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An Integrated Screening and Optimization Strategy

Nathaniel J. Rohbock

A selected project submitted to the faculty of
Brigham Young University
in partial fulfillment of the requirements for the degree of

Master of Science

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ABSTRACT

An Integrated Screening and Optimization Strategy

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Within statistical methods, design of experiments (DOE) is well suited to make good inference from a minimal amount of data. Two types of designs within DOE are screening designs and optimization designs. Traditionally, these approaches have been necessarily separated by a gap between the objectives of each design and the methods available. Despite being so separated, in practice these designs are frequently connected by sequential experimentation. In fact, from the genesis of a project, the experimenter often knows that both designs will be necessary to accomplish his objectives. Due to advances in the understanding of experimental designs with complex aliasing and their analysis, a current topic within statistics is how to desegregate these methods into a more unified and economical approach. This project is one treatment of that issue.

Keywords: experimental design, response surface, optimization, titanium dioxide, definitive, screening, augment

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To Katherine, for her faith.

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INTRODUCTION

1.1 BACKGROUND

The experiments described in this paper have to do with refining the the process used to synthesize TiO_2 (titanium dioxide). This project is a collaboration between myself and a doctoral student, Betsy Olsen, in the chemistry department. Betsy is responsible for the experiments and resulting data used in this report, and she will be referred to as the experimenter hereafter.

The objective of interest for the experimenter is to learn more about the synthesis of TiO_2 to be a catalyst support. My objective of interest is to help the experimenter accomplish her objectives in the most efficient manner. That is, to learn which factors are truly of interest and what their relationship is to the responses of interest by using the fewest possible experiments.

As a catalyst support TiO_2 is used for diverse narrow applications requiring various properties that are characteristic of TiO_2 when (it is thought) the substance is prepared in a certain way. In each of these applications, the catalyst support will maximize the efficiency of the chemical reaction which the support is designed for. TiO_2 is a familiar substance, but the synthesis of TiO_2 is complex, and there are many variables manipulated in the preparation of the substance. The effect of each of these variables is unknown or uncertain in many cases. Measurable characteristics of TiO_2 that are of interest, are our chosen response variables. These variables are outlined in Table 1.1 with the objectives of experimentation for each response.

In these experiments, the experimental unit is the raw set of materials used to synthesize a batch of TiO_2 . Among the initial ingredients are TiCl_4 (titanium chloride), ABC

Table 1.1: Response variables and respective objectives of experimentation

Response Variables	Objective	Units of Measurement
Particle Size	Control	nm
Surface Area	Maximize	m ² /g
Pore Diameter	Control	nm
Pore Volume	Maximize	cm ³ /g

Table 1.2: Independent factors.

Factor	Description
A	Mixing Order
B	Speed of H ₂ O Addition
C	Amount H ₂ O
D	Order of Rinsing
E	Drying Time
F	Drying Temperature
G	Calcination Ramp
H	Calcination Temperature
I	Calcination Time
J	Dopant Amount

(ammonium bicarbonate), and Al(NO)₃ (aluminum nitrate). The order in which these raw materials are mixed for a given batch are labeled as either TiCl₄ and Al(NO)₃ or ABC and Al(NO)₃, or TiCl₄ and ABC. this convention corresponds to factor A, or ‘mixing order.’ The experimental procedure has been designed to be as simple as possible, and only factors thought by the experimenter to be important will be varied in the design. The effort to attain simplicity does not change the fact that the experimental procedure is complex. The factors to be varied in the experimental procedure are outlined in Table 3.1.

In Figure 1.1 a simplified flowchart of the synthesis procedure is illustrated. The red squares are the procedural steps to each run. And the yellow circles help to track the physical state of the experimental units. Note that the factors have not been worked into this diagram, also steps preceded by a dashed line may be reordered, but these issues will be addressed in greater detail in the methods section.

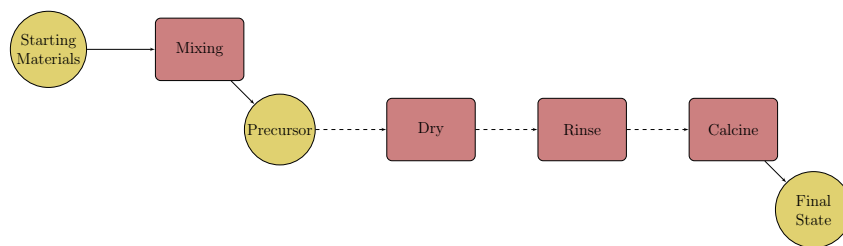


Figure 1.1: A simple diagram of a typical experiment.

There is no need for pilot testing during this research. The experimenter has been working with this process for some time in the laboratory setting, and is very familiar with which factors can be varied easily and controlled, as well as how to accurately measure responses. Due to the high cost of the equipment needed to measure responses, and the academic nature of BYU, the focus of our collaborator's department is research - not production. As a result, there is no need to consider a split plot design since the same equipment will be used for each run performed.

A possibility of lurking variables in these experiments exists. Two of note are associated with specific steps within the synthesis process. The one is grinding or mixing time and the other is the temperature of the reaction during mixing. These are the lurking variables that are most obvious. From what the experimenter has expressed, mixing time is reasonably easy to hold constant. With the equipment being used, however, it is not possible to control the reaction temperature of the reaction. This is because the change in temperature originates from the reaction taking place during the mixing step where the mixing is done by hand and the temperature is not monitored. The experimenter is aware that she must make every reasonable effort to consistently carry out these unmeasured steps in a constant fashion from run to run. These efforts are being made.

Since a run must be completed before another has begun, each piece of equipment must go through the entire cycle of preparation and operation within each experimental run. We are counting on the randomization process to wash out any effects from latent variables that may be in this process.

In order to accomplish the objectives of experimentation shown in Table 1.1, it will be necessary to bring the modeling process to run its course. Careful consideration must be given to the modeling strategy, including what information is to be gathered. A model needs to be proposed and fit to experimental data. The model should be checked against and possibly modified to comply with key assumptions. There are also other challenges to address overcome before the modeling process can be successfully completed.

The initial challenge is that we have 10 factors as candidates for main effects. Effect sparsity implies that few of these candidates are likely important. Our first step is to screen for and identify the important factors. It is then more feasible to proceed to the second step where we propose and execute an augmenting design that can accommodate our objectives.

Screening for important factors in these experiments will not be a trivial task. The first challenge arises from having 10 candidate factors. This is a large number of factors, and in order to have few runs, our factorial will have a high level of confounding. The complexity here is in the possibility of two factor interactions. Such interactions are likely to exist. If so, many possible interactions are likely confounded with main effects in small screening designs. Additionally, the possibility of quadratic terms must be faced since we are considering interpolating within the experimental region. If quadratic effects exist, we will need three levels for each factor in order to discern such an effect. These additional complexities will make screening more difficult.

LITERATURE REVIEW

2.1 SCREENING DESIGNS

The classic screening design is the fractional factorial (Box and Hunter 1961a). Fractional factorials are 2-level factor screening designs, with factor levels chosen to maximize the range of each factor within the resulting data. Among the 10 independent variables listed in Table 3.1, 8 of them are continuous (all but A and D.) The continuity of these factors supports the notion that high order interactions will most likely be negligible. Any reservations on this matter come from treatments A and D. Nevertheless, this dominant characteristic of the factor space makes the use of a fractional factorial design reasonable. The alias structure of the fractional factorials is easily obtained from defining relation. Ideally, a fractional factorial would screen out the significant effects through fitting a saturated model and examining a half normal plot (Daniel 1959) or lenth plot of effects (Lenth 1989). Once the negligible effects are discarded, the reduced model can be fit, hypotheses tested, and confidence intervals estimated.

In addition, significant effects should be cross examined against the alias structure of the design under collaboration with an expert in the field of experimentation. If the number of significant effects is large relative to the number of runs, or confounding with two factor interactions, appears to be an issue then fractional factorial designs may be augmented using a foldover design as discussed in general by (Box and Hunter 1961a). Specifically, (Montgomery and Runger 1996) give detailed strategies on how to increase resolution III designs to resolution IV designs using the foldover method.

Box and Hunter (1961a) show the advantages of a fractional factorial include relatively few runs compared to a full factorial include a natural aptitude for sequential experimenta-

tion, and that it is easy to design the number of runs in a fractional factorial according to the available resources for experimentation. For example, if many effects are expected to be significant but the resources are not available for a full factorial, then a half fraction factorial halves the number of runs from a full factorial and still has relatively few confounded variables. If an interaction is known to be non-existent or particularly unlikely, then that interaction can be used in the defining relation, since confounding on that interaction is unlikely to be an issue. Alternatively, if very few effects are expected to be significant or if resources are particularly scarce $1/4, 1/8, \dots, 1/2^{p-1}$ fractional factorials are possible. The caveat, of course, is that the smaller the fractional factorial, the more confounding there is, making it difficult to correctly confirm significant effects from associated aliases.

In the context of the experimental objective, the main drawback of using a fractional factorial is that they offer no ability to estimate quadratic effects. This is important for our case. In most cases, however, if a fractional factorial is used in order to screen for important variables, center points may then be added for the important factors, without the burden of additional runs and center points for factors that might be screened out (Box and Wilson 1951).

An alternative to fractional factorials within the realm of screening designs are designs said to have complex aliasing or to be non-regular orthogonal designs Xu et al. (2009). In fractional factorials, main effects confounded with interactions within the alias structure are *completely* confounded with each other. The only way to clear a set of confounded variables, short of knowing the real relationship, is to augment the design. In designs with complex aliasing structure, there are more confounded effects, but the confounding is not full confounding. This means that due to effect sparsity, heredity and hidden projection properties; stepwise model selection procedures (among other methods) may be able to find the correct set of effects despite partial confounding.

One set of designs that have complex aliasing are Plackett-Burmann (PB) designs Plackett and Burman (1946). Like fractional factorial designs, Plackett-Burmann designs

have two levels for each factor. They do not, however, have generators or defining relations as in fraction factorial designs and aliasing structure is not as straightforward to obtain. Another difference between Plackett-Burmann and fraction factorial designs is that while fraction factorial designs have runs restricted to powers of two, the number of runs within a Plackett-Burmann design are factors of 4.

2.2 DESIGNS FOR FITTING QUADRATIC MODELS

Classic Designs

Traditionally, when fitting a response surface design, one of two methods have been used. The first is discussed by Box and Wilson (1951) and has since been dubbed the central composite design. The method consists of choosing a screening (usually a factorial) design and then adding center and axial points in order to estimate quadratic effects.

The other method are a class of designs described by Box and Behnken (1960), incidentally referred to as Box-Behnken designs. These designs are made, in the author's own words, by combining two-level factorial designs with incomplete block designs in a particular manner.

The designs discussed in this section are proven designs, but there are also more recently devised designs which require fewer experimental runs (with caveats). In order to give context to the experimental designs in the following section, in the case of 10 factors, running a traditional Box-Behnken design requires 170 experimental runs.

Minimal Run Designs

In the spirit of fractional factorials, though non-regular, the fractional Box-Behnken design (FBBD) is presented as a single step screening and response surface design by Edwards and Mee (2011). In the paper cited, design structures are given for fractional Box-Behnken designs ranging from 4 to 13 factors. In relation to this project, with 10 candidate factors,

the FBBB suggested in the paper is a $(1/2)$ BB10 requiring as few as 81 runs. The FBBB is constructed by taking subsets of the two level factorial designs that make up the Box-Behnken design.

The proposed strategy for fitting a model to a FBBB is outlined by Edwards and Mee (2011) as a three step process. First, a backward elimination step is performed. The purpose of this step is to find a ‘tentative second order model.’ Initially one fits $(T-1)$ -factor second order models and selecting the one that gives the smallest RMSE. Afterwards, an F-statistic is used to test the null that all $t + 1$ parameters involving each individual factor are negligible. If the p-value is greater than a pre-determined α , then the factor is dropped. This second part of the step is iterated until no more factors can be dropped.

Next, an extra diagnostic step is taken. The purpose of this step is to identify factors that may have previously been eliminated but deserve further consideration. For each of T projections onto $T-1$ factors, one estimates the coefficients for the *full* second order model. Treating the modeling error as random error, t -statistics are constructed for the coefficients in each of the T model fits. This should give a sense of importance for each factor. The linear main effect estimates are plotted from the T model fits. Estimates consistently away from 0 indicate an active main effect with partial aliasing with inactive interactions. Inconsistent estimates may also be indicative of an effect and should be investigated in context of the set including its partial aliases.

Finally, the second order model should be fit using all the factors identified in the elimination and diagnostic steps. Edwards and Mee acknowledge that the final result is likely to be overfit, but point out that using the model including insignificant terms is common practice when searching for a stationary point.

The FBBB designs discussed are an attractive approach for our problem, but comes at a cost of at least 81 experimental runs. Herein lies the strength and the weakness of the design. The FBBB approach just discussed allows for the efficient estimation of a second-

order model in at least T-1 Factors, but the design is not friendly to a researcher who does not believe that there is a need to have T-1 factors in his second-order model.

