

EFFICIENT SCREENING OF PROCESS VARIABLES

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Industrial research aimed at process or product improvement requires consideration of a large number of possible variables. An efficient method for screening such variables to select the more important ones is frequently desired. One such method is the Plackett-Burman design, which is based on balanced incomplete blocks (3).

In this article, the use of this design for screening process variables is illustrated by a case history involving the preparation of a new catalyst. The effects of preparation variables on catalytic activity are studied in a Plackett-Burman design to identify the major variables as candidates for further study.

The increasing complexity of chemical processes and the climbing costs of research work in recent years have had a double effect on research workers. First, workers are often confronted with problems in which many variables may be of possible importance. Second, there is considerable pressure to pick out and optimize the most important variables as efficiently as possible.

The classical method—studying one variable at a time while holding all others constant—is extremely inefficient in many cases. In recent years, the literature has encouraged the use of such statistical methods as regression analysis, factorial designs, and EVOP. However, even these either involve a large number of experiments or are too restricted in the number of variables which can be studied. Thus, if the less important variables happen to be chosen, the study may result in much effort and expense and still not provide much useful information.

There is obviously a need for a practical method of screening large numbers of variables of a process. If efficient preliminary screening could be accomplished, a relatively small number of important variables could be selected for further, more detailed study. The statistical procedures mentioned previously could be used for this further study, with the major effort and expense concentrated on the most important variables.

The design of optimum multifactor experiments has been treated by Plackett and Burman (3). These authors proposed a series of basic designs for as many as

MATRIX FOR

RUN NO.	RANDOM ORDER	A	B	C
1	1	+	+	+
2	4	+	+	+
3	7	+	+	-
4	5	+	-	+
5	16	-	+	-
6	10	+	-	+
7	12	-	+	+
8	15	+	+	+
9	6	+	-	-
10	3	-	-	+
11	2	-	+	-
12	13	+	-	-
13	11	-	-	-
14	9	-	-	+
15	14	-	+	+
16	8	-	-	-

100 experiments. These suggested designs are based on balanced incomplete blocks. Each design of N experiments is useful for studying up to $N - 1$ variables. Although the article seems to be fairly well known among statisticians, there are few references in the literature to practical applications of Plackett-Burman designs. Thus, chemists and engineers do not appear to have recognized the utility of these designs. In addition, the language of the original paper conceals from most nonstatisticians the relative ease with which the analysis is applied to the screening of factors in a multifactor investigation.

Williams (4) compared various screening designs in a

problem in which 24 variables were studied. A Plackett-Burman analysis using 28 experiments was compared with a random-balance design using 28 experiments, and with a fractional factorial using 32 experiments. The Plackett-Burman design was generally best suited for efficient screening and accuracy in picking out the important variables. It seems, therefore, that a detailed example of the use of the Plackett-Burman designs might serve to encourage their increased use in programs for the screening of variables.

Example—Process Description

A new catalyst for a specific use had been discovered. A measure of the quality of the catalyst could be obtained readily in a standardized test by measuring the per cent yield of product in the catalyzed reaction. A number of experiments performed in the classical manner had vaguely outlined the area in which a fairly active

16 RUNS

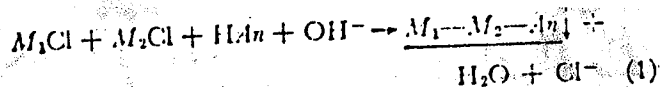
-- = LOW
+ = HIGH

VARIABLE											
D	E	F	(G)	H	I	J	K	L	M	(N)	(O)
+	-	+	-	+	+	-	-	+	-	-	-
-	+	-	+	+	-	-	+	-	-	-	+
+	-	+	+	-	-	+	-	-	-	+	+
-	+	+	-	-	+	-	-	-	+	+	+
+	+	-	-	+	-	-	-	+	+	+	+
+	-	-	+	-	-	-	+	+	+	+	-
-	-	+	-	-	-	+	+	+	+	-	+
-	+	-	-	-	+	+	+	+	-	+	-
+	-	-	-	+	+	+	+	-	+	-	+
-	-	-	+	+	+	+	-	+	-	+	+
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-	+	+	+	+	-	+	-	+	+	-	-
+	+	+	+	-	+	-	+	+	-	-	+
+	+	+	-	+	-	+	+	-	-	+	-
+	+	-	+	-	+	+	-	-	+	-	-
-	-	-	-	-	-	-	-	-	-	-	-

Figure 1. Plackett-Burman matrix for determining the effects of 15 variables at two levels using 16 runs. In the example, catalyst activity was studied using this experimental design. Twelve variables represented real changes in preparation methods, while three were dummy variables used to estimate experimental error.

catalyst could be prepared. However, attempts to optimize the catalyst activity on the basis of these preliminary results indicated that the system was complex, and that many variables might affect catalyst activity.

