares for the full model of the example is

$$SSE_f = 27{,}716 - 31.43(432) - 3.57(1{,}884) - 8.0(921) = 44.4$$

with $n - k - 1 = 7 - 2 - 1 = 4$ degrees of freedom. The estimate of β_0 for the reduced model is $\overline{y}_. = 61.7$, and the experimental error sum of squares is $SSE_r = 1055.4$. The sum of squares for regression is

$$SSR = 1055.4 - 44.4 = 1011.0$$

with $k = 2$ degrees of freedom.

13A.2 Appendix: Location of Coordinates for the Stationary Point

The estimated quadratic model expressed in matrix form is

$$\widehat{y} = \widehat{\beta}_o + \boldsymbol{x}'\boldsymbol{b} + \boldsymbol{x}'\boldsymbol{B}\boldsymbol{x} \tag{13A.1}$$

where

$$\boldsymbol{x}' = [x_1 \quad x_2 \quad \cdots \quad x_k] \qquad \boldsymbol{b}' = [\widehat{\beta}_1 \quad \widehat{\beta}_2 \quad \cdots \quad \widehat{\beta}_k]$$

$$\boldsymbol{B} = \begin{bmatrix} \widehat{\beta}_{11} & \widehat{\beta}_{12}/2 & \widehat{\beta}_{13}/2 & \cdots & \widehat{\beta}_{1k}/2 \\ \widehat{\beta}_{12}/2 & \widehat{\beta}_{22} & \widehat{\beta}_{23}/2 & \cdots & \widehat{\beta}_{2k}/2 \\ \widehat{\beta}_{13}/2 & \widehat{\beta}_{23}/2 & \widehat{\beta}_{33} & \cdots & \widehat{\beta}_{3k}/2 \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ \widehat{\beta}_{1k}/2 & \widehat{\beta}_{2k}/2 & \widehat{\beta}_{3k}/2 & \cdots & \widehat{\beta}_{kk} \end{bmatrix}$$

The stationary point is found by setting the derivative of $\widehat{y}$ with respect to the $\boldsymbol{x}$ vector equal to 0,

$$\frac{\partial \widehat{y}}{\partial \boldsymbol{x}} = \boldsymbol{b} + 2\boldsymbol{B}\boldsymbol{x} = 0 \tag{13A.2}$$

The vector of design coordinates for the stationary point is the solution to Equation (13A.2), or

$$\boldsymbol{x}_s = -\frac{1}{2}\boldsymbol{B}^{-1}\boldsymbol{b} \tag{13A.3}$$

Substituting the solution into Equation (13A.1), the estimated response at the stationary point is

$$\widehat{y}_s = \widehat{\beta}_0 + \frac{1}{2}\boldsymbol{x}_s'\boldsymbol{b} \tag{13A.4}$$

13A.3 Appendix: Canonical Form of the Quadratic Equation

The canonical form of the quadratic response equation is

$$\widehat{y} = \widehat{y}_s + \lambda_1 Z_1^2 + \lambda_2 Z_2^2 + \cdots + \lambda_k Z_k^2 \tag{13A.5}$$

where the λ_i are the eigenvalues of the matrix $\boldsymbol{B}$ in Equation (13A.1) and the Z_i are variables associated with the rotated axes that correspond to the axes of the contours of the response surface. The origin for the rotated coordinate system is the stationary point with all $Z_i = 0$ and response $\widehat{y}_s$.

The eigenvalues of $\boldsymbol{B}$ are the roots of the determinantal equation

$$|\,\boldsymbol{B} - \lambda\boldsymbol{I}\,| = 0 \tag{13A.6}$$

where $\boldsymbol{I}$ is the identity matrix. The relationship between the matrix $\boldsymbol{B}$ and the λ_i is

$$\boldsymbol{B}\boldsymbol{m}_i = \boldsymbol{m}_i\lambda_i \quad i = 1, 2, \ldots, k \tag{13A.7}$$

where the $\boldsymbol{m}_i$ are the eigenvectors corresponding to the λ_i. The $\boldsymbol{m}_i$ are normalized so that $\boldsymbol{m}_i'\boldsymbol{m}_i = 1$.

The relationship between the variables representing the coded factor levels $\boldsymbol{x}$ and the canonical equation variables $\boldsymbol{Z}$ is

$$Z = M'(x - x_s) \tag{13A.8}$$

where the columns of M are the normalized eigenvectors m_i.

The various methods required for the calculations can be found in standard matrix algebra books such as Graybill (1983). The matrix calculations can be performed with many of the common computer programs. The calculations are illustrated with the response equation for Example 13.3,

$$\hat{y} = 169 + 6.747x_1 + 26.385x_2 - 10.875x_1^2 - 21.625x_2^2 - 15.25x_1x_2$$

The determinantal equation is

$$\begin{vmatrix} -10.875 - \lambda & -7.625 \\ -7.625 & -21.625 - \lambda \end{vmatrix} = 0$$

with $\lambda^2 + 32.5\lambda + 177.03 = 0$. The roots of the quadratic equation are $\lambda_1 = -25.58$ and $\lambda_2 = -6.92$. Thus, the canonical equation with $\hat{y}_s = 177.25$ is

$$\hat{y} = 177.25 - 25.58Z_1^2 - 6.92Z_2^2$$

The matrix of normalized eigenvectors is

$$M = \begin{bmatrix} 0.4603 & 0.8877 \\ 0.8877 & -0.4603 \end{bmatrix}$$

The coordinate of the stationary point is $x_{1s} = -0.156$ and $x_{2s} = 0.665$, and the relationship between the canonical variables and the coded factor variables is

$$\begin{bmatrix} Z_1 \\ Z_2 \end{bmatrix} = \begin{bmatrix} 0.4603 & 0.8877 \\ 0.8877 & -0.4603 \end{bmatrix} \begin{bmatrix} (x_1 + 0.156) \\ (x_2 - 0.665) \end{bmatrix}$$

or

$$Z_1 = 0.4603(x_1 + 0.156) + 0.8877(x_2 - 0.665)$$

$$Z_2 = 0.8877(x_1 + 0.156) - 0.4603(x_2 - 0.665)$$

14 Split-Plot Designs

This chapter introduces the split-plot design for experiments with a factorial treatment design and describes some unique features of the design relative to its structure, composition of experimental errors, and analysis. The relative efficiency for split-plot designs is also discussed. Extensions and variations of the design include the split-split-plot and split-block designs.

14.1 Plots of Different Size in the Same Experiment

One factor sometimes requires more experimental material for its evaluation than a second factor in factorial experiments. In agronomic or horticultural field trials a factor such as cultural methods may require the use of equipment that is best-suited for large plots, whereas another factor in the experiment such as cultivar or fertility level may be applied easily to a much smaller plot of land. The larger cultural treatment plot, the *whole plot*, is split into smaller *subplots* to which the different cultivars or fertility treatments are applied. This is known as a *split-plot* design, and in this particular example there are two different sizes of experimental units.

The experiment used for the following example illustrates the creation of a split-plot design when a second factor was introduced to subdivisions of the existing experimental units for an experiment already in progress.

Example 14.1 Nitrogen Fertilizer and Thatch Accumulation in Penncross Creeping Bent Grass

The soil for most golf greens is almost pure sand and frequent irrigation and fertilization are required to maintain the turf. The sandy soil has little capacity to retain nitrogen, and after fertilization the nitrogen quickly leaches from the root zone after irrigation. Administering large initial doses of nitrogen to