For such a researcher, there are definitive screening designs (Jones and Nachtsheim 2011). Suppose the quadratic model

$$\mathbf{y} = \mathbf{X}_1\beta_1 + \mathbf{X}_2\beta_2 + \epsilon$$

approximates the true relationship between \mathbf{y} and the independent variables well. If \mathbf{X}_1 is the design matrix corresponding to the main effects, and \mathbf{X}_2 corresponds to the second order effects, then the estimates for effects in an underfit model using only main effects is given by

$$E(\hat{\beta}_1) = \beta_1 + \mathbf{A}\beta_2.$$

In a definitive screening design, \mathbf{A} is $\mathbf{0}$ and the estimates of main effects are unbiased by any existing linear by linear interactions. This characteristic is only true of a fractional factorial of resolution IV. For $m = 10$ factors, this would require a 2^{10-5} design with 32 runs. Definitive screening designs are so called because they achieve this in fewer runs ($2m + 1 = 21$ in this case) and still offer the ability to fit quadratic terms since every run has a center point for a factor and every factor has 1 center point within the whole experiment. Additionally, for designs with 6-12 factors, response surface models can be fit for three or fewer factors with high efficiency.

From the properties described, for a 10 factor experiment, the definitive screening design allows two factor interactions and quadratic terms as in a resolution V design but in fewer runs than a resolution III fractional factorial design. This precludes any concern about partial confounding among two-factor interactions and quadratic terms within the definitive screening design. Although the FBBD allows for a second-order model, the definitive screening design requires fewer runs and is the clear choice if few factors are expected to be significant.

2.3 ANALYSIS OF DATA FOR QUADRATIC MODELS AND RUN DESIGNS WITH COMPLEX ALIASING

Some of the designs discussed thus far have the ability to provide the data needed to fit the models we need, however, for the most part the discussion thus far has not addressed how to adequately explore the factor space and select a final model. Part of the difficulty is that there are more second-order effects to be screened than there are data points. In order to overcome this, there are diverse methods available some of which are computationally demanding, and do not necessarily have clear advantages one over the other.

Perhaps the least creative approach to select a model is to perform an all subsets regression. Among the results, the models may be sorted by R^2 , or some other measure of model fit, and the most sensible model selected from the top results. This should be done while keeping in mind principles of effect sparsity and heredity. The first drawback to the method is how quickly it renders itself infeasible. An all subsets algorithm has to fit $\binom{n}{p}$ models if n is the number of effects fed to the algorithm and p is the number of terms called for in the final model. Another drawback is that the best performing models are sometimes difficult to interpret in context of effect heredity because the model can be quite overfit using seemingly unrelated sets of factors.

A more clever way presented by Lawson (2002) to help select a good subset of factors is to limit the number of candidate interactions using the alias structure of the design. Once the candidate factor space is reduced, any method of selection including all subsets can be applied to the restricted factor space. This method is useful because it prioritizes how the factor space is explored according to what can most influence the bias of the final model.

Briefly, in Lawson's method, the largest main effects are identified as candidates. Next, candidate interactions are identified by using the alias matrix to see which higher order effects, if they exist, would contribute the most to bias in an underfit model. Then

an all subsets regression, or other search method, is performed simultaneously on the large main effects and on this limited subset of candidate interactions.

There is still a risk for the selection of an underfit model and consequent biased estimates. The strength of the method is that it ensures that if an underfit model is settled on, that the effects left out will be those which would bias the remaining estimates less than those which were included in the model search.

With regard to designs having complex aliasing, there are strategies to make the most of the relatively few runs needed to run the experiment. These strategies attempt to make the most of effect sparsity, heredity and projection properties. One such approach proposed by Hamada and Wu (1992) is a relatively simple 3 step algorithm:

- 1.) Entertain all main effects and interactions that are orthogonal to the main effects. Select candidates using standard methods.
- 2.) Using effect heredity, in addition to selected candidates thus far, entertain two-factor interactions that have at least one component factor among the candidates. Also entertain interactions suggested by the experimenter. Use forward selection to augment existing candidates.
- 3.) Using the existing candidates, use forward selection to identify significant effects. Repeat step 2.

Steps 2 and 3 should be iterated until the selected model in step 3 stops changing. The authors suggest that if a more thorough search is desired, the model at this phase is a good starting model. From this point a search can be made by adding or deleting effects one at a time. This method is preferred by the authors over an all subsets regression, especially since final models are more interpretable in the context of effect heredity.

Weaknesses of this main approach are acknowledged by Hamada and Wu and should be kept in mind. If interaction effects larger than main effects exist, this can cause a failure to detect important main effects when using standard methods such as half normal plots in

step 1 of the algorithm. In such a situation the proposed algorithm would be helpless to find the missed main effects. A similar variation on the theme is seen when interactions of equal size to an unrelated main effect but inheriting significance from another main effect exist. In step 1, the main effect found in the interactions will likely appear significant, but the other unrelated main effect may be masked by the still undiscovered interactions. My observation is that it may be wise to employ the alias matrix as suggested by Lawson initially to identify main effects that may be masked in step 1 of Hamada and Wu's algorithm before proceeding.

Notice that the weaknesses of this approach arise from step 1 of Hamada and Wu's algorithm. They suggest an alternative step 1 in the paper that may be more robust: For each factor X , that X and all interactions of the form XY if $X \neq Y$, should be entertained and shrunk using forward selection. Selected variables from each factor should be pooled and then starting at step 3 iterated through steps 2 and 3 as before. Finally, the authors suggest that if several incompatible but equally plausible models remain after analysis, it is a strong indication that no analytical method will successfully distinguish the most correct model. In such a situation there is no substitute for augmentation.

The success Hamada and Wu's algorithm, was later explained by its utilization of hidden projection (Wang and Wu 1995). In the paper, Wang and Wu explain that a design has a hidden projection property if a subset of interactions can be estimated even if the projected design does not have the right resolution or other combinatorial design property for that subset of interactions to be estimated. Hamada and Wu's algorithm is significant because it demonstrated that designs with complex aliasing, which had been considered only useful for identifying important main effects, could successfully be used to identify interactions as well. This article spurred considerable of research which has advanced the statistician's ability to get more from experimental designs with complex aliasing. In each case this is possible because of the hidden projection property described by Wang and Wu.

The final strategy to be discussed here is the Bayesian variable selection model discussed by Chipman et al. (1997). This approach assigns prior probabilities for each candidate

parameter and builds posterior probabilities for the activity of each parameter. Once the algorithm is done running, those parameters with the binomial posterior greater than a certain cutoff value are selected for the final model. The advantage of this search method is that it theoretically allows the entire model space to be explored. Because the prior probabilities are built to reflect our assumption of effect heredity, the final model will likely reflect that assumption. In the end it has the ability to select a model with main effects and interactions, but without the drawbacks of a forward selection procedure or a restricted candidate parameter space.

3.1 THE SCREENING DESIGN

In order to identify the significant factors in the synthesis procedure, a screening design is most appropriate for the first step of this project. With the selected factors, the screening design for this project is a sixty-fourth fraction factorial (2^{10-6}). This specific design was selected for the number of runs (16) necessary to complete it and because we believed that it would successfully eliminate factors from future experiments. In order to create the design for execution, the drop down menus in JMP were used to produce and randomize the design. The generators used by JMP to produce the design were $E=ABCD$, $F=BCD$, $G=ACD$, $H=CD$, $I=ABD$, and $J=ABC$. From the generators, the defining relation was obtained and the aliasing structure shown in Table 3.1 is derived.

The selected design is resolution III. In practice, this means that main effects are confounded with two-factor interactions and higher order terms. The randomized run scheme for the screening experiment is available for reference in Table A.1 located in the appendix. There is a high level of confounding in this design, however, we expect few factors to be significant with respect to the response variables. The full list of factors is outlined in Table 3.1 along with the levels to be used for screening and the lower order aliasing structure for each factor.

In context of the synthesis procedure, a more complete illustration of the synthesis procedure and the relative sequence of factors is given in Figure 3.1. In the figure, the red squares are the procedural steps to each experiment and the yellow circles help to track the physical state of the experimental units. The addition of the blue diamonds correspond to where factors are controlled.

Table 3.1: Dependent variables and aliasing structure up to first order interactions.

Factor	Description	Levels	Confounded two-factor interactions
A	Mixing Order	ABC & AlNO ₃ , TiCl ₄ & AlNO ₃	FE HG
B	Speed of H ₂ O Addition	Slow, Fast	HF GE
C	Amount H ₂ O	7 ml, 25 ml	HD IE
D	Order of Rinsing	DRC, DCR	JE HC
E	Drying Time	3 hrs, 24 hrs	JD IC BG FA
F	Drying Temperature	25°C, 100°C	HB EA
G	Calcination Ramp	2°C/min, 20°C/min	BE HA
H	Calcination Temperature	400°C, 700°C	FB JI DC GA
I	Calcination Time	2 hrs, 20 hrs	HJ EC
J	Dopant Amount	5%, 22%	HI DE

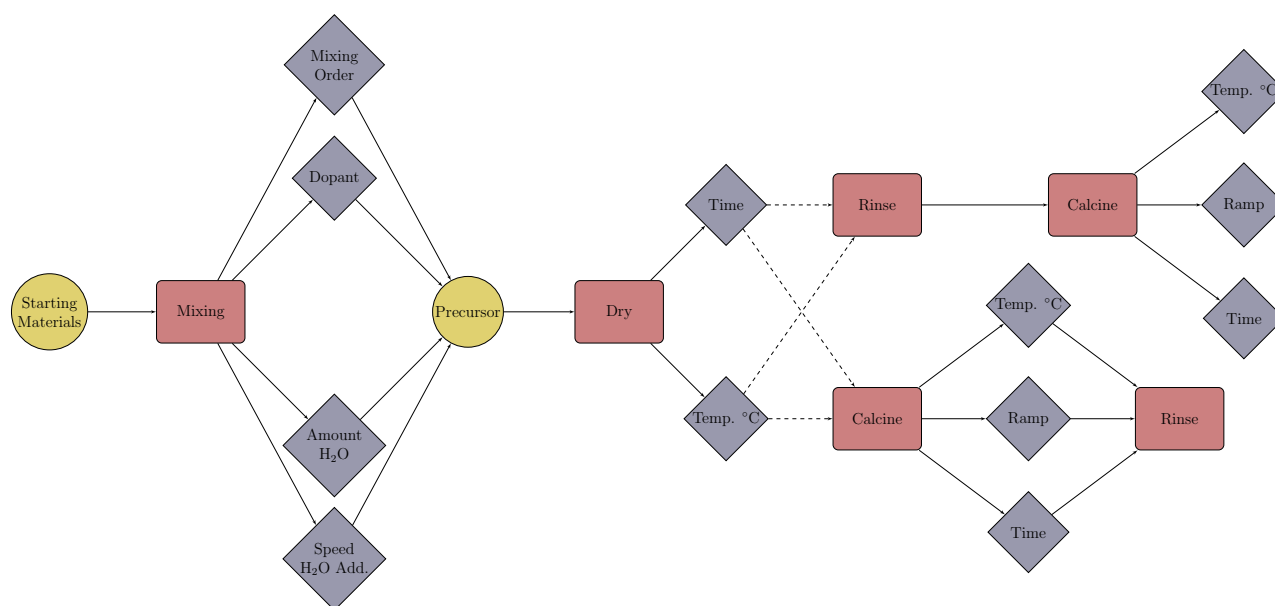


Figure 3.1: A detailed diagram of the experiment process for TiO₂ synthesis.

Again, the purpose of the screening step is to allow our expectation that few factors will be of importance to simplify the process of building a good model. At this stage of the experiment the power to estimate differences at, say, a level of $\alpha = .05$ is of secondary concern. Hence, in the randomized run scheme the 2^{10-6} factorial is made in 16 runs - the minimum that can be used. Since there are no replicates, we have no ability to formally estimate confidence intervals or perform formal tests, but we can use graphical methods to determine what effects are likely to be significant.

Once we have defined our experiment and how many runs will be used, it is a good idea to check what size of effect can be detected at level $\alpha = .05$. As suggested by Wheeler (1974), the size of a practical effect, Δ , may be obtained from the equation:

$$N = ((8\sigma)/\Delta)^2.$$

Given $N = 16$, Δ is solved for giving,

$$\Delta = 2\sigma.$$

From this calculation, we expect that from the design should be able to detect effects that are twice the size of the experimental error with reasonable power.

The experiments for the 2^{10-6} were performed and both half-normal plots of effects and Lenth's plot of effects (Lenth (1989)) were used to detect important effects. The half-normal plots are given in Figure 3.2 and the Lenth plots are in Figure 3.3.

From the Lenth plots, it is clear that H - calcination temperature and D - rinse order are the most important factors for multiple responses. For other factors, the evidence of significance is less strong. Still, we were hesitant to rule out several factors including C - amount H_2O because of the half-normal plot for pore volume and B - speed of H_2O addition because its effect ranks high for a majority of the responses. In the most conservative case, considering only H - calcination temperature and D - rinse order, there is still quite a lot of aliasing as outlined in Table 3.2 respective to response. Unfortunately, H - calcination temperature is the most heavily aliased factor in the fractional factorial. This was partially by design since the experimenter expected H - calcination temperature to be the most significant. The results were discussed with the experimenter, but she after considering which factors she believed could be eliminated from further experimentation, none of the factors could be eliminated for all four responses. As a result between D - rinse order and H - calcination temperature, every factor is present as either a significant factor or in an interaction as an alias for a significant factor.