The catalyst was prepared in the form of an insoluble mixed metal salt, precipitated by neutralizing an acid solution of the metal chlorides with a source of hydroxyl ions.



where M_1Cl = first metal chloride, M_2Cl = second metal chloride, and An = anion.

The soluble chlorides of the two metals, along with the acid, were made into an acid feed solution which was fed into a stirred vessel simultaneously with an ammonium hydroxide solution. The feed rates were such that the pH of the system remained constant within narrow limits. The resulting precipitate was filtered, washed, dried, and treated with additives to produce a form suitable for testing as a catalyst.

Choice of Experimental Design

In a system such as this, many variables which might affect catalyst activity are obvious. These include the temperature of precipitation, the pH, the ratio of one metal to the other, and the amount of excess anion. To attack this problem in the classical manner—that is, by changing one variable at a time while holding the rest constant—would require large expenditures of time and effort. Ideally, effort should be concentrated on studying only the most important variables affecting the catalyst activity.

A four-variable factorial design at two levels requires 16 separate experiments. Five variables at two levels can be studied in 32 experiments, or with 16 experiments using a one-half replicate. Because more than four or five variables may be involved in our example, neither of these factorial designs is satisfactory for screening.

The original Plackett-Burman paper suggests basic designs for 8, 12, 16, 20, up to 100 experiments. In each of these, $N - 1$ factors can be studied. In general, one determines how many variables need be included, and then chooses the design which most nearly satisfies that number. Thus, the design for eight experiments is suitable for up to seven factors. Any factors not assigned can be listed as dummies. As will be shown in the example, such dummy runs can be used to obtain an estimate of the variance.

A further consideration in the choice of the size of the Plackett-Burman design is the possibility of identifying the confounding which exists between the effects of primary variables and the effects of two-factor, three-factor, and higher order interactions. Although the effects of such interactions cannot be separated using this kind of experimental design, identification of the confounding may aid the chemist in interpreting the results. If one prefers to deal with a design in which a table of the confounding of two-factor interactions can be constructed, as shown below, it is necessary to choose a design in which N is an integral power of two. For these reasons, the design chosen for our example was one requiring 16 experiments or runs. The 15 variables which could be studied were sufficient for preliminary screening, while 16 runs appeared to be a convenient number for laboratory precipitations. Incidentally, a complete factorial design for 15 variables at two levels would require 32,768 experiments.

Variables Chosen for Study

- A** = pH of precipitation
B = ratio of M_1 to M_2 in the acid feed solution
C = precipitation temperature
D = excess anion
E = acid solution feed rate
F = source of metal ion M_2 , indicated by sources 1 and 2
(G) = dummy
H = measure of the condition of start-up indicated by conditions 1 and 2
I = aging after precipitation
J = source of OH^- ion, which is either NH_4OH solution or a given mixture of ammonium and sodium hydroxides
K = variable in the adjustment of the pelletor used in pelleting the dried catalyst powder, indicated by conditions 1 and 2
L = amount of additive X which was added to the powder before pelleting
M = amount of additive Y which was added to the powder before pelleting
(N) = dummy
(O) = dummy

Layout of Plackett-Burman Design

Figure 1 shows the matrix of the design, with each variable tried at two levels, "plus" denoting high level, and "minus," low level. Twelve variables were selected for study, leaving three dummy variables from which the error was estimated.

Inspection of variable *A* shows that, during the 16 runs, it appears at its high level eight times and at its low level eight times. This is also true for all of the other variables. The effect of a variable on the response is simply the difference between the average value of the response for the eight runs at the high level and the average value of the response for the eight runs at the low level, as in Equation 2:

$$E_A = \frac{R \text{ at } (+)}{8} - \frac{R \text{ at } (-)}{8} \quad (2)$$

where E_A = effect of *A* and *R* = response or result.