Table 3.2: Factors that were not eliminated by the screening design by response.

Response	Uneliminated Factors	Factor Aliases
Particle Size	H	FB JI DC GA
Surface Area	H	FB JI DC GA
Pore Diameter	D	JE HC
Pore Volume	D	JE HC

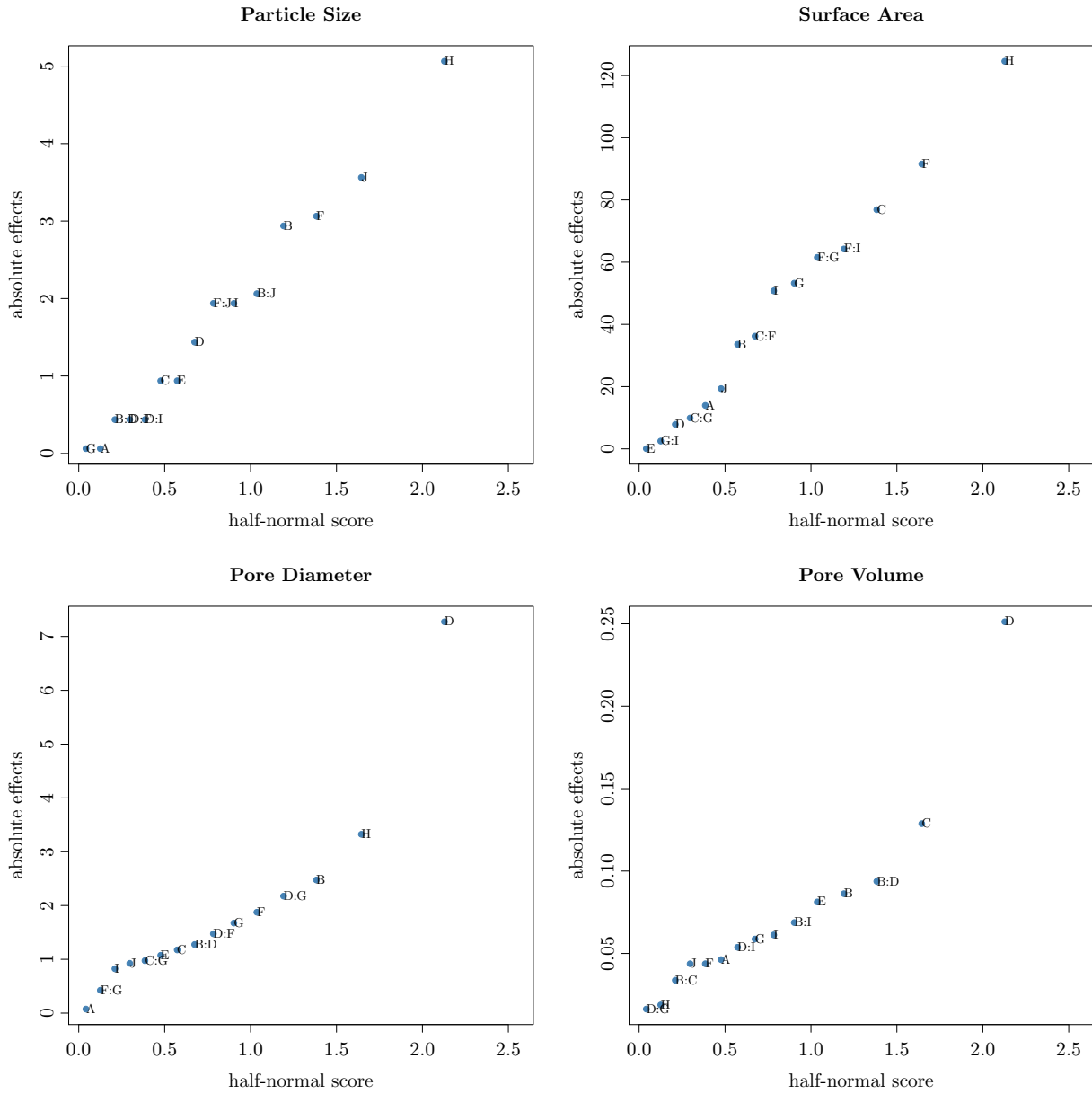


Figure 3.2: Half-Normal plots for each response.

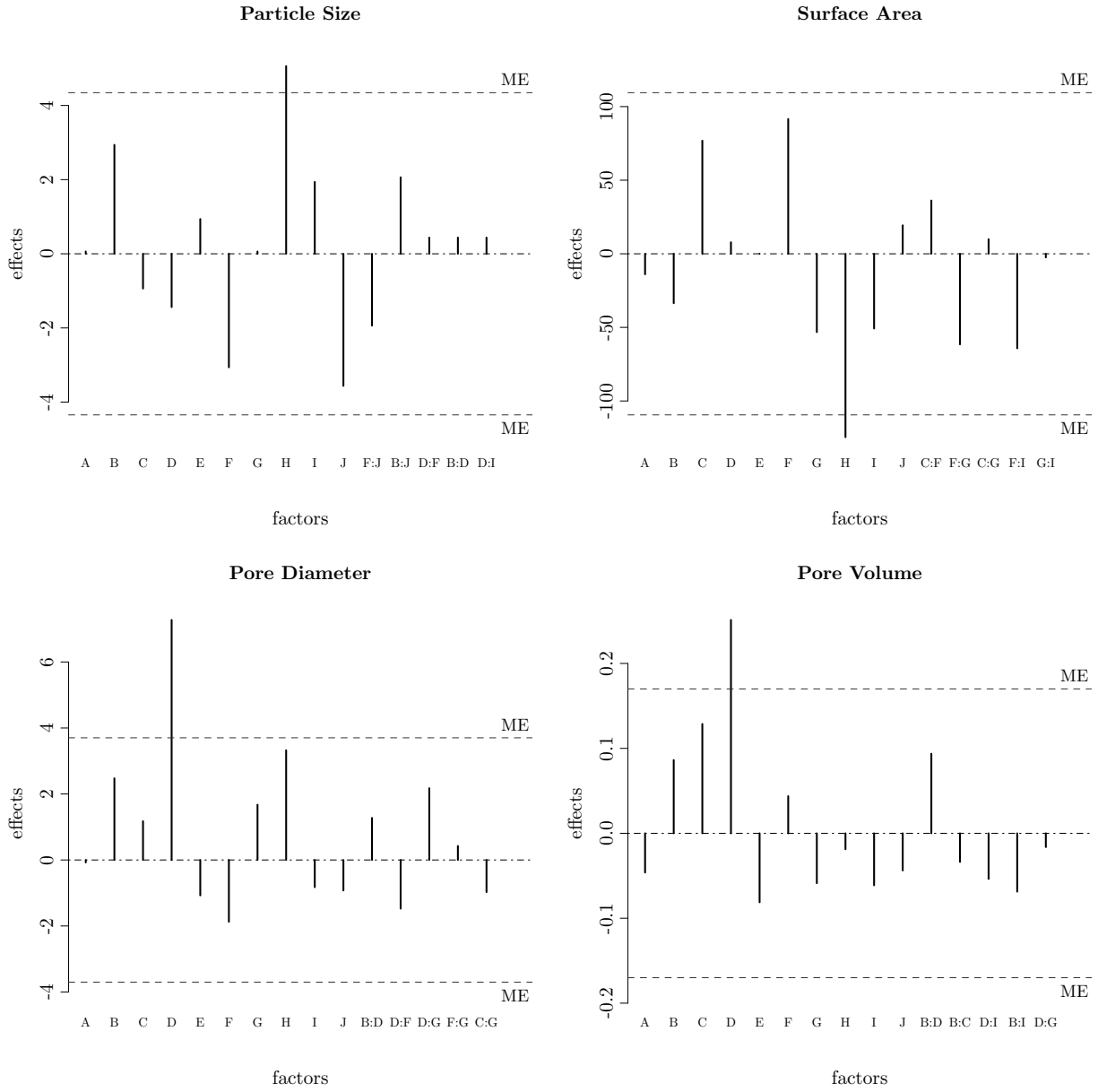


Figure 3.3: Lenth plots for each response, shown for $\alpha = 0.1$.

3.2 AUGMENTING THE DESIGN

In order to keep the total number of experiments required low, but also create the ability to fit a response surface model, we opted to employ the definitive screening design. Since there were still $m = 10$ factors under consideration, there were 21 experiments needed to complete this new step. If this design and the existing data are successful in screening out at least 6 factors for surface area and pore volume, then there should be sufficiently many experiments in order to fit a response surface model on the remaining 4 or fewer factors. Table 3.3 shows the new center points for each factor.

Table 3.3: Dependent variables with the three factor levels listed for each.

Factor	Description	Levels
A	Mixing Order	ABC & AlNO ₃ , TiCl ₄ & AlNO ₃ , TiCl ₄ and ABC
B	Speed of H ₂ O Addition	Very Slow, Slow, Fast
C	Amount H ₂ O	4 ml, 7 ml, 25 ml
D	Order of Rinsing	DRC, DCR, RDC
E	Drying Time	3 hrs, 6 hrs, 24 hrs
F	Drying Temperature	25°C, 50°C, 100°C
G	Calcination Ramp	2°C/min, 10°C/min, 20°C/min
H	Calcination Temperature	400°C, 550°C, 700°C
I	Calcination Time	2 hrs, 20 hrs, 40 hrs
J	Dopant Amount	5%, 14%, 22%

The list of randomized experiments for the augmenting design are given in Table A.2 in the appendix. As the factorial design before, this list was generated by using the drop-down menus in JMP after installing the definitive screening add-in from the SAS website. Examining the list, one can see that there is one center point for each experiment and one experiment with only center points. This comes out to three center points for each factor.

The strategy moving forward differs from the traditional approach in screening and optimization studies. The traditional approach can be outlined as follows:

- i.) Complete a resolution III screening design.

Use graphical procedures to identify large effects. In this project, this was done in 16 experiments.

- ii.) Complete a foldover design.

This augmentation would deconfound main effects from 2-factor interactions. Graphical techniques would be used in this step in order to identify whether main effects or strings of interactions are important. This would require 16 additional experiments, cumulatively 32 experiments.

- iii.) Perform further augmentation experiments.

This step is meant to deconfound significant 2-factor interactions from each other, and strings of confounded 2-factor interactions. Regression could be used in order to identify important main effects and two factor interactions after the experiments are run. This would likely require 16-32 additional runs, or 48-64 runs to this point.

- iv.) Complete final augmentation experiments.

This last augmentation would be to add center and axial points for the important variables. Once the experiments are run, a response surface model would be fit in the important factors. This would likely require 10 to 16 additional experiments, bringing the grand total number of experiments run to be between 58 and 80.

The strategy I employed is outlined differently:

- i.) Run the resolution III screening design.

Use graphical procedures to identify large effects. In this project, this was done in 16 experiments.

- ii.) Augment the resolution III screening design with a definitive screening design.

This augmenting design would partially unconfound main effects from both 2-factor interactions and from quadratic effects. Once these final experiments are completed, a

variable selection algorithm will be needed in order to find the important factors. This was done in 21 additional experiments, bringing the current total (hopefully the grand total) to 37 experiments.

3.3 SELECTING VARIABLES FOR THE FINAL MODELS

Once the experiments involved in my approach were run and the data collected, the next step was to perform the factor selection procedure. For this challenge, two strategies were implemented: a Bayesian variable selection (BVS) algorithm and an alias matrix guided search.

The alias guided search aided in selecting a subset of factors for an all-subsets regression. The factors selected for the all-subsets regression are given in Figures 3.4 through 3.7. Once the all-subsets regression was completed, MSE plots were produced for the top 10 model results in each category, where category is determined by the number of parameters in a model, ranging from 2 to 7 parameters. These plots are also included in Figures 3.4 to 3.7. A reasonable model size is chosen based on where diminishing returns with respect to reducing MSE appears to take place. The factors selected are shown by response in Table 3.4.

In order to run the BVS algorithm it is necessary to first specify the priors δ_i for the activity of each effect included in the search. Using A and B as example effects, the priors are chosen as follows:

$$Pr(\delta_A = 1) \stackrel{def}{=} \pi \stackrel{set}{=} 0.25$$

$$Pr(\delta_{AB} = 1) \stackrel{def}{=} \begin{cases} p_{00}/\pi \stackrel{set}{=} 0.1 & \text{if } \delta_A = \delta_B = 0 \\ p_{10}/\pi \stackrel{set}{=} 0.5 & \text{if } \delta_A \neq \delta_B \\ p_{11}/\pi \stackrel{set}{=} 1.0 & \text{if } \delta_A = \delta_B = 1 \end{cases}$$

Once the priors were specified, the BVS algorithm returned posteriors for each effect in a full second order model as illustrated in Figure 3.8. From the results, we picked factors

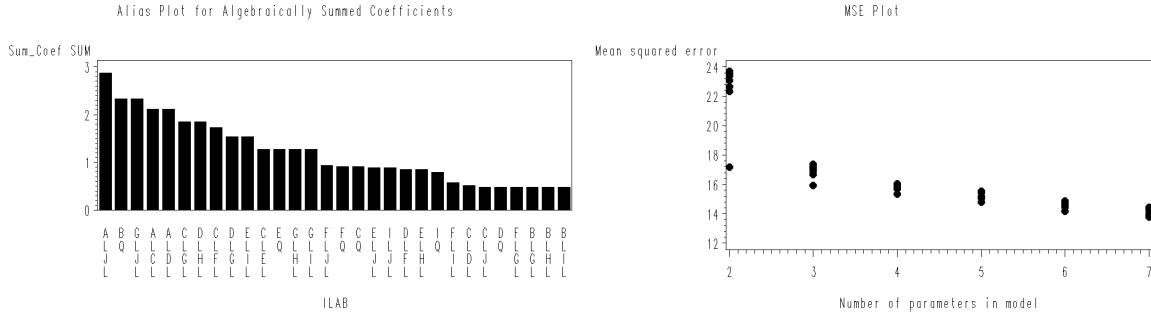


Figure 3.5: These are the sums of the bias coefficients for the model with particle size as the response. From this plot, $A \times J$ through C^2 , as listed on the plot, were selected for the all subsets regression. On the right, from the MSE plot it appears that 4 variables is a reasonable number to select.

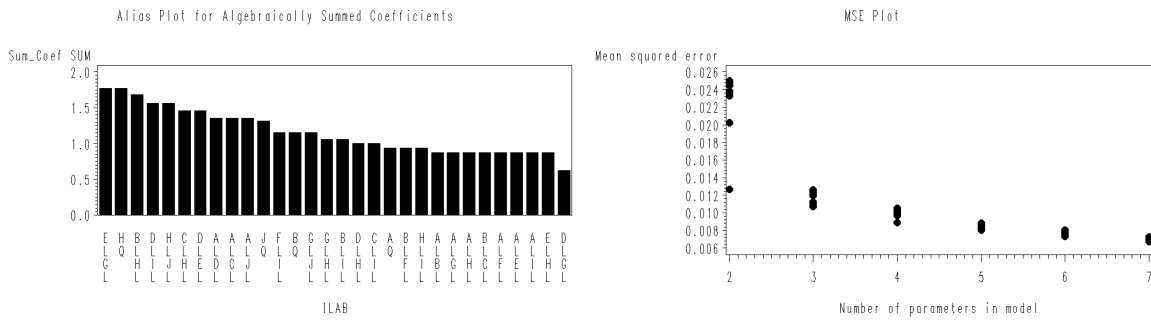


Figure 3.6: These are the sums of the bias coefficients for the model with pore volume as the response. From this plot, $E \times G$ through $H \times I$, as listed on the plot, were selected for the all subsets regression. On the right, the MSE plot appears to indicate that 3 or 4 parameters is a reasonable model size.

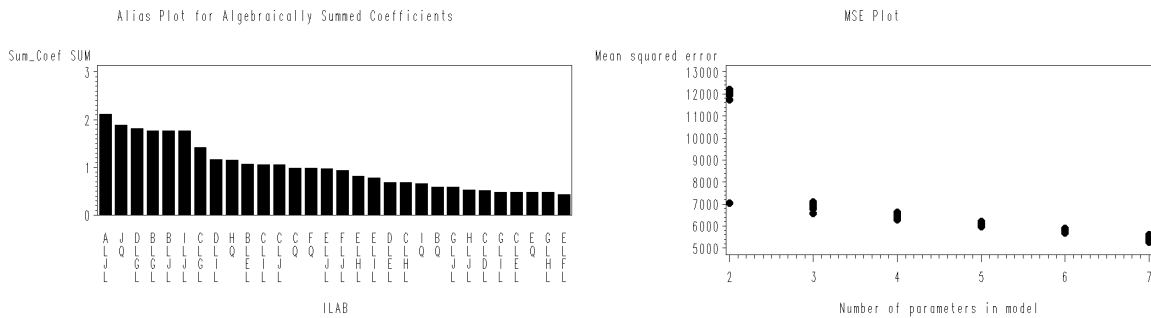


Figure 3.7: These are the sums of the bias coefficients for the model with surface area as the response. From this plot, $A \times J$ through $E \times I$, as listed on the plot, were selected for the all subsets regression. On the right, the MSE plot seems to indicate that 3 parameters is a reasonable model size.

Posterior Probabilities for Candidate Effects

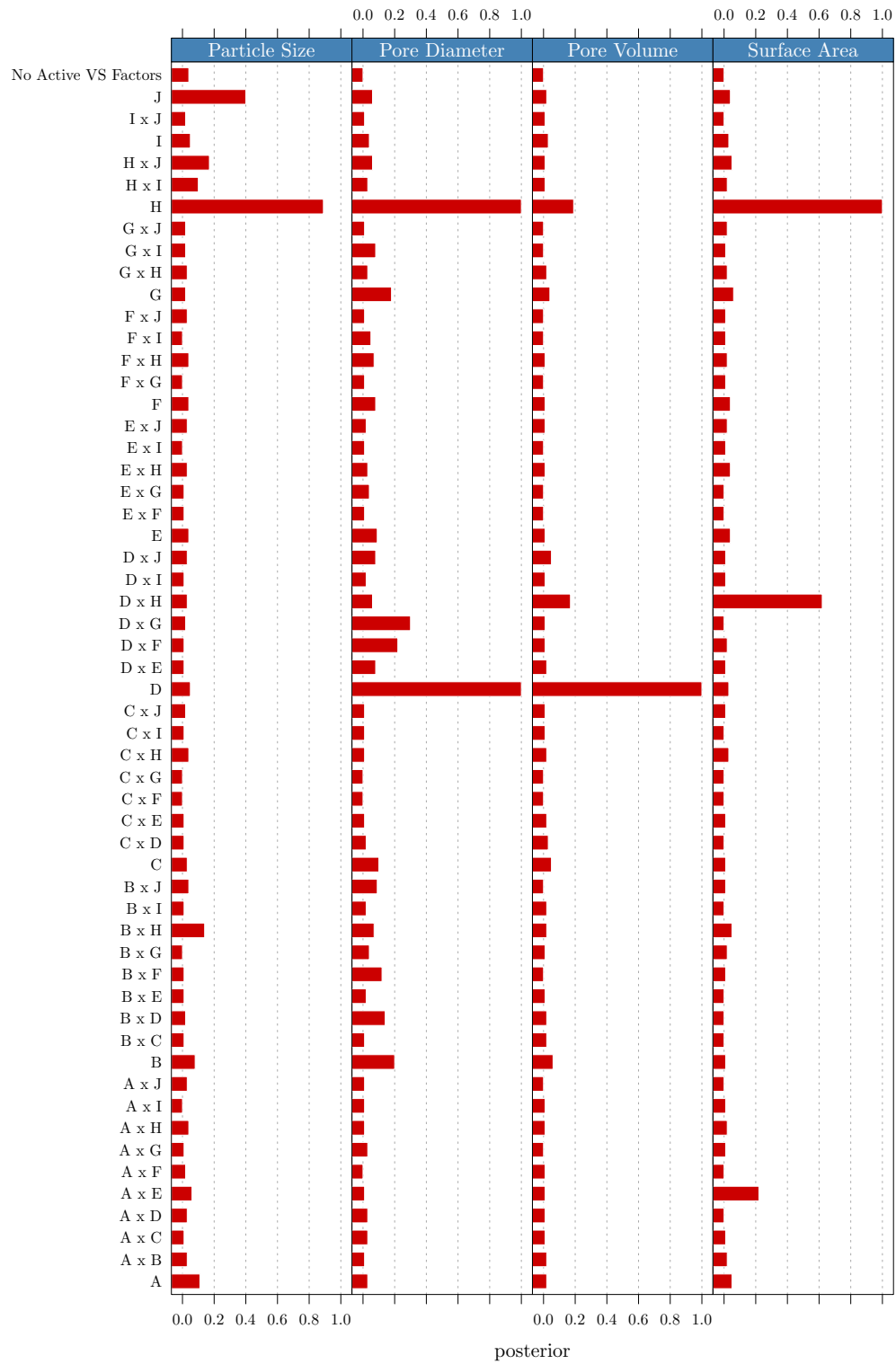


Figure 3.8: Using the posterior probabilities above, the top factors (up to 3 or 4) were selected as outlined in Table 3.4

RESULTS

4.1 FINAL MODELS

Particle Size

For particle size, the variables selected for the initial model were B - speed of H₂O addition, D - rinse order, H - calcination temperature, and J - dopant amount. The objective for particle size is to simply to find a model to aid in controlling the response. In order to do this, I simply started with a full model including the four factors previously selected and eliminated the most insignificant variable in a stepwise fashion. This gives the model summarized in Table 4.1.

Table 4.1: The table below summarizes the final model for particle size. J - dopant amount perhaps should have been eliminated, I felt that the information was useful.

	Estimate	Std. Error	t value	Pr(> t)
(Intercept)	9.8860	0.6556	15.08	0.0000
H	2.6618	0.6829	3.90	0.0004
J	-1.2785	0.6426	-1.99	0.0547

Qualitatively this says the following: that as H - calcination temperature is increased, particle size also increases and that as J - dopant amount is increased, particle size decreases. In order to apply this model in practice, use the following equation:

$$\text{Particle Size} = 9.8860 + 2.6618 \frac{(H - 550)}{150} - 1.2785 \frac{(J - 14)}{8}$$

if $400^\circ < H < 700^\circ$ and $5 < J < 22$.

Surface Area

The objective with regard to surface area is to maximize the response. As previously mentioned, the variable selection methods suggested quite different models, both were fit, and the variables selected by the Bayesian selection algorithm yielded the better model. The variables suggested by the Bayesian selection algorithm were A - mixing order, D - rinse order, E - drying time, and H - calcination temperature. Note that E - drying time and H - calcination temperature are continuous variables and that A - mixing order and D - rinse order are categorical factors with no natural ordering.

To simplify the discussion, consider the categorical factor D - rinse order. Since there was a need to fit a response surface model for this response, D - rinse order presented difficulties. If D - rinse order is crossed with other factors it is difficult to interpret. If D - rinse order is not crossed with other factors, but left in as a lone categorical factor, any effect it has is analogous to producing a separate response surface for each level of D - rinse order. This produces a higher level of variability in the model and, in fact, there is no reason to expect the shape of the response surface to even be similar from level to level of D - rinse order. Additionally, this latter approach could also decrease our power to test the significance of other variables in the model. A presents the same difficulties as D - rinse order, so for these reasons, we decided to subset on the two extreme levels of A - mixing order and D - rinse order and to exclude the center point runs from the final model fits.

There were not enough runs to fit subsets of the experiments by holding A - mixing order and D - rinse order constant for each of the four combinations of interest and fit a model with each, so I explored them separately. I did this by fitting a response surface where A - mixing order or D - rinse order was held constant at one level, then proceeded to fit the second order model in E - drying time and H - calcination temperature. I did this for each of the levels of A - mixing order and D - rinse order and observed that holding A - mixing order constant at $\text{TiCl}_4(\text{NO}_3)$ appears to give the maximum response. This approach led

me to hold A - mixing order constant at that level for the final model summarized in Table 4.2.

Table 4.2: The table below summarizes the final model for surface area where A is held constant at TiCl_4 & $\text{Al}(\text{NO}_3)$, and a second order model is fit in E and H.

	Estimate	Std. Error	t value	Pr(> t)
(Intercept)	130.2833	35.1096	3.71	0.0034
E	115.5834	21.2822	5.43	0.0002
H	-79.0208	6.2961	-12.55	0.0000
E:H	-52.7925	6.4622	-8.17	0.0000
E ²	-92.6548	31.6327	-2.93	0.0137
H ²	94.3375	25.1588	3.75	0.0032

The response surface area for this model is low-dimensional enough to be illustrated, and this illustration is given in Figure 4.1. From the diagram it is clear that the response maximizes near the experimental points of $H = -1$ and $E = 1$. These points correspond to an H - calcination temperature of 400° celsius and a E - drying time of 18 hours.

Pore Diameter

For pore diameter, the objective (as with particle size), is to make a model to understand how to control the response. To this end, I started with the full model using the four factors suggested by both variable selection methods used. From this step, although G - calcination ramp could have been eliminated for a p-value greater than 0.05, I chose not to reduce the model any further. The summarized model is given in Table 4.3

Table 4.3: The table below summarizes the final model for pore diameter.

	Estimate	Std. Error	t value	Pr(> t)
(Intercept)	11.0076	0.5733	19.20	0.0000
D (DRC)	-6.7218	0.7378	-9.11	0.0000
D (RDC)	-6.4831	1.3673	-4.74	0.0000
B	1.4255	0.5701	2.50	0.0179
H	1.7474	0.3689	4.74	0.0000
G	0.6263	0.3277	1.91	0.0653

Conceptually this says that for D - rinse order of DCR, the response starts highest. As B - speed of H₂O addition increases, so does pore diameter. That as H - calcination temperature is increased, pore diameter also increases. Finally that as G - calcination temperature ramp is increased, pore diameter also increases. In order to apply this model in practice, use the following equation:

$$\text{Pore Diameter} = \begin{cases} 11.0076 + 1.4255B + 1.7474\frac{(H-550)}{150} + 0.6263\frac{(G-10)}{8} & \text{if D = DCR} \\ 11.0076 - 6.7218 + 1.4255B + 1.7474\frac{(H-550)}{150} + 0.6263\frac{(G-10)}{8} & \text{if D = DRC} \\ 11.0076 - 6.4831 + 1.4255B + 1.7474\frac{(H-550)}{150} + 0.6263\frac{(G-10)}{8} & \text{if D = RDC} \end{cases}$$

if B = -1 for very slow, 0 for slow, and 1 for fast. Also the following must hold: $400^\circ < H < 700^\circ$, B must be somewhere in between very slow and fast as the experimenter has defined it to be (qualitatively), $2^\circ \text{ C/min} < G < 20^\circ \text{ C/min}$.