Although this seems reasonable enough, the question arises, "How can one variable be distinguished when all the other variables are changing at the same time?" Further examination of the matrix shows that when variable *A* is at its high level, *B* is high four times and low four times. Likewise, when *A* is at its low level, *B* is high in four runs and low in the other four. Thus, the net effect of changing variable *B* cancels out in calculating the effect of *A*. The remaining variables balance in this same way so that the net difference is only the effect of *A*.

Note the simplicity of the calculation. This feature is especially attractive to the research worker who might not have access to a modern computer. Despite the simplicity, the results are mathematically and numerically equivalent to those obtainable by a computer using a complete multiple regression analysis program.

Variables (G), (N), and (O) in the matrix of Figure 1 are dummy variables—no changes are made correspond-

ing to the pluses and minuses in these columns. The effects of the dummy variables are calculated in the same way as the effects of the real variables. If there are no interactions and all levels are reproduced perfectly with no error in measuring the response, the effect shown by a dummy variable will be 0. If the effect is not equal to 0, it is assumed to be a measure of the lack of experimental precision plus any analytical error in measuring the response. Usually, three such estimates of experimental error—that is, three dummy variables—will provide adequate confidence. However, more can be used if fewer real variables need be studied.

Equation 3 shows how the dummy effects are combined to estimate the variance of an effect.

$$V_{\text{eff}} = \frac{\sum (E_d)^2}{n} \quad (3)$$

$$= \frac{E_{(G)}^2 + E_{(N)}^2 + E_{(O)}^2}{3}$$

where V_{eff} = variance of an effect, E_d = effect shown by a dummy, and n = number of dummy variables.

The equation shows that the variance is equal to the average of the squares of the dummy effects. This equation was derived from the equation for variance given by Plackett and Burman, and lends itself readily to hand calculation.

The relationship between the variance of an effect and the standard error of an effect is shown in Equation 4:

$$S. E_{\text{eff}} = \sqrt{V_{\text{eff}}} \quad (4)$$

Thus, from the responses of the 16 runs, it is possible to calculate the effect of each of the 12 real variables and to find the standard error of the effects from the effects of the dummies. The significance of each effect can then be determined by using the familiar *t*-test:

$$t = \frac{\text{effect}}{S. E_{\text{eff}}} \quad (5)$$

In this example, the three dummy variables provide three degrees of freedom for entering the tabulated values of *t* (2).

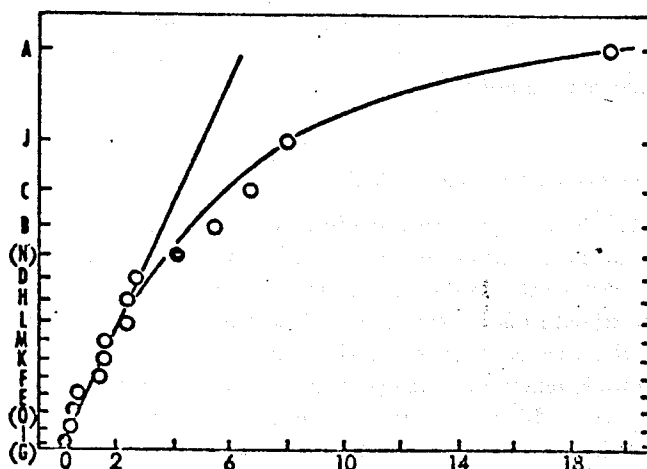


Figure 2. Half-normal probability plot of the experimental results. If the apparent effects of the variables were all due to normally distributed random error, the points would fall along a straight line.

TABLE I. VARIABLES AND EFFECTS

Code	Variable Name	Levels		Effect, (-) to (+)	Relative Significance, <i>t</i> -Test	
		Low (-)	High (+)			
A	pH	Low	High	-19.4	6.43	99%
B	M ₁ /M ₂ ratio	Low	High	5.3	2.29	80%
C	Temperature	20°	30°	6.6	2.85	90%
D	Excess anion	5%	20%	2.6	1.14	70%
E	Feed rate	2 liters/hr.	3 liters/hr.	0.5	0.19	
F	M ₂ source	1	2	1.2	0.52	
(G)	Dummy	0.116		
H	Start-up	1	2	2.3	0.99	
I	Digestion time	0	20 hr.	0.3	0.12	
J	OH ⁻ source	NH ₄ ⁺	NH ₄ ⁺ + Na ⁺	-7.8	3.37	95%
K	Pelleting	1	2	1.5	0.66	
L	Amount of X	0	X	2.2	0.95	
M	Amount of Y	0	Y	-1.5	0.66	
(N)	Dummy	3.974		
(O)	Dummy	0.354		