Pore Volume

As with surface area, the objective for pore volume is to maximize the response. The challenge that existed for the use of the factor D - rinse order is the same for pore volume, and the approach employed to identify the correct level of D - rinse order to hold constant was the same. As a result, for the final model, D - rinse order is held constant at DRC and the second order model is fit in B - speed H₂O addition and H calcination temperature. The final model is summarized in Table 4.4.

Table 4.4: The table below summarizes the final model for pore volume where D is held constant at DRC, and a second order model is fit in B and H.

	Estimate	Std. Error	t value	Pr(> t)
(Intercept)	0.0330	0.0548	0.60	0.5589
B	-0.0295	0.0371	-0.80	0.4429
H	-0.0889	0.0209	-4.25	0.0014
B:H	0.0367	0.0285	1.29	0.2236
B ²	0.0512	0.0440	1.16	0.2687
H ²	0.1579	0.0586	2.69	0.0209

The response surface area for this model is also low-dimensional enough to be illustrated, and this illustration is given in Figure 4.2. From the diagram it is easy to see that the response maximizes near the experimental points of $H = -1$ and $B = -1$. These points correspond to an H - calcination temperature of 400° celsius and a B - addition of H_2O speed of very slow. This is valid for when D - rinse order is held constant at DRC.

4.2 MODEL ASSESSMENT

In order to assess the validity of the response surface models, perhaps the most crucial diagnostic to check is where the experimental points are on each surface and how many of them there are. This is necessary since we subset on the levels of D - rinse order and A - mixing order depending on the response. By subsetting in this manner, it turns out that the experimental region is no longer covered by experimental points as was intended by the original design.

For surface area, 3D surface plot and contour plot with the latter overlaid with the experimental points are given in Figure 4.1. Note that in the contour plot in the lower right region, where drying time is long and calcination temperature is low and where the response appears to be maximized, is covered by experimental points. This is the most crucial support we need for this model's validity.

For pore volume the same plots were created as for surface area and is given in Figure 4.2. Upon examining the placement of the experimental points on the contour plot, it is clear that the region where the response appears to be maximized (in the lower left region where calcination temperature is low and the speed of adding water is also low) is not covered by the experimental points. This is a concern, and it will be necessary for the experimenter to run confirmatory experiments in order to validate the extrapolation given by the response surface illustrated in the figure. Despite this drawback, there are experimental points at $B = 0$ and $H = -1$ which support a trend that are consistent with the conclusion given by the model. I think it is reasonable to expect confirmation of the response illustrated.

The next assessment are the predicted versus residual plots shown in Figures 4.3 and 4.4. The residuals in these plots are colored by A - mixing order or D - rinse order as specified in the titles and legends. The reason for coloring the residuals by these colors is simply to see if there is any obviously unexpected behavior that can be associated with these two categorical variables.

From the two sets of plots, I don't see anything concerning about the residuals that I can associate with either A - mixing order or D - rinse order, except in the pore diameter model. I should acknowledge that in the pore diameter plot colored by D - rinse order there is a distinct vertical separation of the predicted values of the response, but this is expected as is clear from the fact that D - rinse order is a predictor in the model. The concern that does arise is the apparent heterogeneity of variance that appears in Figure 4.4. One way that this could be addressed is by fitting a mixed model that fits different variances by the levels of D - rinse order.

Aside from what is observable in relation to A - mixing order and D - rinse order, there is little that stands out. It is clear that there are distinguishable separations on the predicted axis; most notable for particle size, surface area, and less noticeably for pore volume, but this is expected from the separation of experimental points and the fact that there are generally only 2 or 3 main effects in each model. The most concerning feature of these plots is the wedge shape of the residuals for pore volume along the predicted axis. Part of this may be attributable to there being few predicted points less than 0.15, but concerning all the same. This could possibly be helped by a log transformation of the response.

The final diagnostic of interest is more of an investigation into a suspicion that the response variables are correlated. This does not have as much bearing on the validity of the individual models, but should be of interest to the experimenter. The plots are given in Figure 4.5.

Surface Area

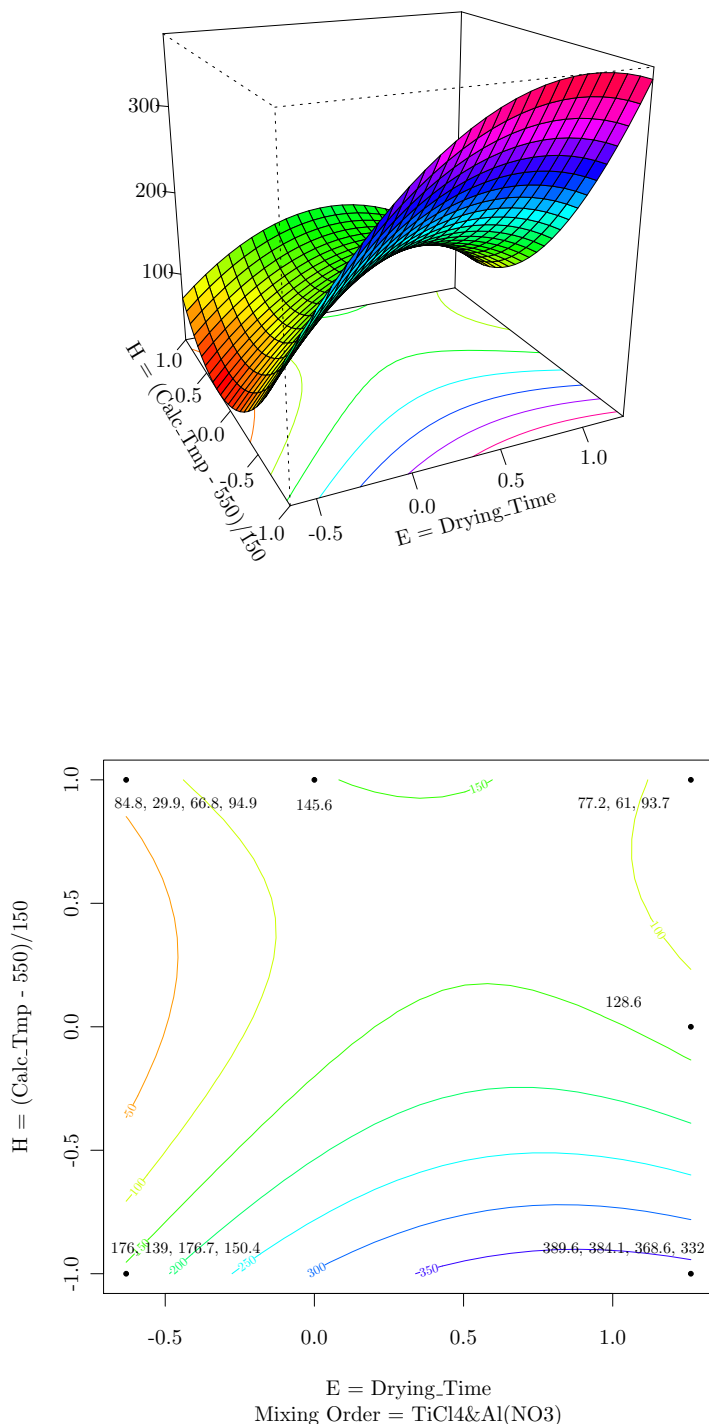


Figure 4.1: Diagnostic plots for the surface area response surface model. Drying time is coded as follows: $((\log(\text{Drying Time}) - \log(6)) / \log(3))$.

Pore Volume

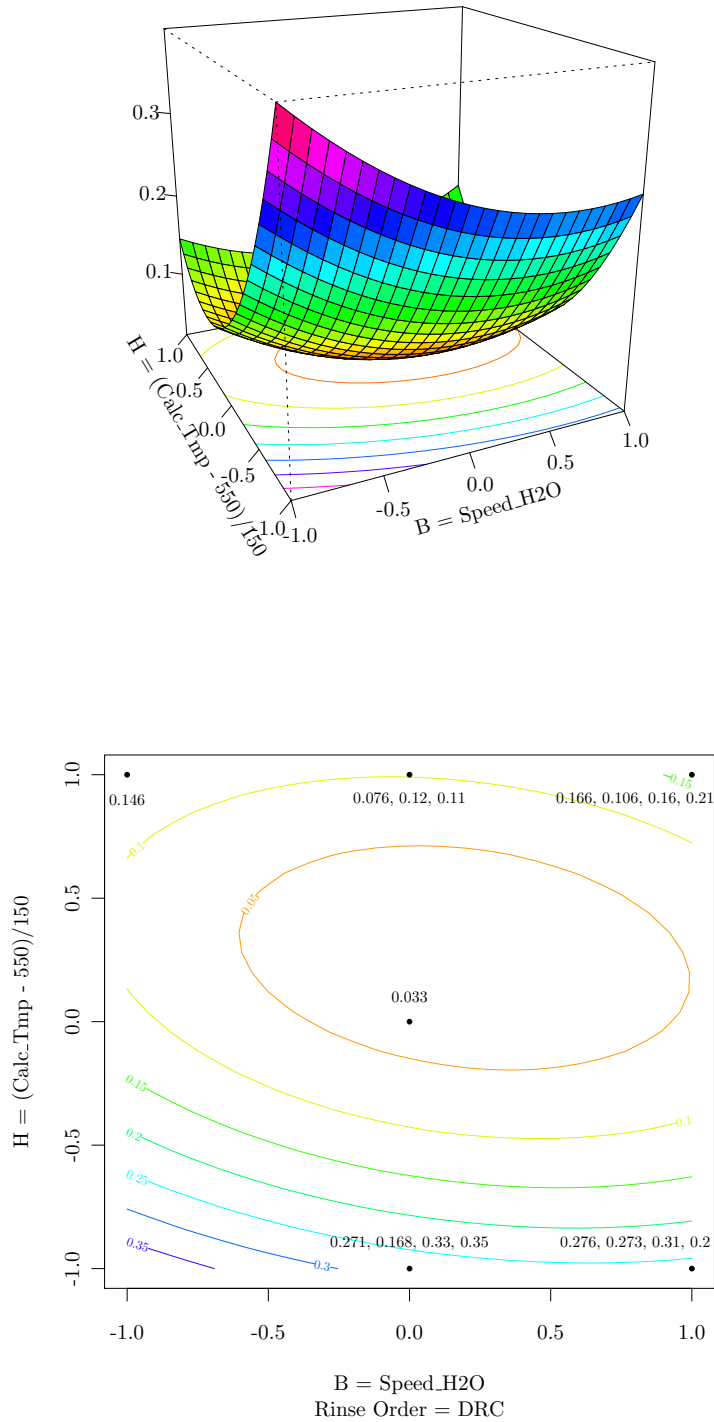


Figure 4.2: Diagnostic plot for the pore volume response surface model. Note that for B, -1 is for very slow, 0 is for slow, and 1 is for fast.

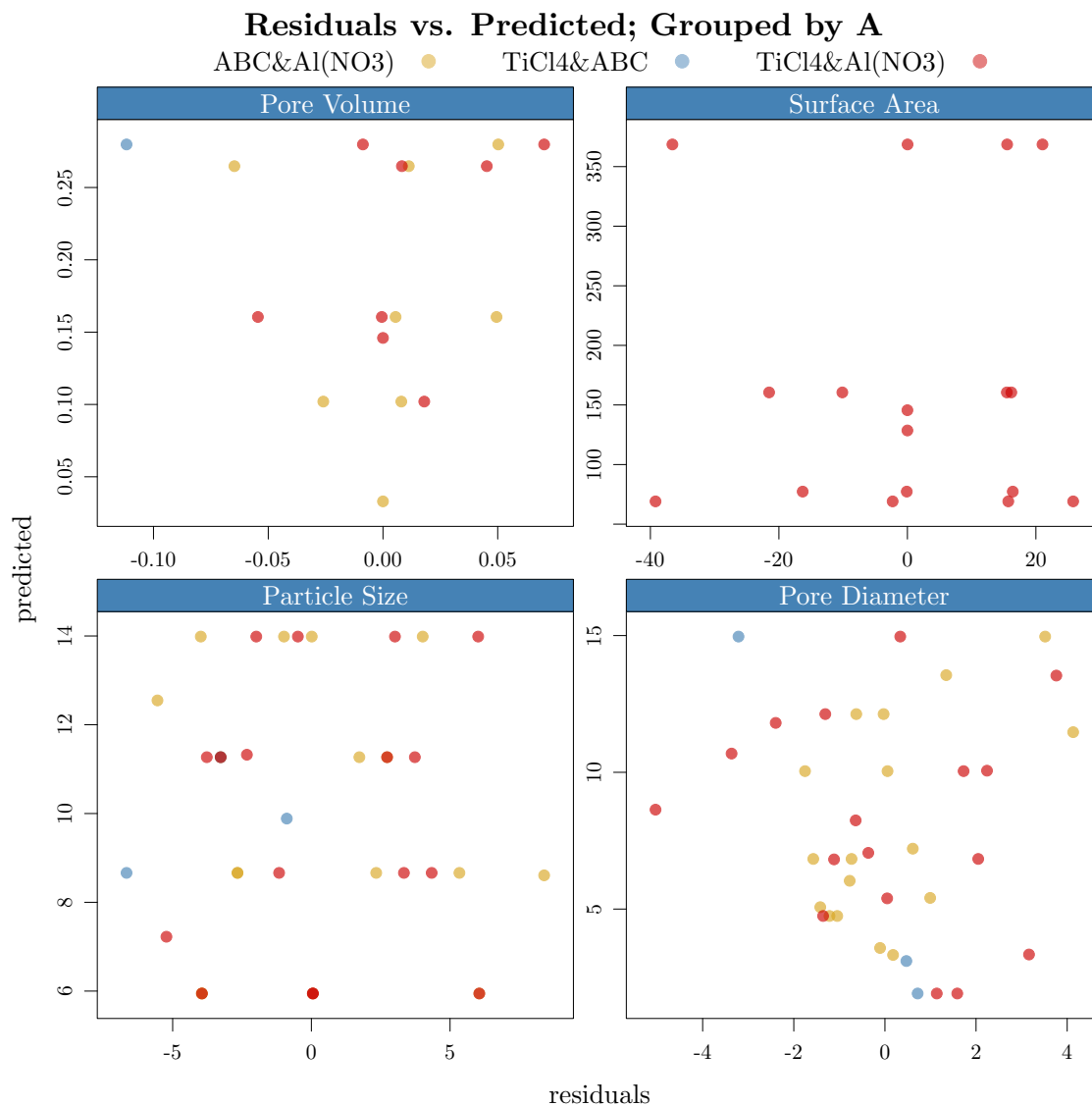


Figure 4.3: Predicted versus residual plots colored by A.

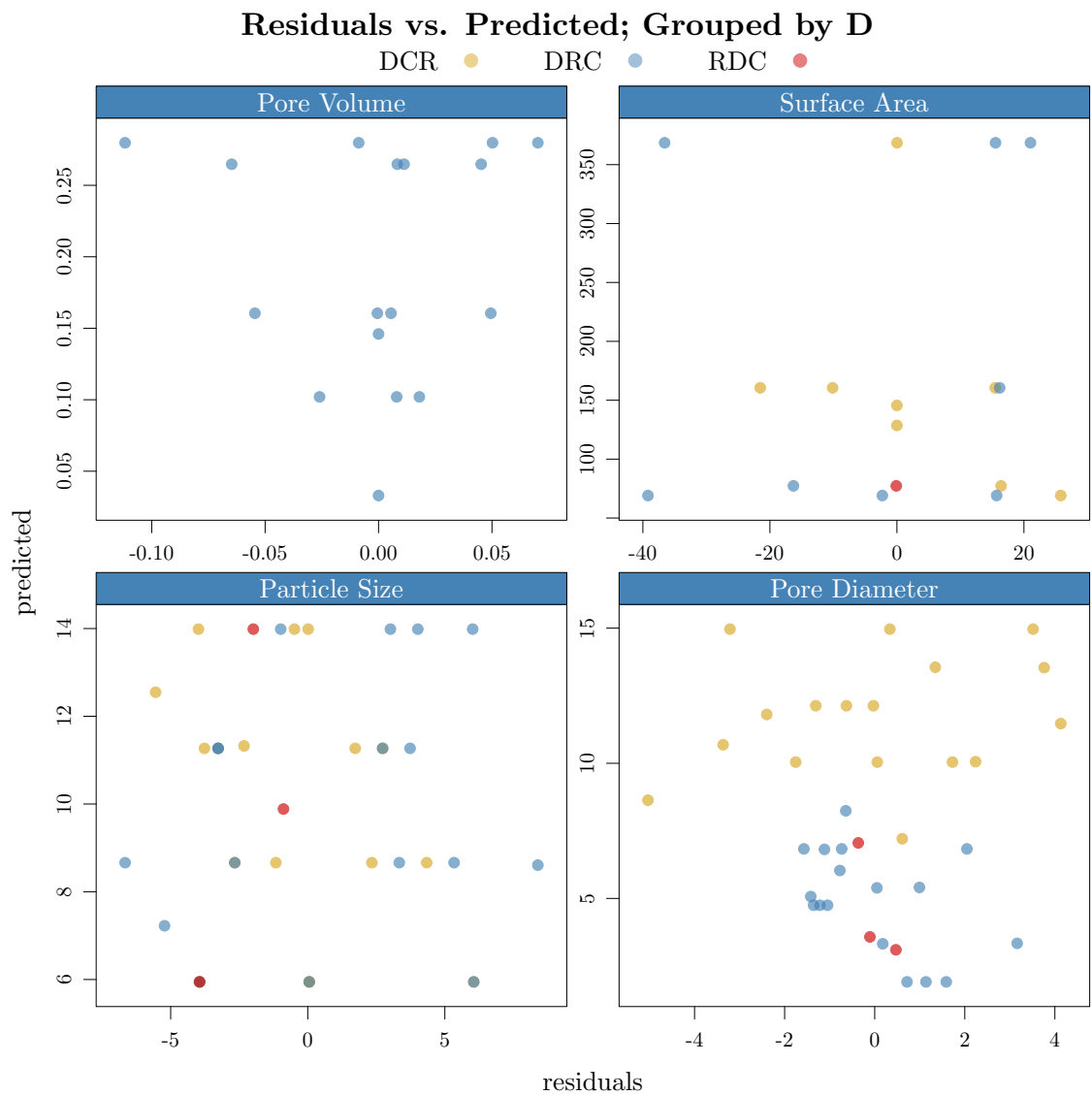
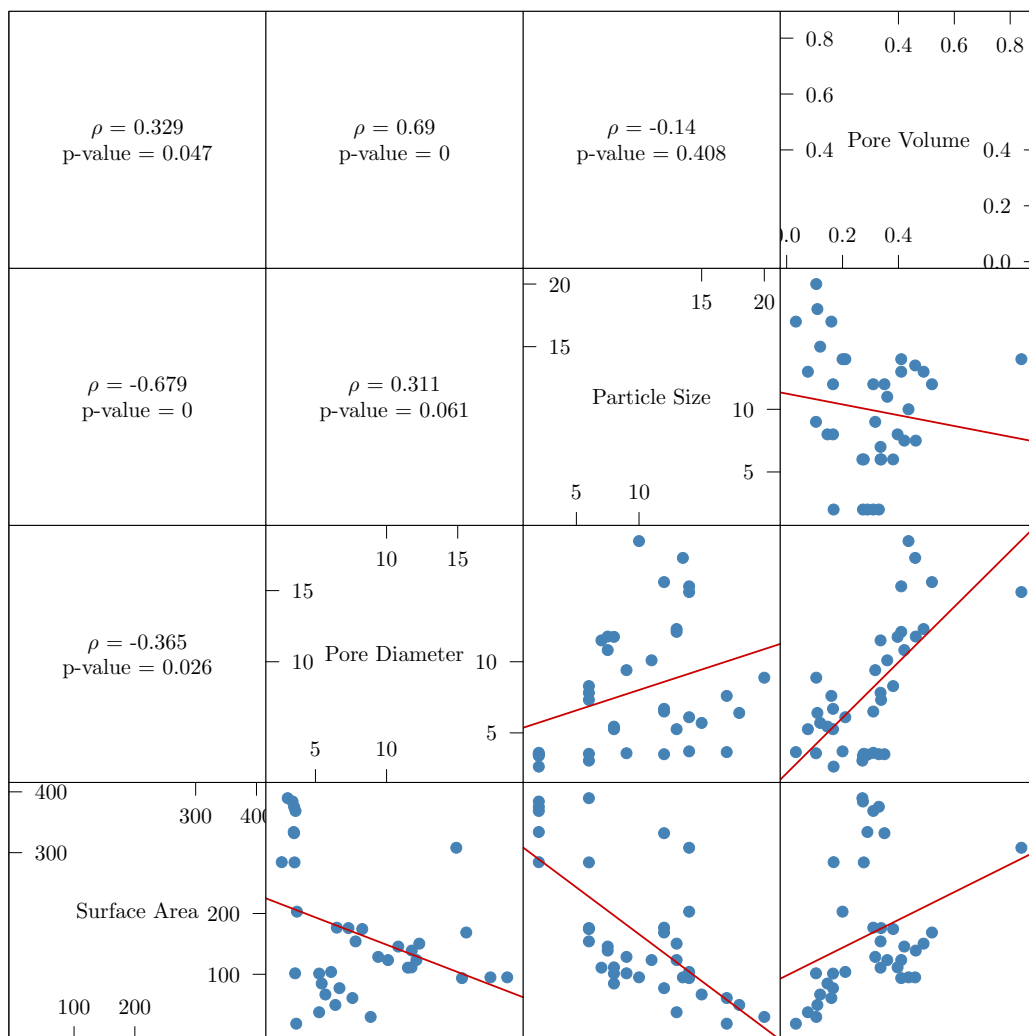


Figure 4.4: Predicted versus residual plots colored by D.

Correlations of Response Variables



Scatter Plot Matrix

Figure 4.5: Each response plotted against each other. Nothing too surprising here, although I think that the correlation between surface area and pore diameter is interesting.

4.3 CONCLUSIONS

For the experiment objectives of maximizing pore volume and surface area of the TiO_2 particles, and creating useful models to control the other two responses, seem to be met. After initially viewing the augmented data, I recall being overwhelmed at confronting the task of selecting the variables to include in the model. There seemed to be so much noise with regard to each response that I would have been happy to come up with one or two good models. In this sense I would attribute the success of the experiment to the variable selection methods used. The definitive screening design presented difficulties, especially when it became necessary to subset on A - mixing order and D - rinse order.

The last point made brings me to the subject of how I would have done things differently in retrospect. In this context I would say that the difficulties associated with the definitive screening design are more due to shortcomings in my planning the experiment than they are due to the design itself. That is, if I were to do it over again, I would not have varied A - mixing order and D - rinse order in the experiments themselves. I would have taken the 4 combinations of the high and low levels of these two factors, held them constant, and varied the other 8 factors. this would have resulted in $4(2^8) + 1 = 68$ experiments in addition to the 16 experiments for screening.

Were these 68 runs made without the initial screening design, we would have more solid response surface models. Additionally, we would have a response surface model for each combination of A - mixing order and D - rinse order, which we don't expect to behave the same anyway. There should be enough information in such an experiment to select which model would be the best candidate for maximizing the response, and if there were more important factors than we could fit a full response surface model to, then we could consider our first augmentation. In the end, I think that although 68 runs is quite a few, they provide a good return on investment. This introspection aside, I would say that my objective of

creating useful models for the experimenter in a small number of runs was met quite well in 37 experiments.

A relevant critical viewpoint that should be acknowledged is the bungled selection of factor levels. First, when I asked the experimenter to provide center points, I failed to catch that one of these was a new low point for the factor. This was missed due to a misunderstanding in naming the levels, but was certainly unfortunate. Additionally, I did not realize the importance of evenly spacing the factors, but this became apparent as I coded the factors. These are all easy to fix aspects of planning the experiment that I will not miss in the future.

From another critical viewpoint, the weakness of the proposed method is that there are more effects to be explored than there are experiments run. To be precise, there are 10 linear effects, 10 quadratic effects, and $\binom{10}{2} = 45$ 2-factor interactions totaling 65 terms as opposed to 37 completed experiments. This means that a regression model cannot simply be fit and important terms kept. This was a difficulty here where I combined a fractional factorial and a definitive screening design, so it will be a pervasive challenge for the use of any definitive screening design. In the literature review, methods were discussed which can be used to find a model in this situation. Two of these methods: the BVS algorithm (Chipman et al. (1997)) and Lawson's alias guided search method were employed, but were only simple in implementation due to the availability of software made freely available by Phil Woodward (Woodward (2012)) and SAS scripts from John Lawson.