TABLE II. PRIMARY AND TWO-FACTOR EFFECT CONFOUNDING

Variable								
-A	pH	BM	CJ	DE	FK	GI	HN	LO
-B	M ₁ /M ₂ ratio	AM	CN	DK	EF	GL	IJ	IO
-C	Temperature	AJ	BN	DO	EL	FG	HM	IK
-D	Excess anion	AE	BK	CO	FM	GH	IN	JL
-E	Feed rate	AD	BF	CL	GN	HI	JO	KM
-F	M ₂ source	AK	BE	CG	DM	HO	IJ	LN
-(G)	Dummy	AI	BL	CF	DH	EN	JK	MO
-H	Start-up	AN	BJ	CM	DG	EI	FO	KL
-I	Digestion time	AG	BO	CK	DN	FH	FJ	LM
-J	OH ⁻ source	AC	BH	DL	EO	FI	GK	MN
-K	Pelleting	AF	BD	CI	EM	GJ	HL	NO
-L	Amount of X	AO	BG	CE	DJ	FN	HK	IM
-M	Amount of Y	AB	CH	DF	EK	GO	IL	JN
-(N)	Dummy	AH	BC	DI	EG	FL	JM	KO
-(O)	Dummy	AL	BI	CD	EJ	FI	GM	KN

The *t*-test for any individual effect allows an evaluation of the probability of finding an effect this large purely by chance when, in fact, no effect really exists. If this probability is sufficiently small, the idea that the effect was caused by the change in levels of that particular variable is accepted. The confidence level of being correct in accepting the idea is 100 × (1 - probability due to chance).

A list of the variables and the levels at which they were studied is shown in Table I. The plus and minus levels of some variables are listed as merely high and low, or coded as run by the operator. They represent definite experimental conditions for the variables, however.

Experimental Effects of Variables

The 16 catalysts were made in random order according to the conditions of the variables listed in the matrix. Each catalyst was then submitted to a standard catalytic test to determine its activity. The activity is expressed as the per cent yield of the desired product in the catalytic reaction. These experimental results are the 16 responses which are used to calculate the effects.

Table I also shows the effects found for each of the variables. Confidence levels are shown only to the 70% level; the remaining variables are considered to have insignificant effect on catalyst activity within the

ranges studied. This table illustrates the screening power of the Plackett-Burman designs because it can be seen that only four, or perhaps five, variables need be considered in further development work.

It is of interest to focus attention on the three dummy variables. Dummy variable (N) is large compared with the others. In fact, it is as large as all but the first four variables. As discussed later, this large value may be due to one of the possible two-factor interactions with which (N) is confounded. Alternatively, it may be due to experimental error. In any case, it is included in the calculation of the variance as a measure of the uncertainty of the results.

An alternate way of presenting the data of Table I is the half-normal plot proposed by Daniel (1). Figure 2 shows the variables plotted in order of the size of their effect. The probability scale used for the vertical axis is the one described by Daniel for 16-run experiments. According to theory, if the effects are due to a normally distributed random error, they should fall along a straight line when plotted in this manner. Deviation of the larger values from a straight line indicates significance. The deviation observed in the figure reinforces the conclusions evident in Table I. That is, variables A, J, C, B, and the dummy variable (N) show significant effects and appear to be real.

Discussion

It is now the investigator's duty to relate the statistical interpretation to the chemical nature of the system. The chemist would like to know why an effect calculated for a dummy variable in which no change was made appears as one of the significant effects.

It is necessary to point out that in designs which are not complete factorials, confounding exists among the variables. The Plackett-Burman design is highly fractionated. The 16-run design described here is, in effect, a 2⁻¹¹ fraction of a 2¹⁶ factorial. The main effects are not confounded with each other, but because of the high fractionation, each main effect is confounded with large numbers of two-factor, three-factor, and higher order interactions. The original Plackett-Burman treatment, although recognizing the confounding which existed, made no attempt to identify which two-factor effects were confounded with each main effect. Thus, there might be a tendency to ignore the confounding because the identities were not known.