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APPENDICES

APPENDIX A

RANDOMIZED RUNS

A=Mixing Order	B=Speed H2O Addition	C=Amount H2O	D=Order of Rinsing	E=Drying Time	F=Drying Temp	G=Calcination ramp	H=Calcination Temp	I=Calcination Time	J=Dopant amount
ABC and Al(NO3)	Fast	25 ml	DCR	3 hrs	100 °C	2 dg/min	700 °C	2 hrs	5%
ABC and Al(NO3)	Fast	7 ml	DCR	24 hrs	25 °C	20 dg/min	400 °C	2 hrs	22%
TiCl4 and AlNO3	Slow	7 ml	DRC	3 hrs	25 °C	20 dg/min	700 °C	20 hrs	22%
TiCl4 and AlNO3	Fast	7 ml	DRC	24 hrs	100 °C	20 dg/min	700 °C	2 hrs	5%
TiCl4 and AlNO3	Slow	7 ml	DCR	24 hrs	100 °C	2 dg/min	400 °C	2 hrs	22%
ABC and Al(NO3)	Slow	7 ml	DCR	3 hrs	100 °C	20 dg/min	400 °C	20 hrs	5%
TiCl4 and AlNO3	Fast	25 ml	DRC	3 hrs	25 °C	2 dg/min	400 °C	2 hrs	22%
TiCl4 and AlNO3	Fast	25 ml	DCR	24 hrs	100 °C	20 dg/min	700 °C	20 hrs	22%
ABC and Al(NO3)	Slow	7 ml	DRC	24 hrs	25 °C	2 dg/min	700 °C	2 hrs	5%
TiCl4 and AlNO3	Slow	25 ml	DCR	3 hrs	25 °C	20 dg/min	700 °C	2 hrs	5%
TiCl4 and AlNO3	Fast	7 ml	DCR	3 hrs	25 °C	2 dg/min	400 °C	20 hrs	5%
ABC and Al(NO3)	Fast	7 ml	DRC	3 hrs	100 °C	2 dg/min	700 °C	20 hrs	22%
ABC and Al(NO3)	Fast	25 ml	DRC	24 hrs	25 °C	20 dg/min	400 °C	20 hrs	5%
ABC and Al(NO3)	Slow	25 ml	DRC	3 hrs	100 °C	20 dg/min	400 °C	2 hrs	22%
TiCl4 and AlNO3	Slow	25 ml	DRC	24 hrs	100 °C	2 dg/min	400 °C	20 hrs	5%
ABC and Al(NO3)	Slow	25 ml	DCR	24 hrs	25 °C	2 dg/min	700 °C	20 hrs	22%

Table A.1: This is the randomization scheme used for the fractional factorial.

A=Mixing Order	B=Speed H2O Addition	C=Amount H2O	D=Order of Rinsing	E=Drying Time	F=Drying Temp	G=Calcination ramp	H=Calcination Temp	I=Calcination Time	J=Dopant amount
ABC and Al(NO3)	Slow	25 ml	DRC	3 hrs	100 °C	20 dg/min	550 °C	2 hrs	22%
TiCl4 and AlNO3	Slow	4 ml	DRC	24 hrs	100 °C	2 dg/min	400 °C	20 hrs	22%
ABC and Al(NO3)	Fast	4 ml	DCR	3 hrs	25 °C	20 dg/min	700 °C	2 hrs	5%
TiCl4 and AlNO3	Slow	7 ml	RDC	24 hrs	100 °C	20 dg/min	700 °C	2 hrs	5%
ABC and Al(NO3)	Slow	7 ml	DCR	3 hrs	100 °C	2 dg/min	700 °C	20 hrs	14%
TiCl4 and AlNO3	Very Slow	7 ml	DRC	3 hrs	25 °C	20 dg/min	700 °C	20 hrs	22%
TiCl4 and AlNO3	Fast	7 ml	DCR	3 hrs	100 °C	10 dg/min	400 °C	2 hrs	22%
ABC and Al(NO3)	Fast	7 ml	DRC	24 hrs	50 °C	2 dg/min	700 °C	2 hrs	22%
ABC and Al(NO3)	Slow	7 ml	DCR	24 hrs	25 °C	20 dg/min	400 °C	40 hrs	22%
TiCl4 and AlNO3	Fast	25 ml	DRC	3 hrs	100 °C	2 dg/min	700 °C	40 hrs	5%
TiCl4 and ABC	Fast	25 ml	DCR	24 hrs	100 °C	20 dg/min	700 °C	20 hrs	22%
ABC and Al(NO3)	Fast	7 ml	DRC	6 hrs	100 °C	20 dg/min	400 °C	20 hrs	5%
ABC and Al(NO3)	Very Slow	25 ml	DCR	24 hrs	100 °C	2 dg/min	400 °C	2 hrs	5%
TiCl4 and AlNO3	Fast	7 ml	DCR	24 hrs	25 °C	2 dg/min	550 °C	20 hrs	5%
TiCl4 and AlNO3	Slow	25 ml	DCR	6 hrs	25 °C	2 dg/min	700 °C	2 hrs	22%
ABC and Al(NO3)	Slow	25 ml	DRC	24 hrs	25 °C	10 dg/min	700 °C	20 hrs	5%
TiCl4 and AlNO3	Fast	25 ml	DRC	24 hrs	25 °C	20 dg/min	400 °C	2 hrs	14%
TiCl4 and AlNO3	Slow	25 ml	DCR	3 hrs	50 °C	20 dg/min	400 °C	20 hrs	5%
ABC and Al(NO3)	Fast	25 ml	RDC	3 hrs	25 °C	2 dg/min	400 °C	20 hrs	22%
TiCl4 and ABC	Very Slow	4 ml	RDC	6 hrs	50 °C	10 dg/min	550 °C	40 hrs	14%
TiCl4 and ABC	Slow	7 ml	DRC	3 hrs	25 °C	2 dg/min	400 °C	2 hrs	5%

Table A.2: This is the randomization scheme used for the definitive screening design.


```

LenthPlot(sA, alpha = .1, main = 'Surface Area', cex.lab = 1.5, cex.axis = 1.5, cex.main = 1.5)
LenthPlot(pD, alpha = .1, main = 'Pore Diameter', cex.lab = 1.5, cex.axis = 1.5, cex.main = 1.5)
LenthPlot(pV, alpha = .1, main = 'Pore Volume', cex.lab = 1.5, cex.axis = 1.5, cex.main = 1.5)
dev.off()

tikz(file = 'hn_plots.tex', standAlone = T, sanitize = F, width = 10, height = 10)
layout(mat=matrix(c(1,2,3,4),nrow = 2, ncol=2, byrow = TRUE), respect=T)
DanielPlot(pS, half = T, pch = 19, col = 'steelblue', datax = F, main = 'Particle Size',
  cex.lab = 1.5, cex.axis = 1.5, cex.main = 1.5)
DanielPlot(sA, half = T, pch = 19, col = 'steelblue', datax = F, main = 'Surface Area',
  cex.lab = 1.5, cex.axis = 1.5, cex.main = 1.5)
DanielPlot(pD, half = T, pch = 19, col = 'steelblue', datax = F, main = 'Pore Diameter',
  cex.lab = 1.5, cex.axis = 1.5, cex.main = 1.5)
DanielPlot(pV, half = T, pch = 19, col = 'steelblue', datax = F, main = 'Pore Volume',
  cex.lab = 1.5, cex.axis = 1.5, cex.main = 1.5)
dev.off()

# WWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWW
# Code for coding the factors and fitting the final models:
# WWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWW

setwd('~/Documents/masters_project/results')
library(rsm)

# WWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWW
# Prep the data
# WWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWWW

dat <- read.csv('../tio2Data/all_prepped.csv')
options(width=160)
names(dat) <- c('Mixing_Order', 'Speed_H2O', 'Amt_H2O', 'Rinse_Order', 'Drying_Time', 'Drying_Tmp',
  'Calc_Ramp', 'Calc_Tmp', 'Calc_Time', 'Dopant_Amt', 'Surface_Area', 'Pore_Diameter',
  'Particle_Size', 'Pore_Volume')

# prep the unevenly spaced factors:
dat$Amt_H2O <- (log(dat$Amt_H2O)-log(7))/log(7-4)
dat$Drying_Time <- (log(dat$Drying_Time)-log(6))/log(6-3)
dat$Drying_Tmp <- (log(dat$Drying_Tmp)-log(50))/log(50-25)

```



```

names(X2)[c(1,4)] <- c('A','D')

pSfin <- lm(Particle_Size ~ H + J, data=coded)
res3 <- residuals(pSfin)
pred3 <- predict(pSfin)
lab3 <- rep('Particle Size', length(res3))

pDfin <- lm(Pore_Diameter ~ Rinse_Order + B + H + G, x = T, data=coded)
res4 <- residuals(pDfin)
pred4 <- predict(pDfin)
lab4 <- rep('Pore Diameter', length(res4))
X4 <- coded[,c(1:10)]
names(X4)[c(1,4)] <- c('A','D')

# combine all of the data
all <- cbind(c(res1,res2,res3,res4),c(pred1,pred2,pred3,pred4),c(lab1,lab2,lab3,lab4),
  rbind(X1,X2,X4,X4))
names(all)[1:3] <- c('residuals','predicted','label')

# make diagnostic graphics (residual plots)
library(tikzDevice)
library(lattice)
tikz('resByA.tex',standAlone = T,sanitize = T)
xyplot(predicted~residuals|label,
  groups = A,
  scales = list(relation = 'free'),
  pch = 19,
  alpha = .65,
  col = c('goldenrod','steelblue','red3'),
  data = all,
  strip = strip.custom(bg = 'steelblue', par.strip.text = list(col = 'white'), horizontal = T),
  main = 'Residuals vs. Predicted; Grouped by A',
  key = list(space = 'top', columns = 3, text = list(levels(all$A)), points = list(alpha = .5,
    pch = 19, col = c('goldenrod','steelblue','red3'))))
)
dev.off()
tikz('resByD.tex', standAlone = T)
xyplot(predicted~residuals|label,
  groups = D,

```

```

scales = list(relation = 'free'), pch = 19, alpha = .65, col = c('goldenrod','steelblue','red3'),
data = all,
strip = strip.custom(bg = 'steelblue', par.strip.text = list(col = 'white'), horizontal = T),
main = 'Residuals vs. Predicted; Grouped by D',
key = list(space = 'top', columns = 3, text = list(levels(all$D)), points = list(alpha = .5,
  pch = 19, col = c('goldenrod','steelblue','red3')))
)
dev.off()

```

```

# check the correlation of 4 responses, make graphics:

```

```

library(grid)
tikz('splom.tex', standAlone = T)
splom(~coded[,c(11:14)],
  upper.panel = function(...){
    #print(list(...))
    grid.text(paste('$\rho$ = ', round(cor(list(...)$x,list(...)$y),3)),x = unit(.5,'npc'),
      y = unit(.55,'npc'), just = 'center',gp=gpar(cex = .8))
    grid.text(paste('p-value = ', round(cor.test(list(...)$x,list(...)$y)$p.value,3)),
      x = unit(.5,'npc'), y = unit(.45,'npc'), just = 'center',gp=gpar(cex = .8))
  },
  lower.panel = function(...){
    panel.xyplot(...)
    panel.lmline(x = list(...)$x,y = list(...)$y,col = 'red3',lwd=2)
  },
  pch = 19,
  col = 'steelblue',
  varnames = c('Surface Area', 'Pore Diameter', 'Particle Size', 'Pore Volume'),
  varname.cex = .8,
  main = 'Correlations of Response Variables',
)
dev.off()

```


APPENDIX C

SAS CODE

```
*This is an adaptation of code provided by Dr. Lawson for the alias matrix guided search;
*options notes source source2 nonumber nodate mprint symbolgen mtrace ps=55 ls=80;
*options nonotes nosource nosource2 nonumber nodate nomprint nosymbolgen nomtrace ps=55 ls=80;
*ods html close;
goptions ftext="Arial";

dm 'log;clear;output;clear;

*NOTE Here is the program for half normal plot and Alias Plot;
*Note Change the Variable Names Nobs, and Effect for which you want the Alias Plot;
%macro aliasplt;
data efpl6; set effpl2;
if etype=1 then P=sign(effect); else P=0;
keep P;
proc iml;
use alias; read all var _all_ into a;
use efpl6; read all var _all_ into p;
PRINT p;
zero={0};
p=zero//p;
ap=a*p;
ap=abs(ap);
create apl from ap[colname='Sum_Coef'];
append from ap;
quit;
data arep; merge lab1 alias;
proc print; Title 'Alias Report';
data aplot; merge lab1 apl;
row=_N_;
proc sort data=aplot; by descending Sum_Coef;
data aplot; set aplot; if _N_<=30;
run;
pattern1 value=solid color=black;
```

```

proc gchart data=aplot; vbar ILAB /descending sumvar=Sum_Coef ;
Title 'Alias Plot for Algebraically Summed Coefficients' ;
axis1 label=('Alias' justify=right 'Coefficient');
axis2 label=none length=6;
run;
%mend aliasplt;

%macro iter;
/*
*%put Enter a list of Interactions to be entertained: ;
*%input;
*/

%window getint color=white
#5 @5 'Enter a list of interactions:' attr=highlight color=blue
#7 @5 infield 80 attr=underline
#12 @29 'Press ENTER to continue.';
%display getint;

proc rsquare data=&dataf outest=cpl mse cp;
model Y=&maineff &infield /ADJRSQ RMSE CP SELECT=10 STOP=6 SIGMA=1.0;
proc Gplot; plot _MSE*_P_;
symbol1 color=black interpol=none value=dot;
Title 'MSE Plot';
run; quit;
proc Gplot; plot _CP*_P_;
symbol1 color=black interpol=none value=dot;
Title 'Cp Plot';
run; quit;
%mend iter;

%macro calc;
proc iml;
*Gets the 95th percentiles of the LGB statistic Rn;
no={8,
    9,
    12,
    16,