Table II shows the confounding of the main effects with the two-factor interactions. Each main variable is indistinguishable from seven such interactions. The primary variable A is confounded with CJ, among others. Also, -C equals AJ and -J equals AC, as can be verified in the table. Because A, C, and J were all highly important variables, one might suspect that one of them may actually be only the interaction of the other two. For instance, the effect of J may be merely the -AC interaction. Similarly, one might suspect that the large dummy effect (N) could be due to the BC interaction. There is no way to verify these suspicions without further experimental work. Reference to the table of two-factor

interactions, along with chemical knowledge of the system, aids in the selection of candidates for future study and emphasizes the necessity for care.

The construction of the table is simple. Taking variable A , for example; it is noted that $-A$ equals DE . The matrix in Figure 1 shows that algebraic multiplication of $D \times E$ gives a minus in the first row. Likewise, this operation gives a minus in the second, third, and fourth rows and a plus in the fifth row. In each of these cases, the resulting sign will be opposite to the sign in the column headed by A —i.e., equal to a $(-A)$. This equality holds true throughout the remaining rows of D and E . The table of interactions is constructed in this manner for each variable by finding the seven different sets of columns which, when multiplied together, produce a sign opposite to the sign of the variable. The situation with regard to the confounding of main effects with two-factor effects is more complex than the usual confounding encountered in designs of lower fractionation. This, of course, is the price paid for the ability to handle so many primary variables in so few experiments.

Other tables could be constructed to show three-factor, four-factor, and higher order interactions, but their interpretive value is limited because of the increasing complexity of the higher order confounding.

At this point, it seems that one might question the value of reserving at least three degrees of freedom for dummy variables merely to attach significance and confidence levels to the results. If the investigator is willing to forego the luxury of significance tests, all of the available degrees of freedom can be utilized for real variables. Because the size of a dummy effect may be partially due to the presence of a sizable interaction, dummy variables are not so valuable for estimating the experimental error as they might appear to be.

Table I shows that the first four or five ranked factors would probably be chosen for further study, regardless of the significance tests assigned to them. Thus, one might almost as well make use of the three remaining degrees of freedom to screen more variables in the original set. Because further work is generally anticipated, the confidence level obtained is less important.

In conclusion, there are several other incidental consequences of using the Plackett-Burman type of design in an industrial setting. The Plackett-Burman analysis is similar to other designed experiments in that there is no restriction on the number of responses which can be measured. Thus, other responses, such as per cent impurity, precipitation efficiency or activity toward other catalytic reactions, are only a few which might be of interest. The investigator needs only to measure these properties during the 16 runs in order to include them in the study. Second, problems of the sort described here are often so complex that *a priori* judgment on the validity of some experimental variables is impos-

sible. There is ample room in this type of design to include several potential variables on the chance that they can be eliminated during the screening experiment. The rewards of doing this are substantial and should not be overlooked. A final consequence arises from the fact that planning is all done prior to beginning the runs, freeing the investigator from further daily planning. Thus, considerable periods of time are made available for reflective thought, creativity, or planning future work.

For Other Values of N

Several of the smaller designs listed by Plackett and Burman are listed below. The first row of the basic cyclic design is given opposite N , the number of experiments. For any particular value of N , the appropriate row is selected and written down as the first row. The remainder of the matrix is generated by shifting this row cyclically one place $N - 2$ times and then adding a final row of minus signs. The result will be a matrix containing N rows and $N - 1$ columns. The matrix in Figure 1 was constructed in this way for $N = 16$.

$N = 8$; + + + - + - - -
 $N = 12$; + + - + + + - - - + -
 $N = 16$; + + + + - + - + + - - + - - -
 $N = 20$; + + - - + + + + - + - + - - - - + + -
 $N = 24$; + + + + + - + - + + - - + + - - + - +
 - - - - -
 $N = 32$; - - - - + - + - + + + - + + - - - - + +
 + + + - - - + + - + - - - +

For $N = 28$, the three blocks representing the first nine rows are permuted cyclically among themselves. The final row of minus signs is then added.

+ - + + + + - - - -	- + - - - + - - - +	
+ + - + + + - - - -	- - + + - - + - - -	
- + + + + + - - - -	+ - - - + - - - + -	
- - - + - + + + + +	- - + - + - - - + -	
- - - + + - + + + +	+ - - - - + + - - -	
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