```

```

18,
21,
27,
32,
37};
rn95pct={1.534,
         1.385,
         1.291,
         1.201,
         1.178,
         1.178,
         1.115,
         1.099,
         1.099};

*Gets X-matrix and y-vector;
use x1; read all var _all_ into x1;
use x2; read all var _all_ into x2;
use Y;  read all var _all_ into Y;

*Calculates beta, absolute betas and ranks of absolute betas;
n=ncol(x1);
xpxi=inv(x1'*x1);
s=sqrt(diag(xpxi));
a=inv(s)*xpxi*(x1'*x2);
aprime=a';
xpy=x1'*Y;
rawb=xpxi*xpy;
rawb=rawb[2:n, ];
beta=inv(s)*xpxi*xpy;
abeta=abs(beta[2:n, ]);
beta=beta[2:n,1];
rbeta=rank(abeta);
lab=&vname2';

*Uses effect names in macro variable vname1;
eff=&vname1[ ,2:n]';

*Creates output files of calculated quantities;
create alias from aprime[colname=&vname1];
append from aprime;
create lab1 from lab[colname='ILAB'];
append from lab;

```

```

create eff from abeta[colname= 'abseff'];
append from abeta;
create efr from rbeta[colname='rank'];
append from rbeta;
create efl from eff[colname='ELAB'];
append from eff;
create efs from beta[colname='effect'];
append from beta;
create efu from rawb[colname='reffect'];
append from rawb;
create numobs from no[colname='numob'];
append from no;
create rnpct from rn95pct[colname='rn95pct'];
append from rn95pct;
quit;
*Merges file of LGB statistic percentiles;
data rnpctl1; merge numobs rnpct;
data effplt; merge efu efs eff efr efl;
zscore=probit(((rank-.5)/(&nobs-1)+1)/2); ID=1;
*Calculates inliers and outliers and keeps labels for largest effects;
proc univariate data=effplt noprint; var abseff; output out=effpl1 median=med;
data effpl1; set effpl1; ID=1;
data effpl2; merge effplt effpl1; by ID;
data effpl2; set effpl2;
s0=1.5*med;
if abseff>2.5*s0 then y=.; else y=abseff;
x=zscore; LAB2=ELAB;
if rank<(10/2) then ELAB=' '; * 10 is the number of main effects;
if ELAB=LAB2 then etype=1; else etype=0;
if _N_ <= &nest and abseff>2.5*s0 then etype=0;
keep ID x y reffect effect abseff rank zscore s0 ELAB LAB2 etype;
proc univariate data=effpl2 noprint; var y ; output out=pse median=pse;
data pse; set pse; pse=1.5*pse; ID=1;
data effpl2; merge effpl2 pse; by ID;
proc print;
run;
*Calculates the LGB statistic slopes and upper prediction limits;
proc reg data=effpl2 noprint outest=slope2;
model y=x/noint; output out=effpl3 u95=abseffcl p=abseffp;

```

```

proc reg data=effpl2 noprint outest=slope1;
model abseff=zscore/noint;
*Makes file for Half normal plot;
data effpl4; set effpl3; abseffec=abseff; code='abs(effects)'; output;
      abseffec=abseffp; code='line'; output;
      abseffec=abseffcl; code='Ulimit'; output;

proc sort; by code zscore;
*Determines significance of effects using LGB statistic;
data Rn; merge slope1 slope2; Rn=zscore/x; keep Rn;
data rnpctls; set rnpctls; if numob=&nobs; id2=1;
data effpl3; set effpl3; id2=1;
data Rn; set Rn; id2=1;
data effpl5; merge effpl3 Rn rnpctls; by id2;
sig='no '; if Rn>rn95pct and abseff>abseffcl then sig='yes';

%mend calc;
%macro printeff;
*Prints Results;
*proc print data=effpl5; *title effpl5;
data _null_; set effpl5 end=eof; file print header=H notitles;
if eof then goto F;
put @10 LAB2 @20 reffect best6. @30 effect best6. @42 abseff best6. @52 rank @62 sig 3.;
if _N_=&nobs-1 then goto S;
Return;
H: put / @20 'Standardized Effects Report'
      /
      /@30 'Standardized'
          / @10 'Label' @20 'Effect' @30 'Effect' @39 'Abs(Effect)' @51 'Rank' @60 'Sig(at 0.05)' /;
Return;
F: put / / @11 'Lawson, Grimshaw & Burt Rn Statistic=' @48 Rn best5.
      / @11 '95th percentile of Rn=' @34 rn95pct best5.;
Return;
S: put / / @11 'PSE=' pse best7.;
run;
%mend printeff;

%macro hlfnpplt;
data anno1; length function color $8;

```

```

retain xsys '2' ysys '2' hsys '1' when 'a';
set effpl4;
function='label'; position='3'; color='black'; text=ELAB; x=zscore; y=abseff; output;
proc gplot; plot abseffec*zscore=code/annotate=anno1 frame vaxis=axis1 haxis=axis2;
axis1 label=('Absolute' justify=left 'Effects');
axis2 label=('Half Normal Score');
symbol2 color=black interpol=none value=dot;
symbol3 color=black interpol=join value=none;
symbol1 color=black interpol=spline value=none line=2;
title 'Half Normal Plot of Effects';
run; quit;
%mend hlnfplt;

```

```

%let dataf=bloodg;
%let nobs=37;
%let nest=10;
/* these are the names for the effects in X1 */
%let vname1={'I' 'AL' 'BL' 'CL' 'DL' 'EL' 'FL' 'GL' 'HL' 'IL' 'JL'};
/* these are the names of the effects in X2 */
%let vname2={'ALBL' 'ALCL' 'ALDL' 'ALEL' 'ALFL' 'ALGL' 'ALHL' 'ALIL' 'ALJL'
            'BLCL' 'BLDL' 'BLEL' 'BLFL' 'BLGL' 'BLHL' 'BLIL' 'BLJL'
            'CLDL' 'CLEL' 'CLFL' 'CLGL' 'CLHL' 'CLIL' 'CLJL'
            'DLEL' 'DLFL' 'DLGL' 'DLHL' 'DLIL' 'DLJL'
            'ELFL' 'ELGL' 'ELHL' 'ELIL' 'ELJL'
            'FLGL' 'FLHL' 'FLIL' 'FLJL'
            'GLHL' 'GLIL' 'GLJL'
            'HLIL' 'HLJL'
            'ILJL'
            'AQ' 'BQ' 'CQ' 'DQ' 'EQ' 'FQ' 'GQ' 'HQ' 'IQ' 'JQ'};

```

```

%let maineff= AL BL CL DL EL FL GL HL IL JL;
data bloodg;
*Note Defines Constant;
I=1;
input A B C D E F G H I2 J Y;
*NOTE Defines Linear and Quadratic Main Effects;
if A =-1 then AL =-1/sqrt(2);
if A = 0 then AL = 0;
if A = 1 then AL = 1/sqrt(2);
if A =-1 then AQ = 1/sqrt(6);

```

```

if A = 0 then AQ = -2/sqrt(6);
if A = 1 then AQ = 1/sqrt(6);
if B = -1 then BL = -1/sqrt(2);
if B = 0 then BL = 0 ;
if B = 1 then BL = 1/sqrt(2);
if B = -1 then BQ = 1/sqrt(6);
if B = 0 then BQ = -2/sqrt(6);
if B = 1 then BQ = 1/sqrt(6);
if C = -1 then CL = -1/sqrt(2);
if C = 0 then CL = 0 ;
if C = 1 then CL = 1/sqrt(2) ;
if C = -1 then CQ = 1/sqrt(6);
if C = 0 then CQ = -2/sqrt(6);
if C = 1 then CQ = 1/sqrt(6);
if D = -1 then DL = -1/sqrt(2);
if D = 0 then DL = 0 ;
if D = 1 then DL = 1/sqrt(2) ;
if D = -1 then DQ = 1/sqrt(6);
if D = 0 then DQ = -2/sqrt(6);
if D = 1 then DQ = 1/sqrt(6);
if E = -1 then EL = -1/sqrt(2);
if E = 0 then EL = 0 ;
if E = 1 then EL = 1/sqrt(2) ;
if E = -1 then EQ = 1/sqrt(6);
if E = 0 then EQ = -2/sqrt(6);
if E = 1 then EQ = 1/sqrt(6);
if F = -1 then FL = -1/sqrt(2);
if F = 0 then FL = 0 ;
if F = 1 then FL = 1/sqrt(2) ;
if F = -1 then FQ = 1/sqrt(6);
if F = 0 then FQ = -2/sqrt(6);
if F = 1 then FQ = 1/sqrt(6);
if G = -1 then GL = -1/sqrt(2);
if G = 0 then GL = 0 ;
if G = 1 then GL = 1/sqrt(2) ;
if G = -1 then GQ = 1/sqrt(6);
if G = 0 then GQ = -2/sqrt(6);
if G = 1 then GQ = 1/sqrt(6);
if H = -1 then HL = -1/sqrt(2);

```

```

if H = 0 then HL = 0 ;
if H = 1 then HL = 1/sqrt(2) ;
if H = -1 then HQ = 1/sqrt(6);
if H = 0 then HQ = -2/sqrt(6);
if H = 1 then HQ = 1/sqrt(6);
if I2 = -1 then IL = -1/sqrt(2);
if I2 = 0 then IL = 0 ;
if I2 = 1 then IL = 1/sqrt(2) ;
if I2 = -1 then IQ = 1/sqrt(6);
if I2 = 0 then IQ = -2/sqrt(6);
if I2 = 1 then IQ = 1/sqrt(6);
if J = -1 then JL = -1/sqrt(2);
if J = 0 then JL = 0 ;
if J = 1 then JL = 1/sqrt(2) ;
if J = -1 then JQ = 1/sqrt(6);
if J = 0 then JQ = -2/sqrt(6);
if J = 1 then JQ = 1/sqrt(6);
*Note Defines Interactions to Make X1 Full Rank;
*NOTE Defines Linear by Linear Interaction Effects;
ALBL=AL*BL;
ALCL=AL*CL; ALDL=AL*DL; ALEL=AL*EL; ALFL=AL*FL; ALGL=AL*GL; ALHL=AL*HL; ALIL=AL*IL; ALJL=AL*JL;
BLCL=BL*CL; BLDL=BL*DL; BLEL=BL*EL; BLFL=BL*FL; BLGL=BL*GL; BLHL=BL*HL; BLIL=BL*IL; BLJL=BL*JL;
CLDL=CL*DL; CLEL=CL*EL; CLFL=CL*FL; CLGL=CL*GL; CLHL=CL*HL; CLIL=CL*IL; CLJL=CL*JL;
DLEL=DL*EL; DLFL=DL*FL; DLGL=DL*GL; DLHL=DL*HL; DLIL=DL*IL; DLJL=DL*JL;
ELFL=EL*FL; ELGL=EL*GL; ELHL=EL*HL; ELIL=EL*IL; ELJL=EL*JL;
FLGL=FL*GL; FLHL=FL*HL; FLIL=FL*IL; FLJL=FL*JL;
GLHL=GL*HL; GLIL=GL*IL; GLJL=GL*JL;
HLIL=HL*IL; HLJL=HL*JL;
ILJL=IL*JL;

cards;

*Input the datalines:

*For some reason, SAS won't accept the data from here as it normally would, but just put it in the;
*prompt from another file and it runs;
*NOTE Defines Estimable and Inestimable Contrasts and Response;
data y; set bloodg; keep Y;
data x1; set bloodg; keep I AL BL CL DL EL FL GL HL IL JL;

```



```
data x2; set bloodg; keep ALBL ALCL ALDL ALEL ALFL ALGL ALHL ALIL ALJL
      BLCL BLDL BLEL BLFL BLGL BLHL BLIL BLJL
      CLDL CLEL CLFL CLGL CLHL CLIL CLJL
      DLEL DFLD DLGL DLHL DLIL DLJL
      ELFL ELGL ELHL ELIL ELJL
      FLGL FLHL FLIL FLJL
      GLHL GLIL GLJL
      HLIL HLJL
      ILJL
AQ BQ CQ DQ EQ FQ GQ HQ IQ JQ;
```

```
%calc
%printeff
%hlfnp1t
%aliasplt
%iter
```

BUGSXL A DOCUMENTATION

This appendix documents the use of BugsXLA in this project. The authoritative source on this process is Spiegelhalter et al. (1999), and provides information on where to obtain WinBUGS and BugsXLA with installation instructions. Assuming that WinBUGS is successfully installed on your system, to proceed, one must download the Excel add-in for BugsXLA from <http://www.philwoodward.co.uk/bugsxla/download.html>.

To start the process, I import the experimental data into Excel. Once the data is imported, I select the add-ins tab in excell where an icon for BugsXLA appears in custom toolbars. Once the icon is selected, a form, 'Bayesian Model Specification,' appears as illustrated in Figure D.1. Select the data range field in the form. Then I select the data you want used by dragging your mouse from the upper left corner of the data to the bottom right. Next, I select the 'Response' field in the form. I enter the name of the response variable identically to how it shows in top of its column in the Excel table. I then select the 'VS Factors' field and name my varied factors. I enter $(A*B*C*D*E*F*G*H*I)@2$ to indicate that I want all of the variables named in a second order model. This will include all main effects and linear by linear interactions in the Bayesian variable selection process.

Before I can press 'OK' for the form, I press the 'Set Variable Types' button near the data range field in the form. A box as pictured in Figure D.2 appears. I move all of my factors to the 'Factors' box and leave the response variable in the 'Variates' box. I highlight all of my factors and press 'Edit Factor Levels'. I then cycle through each factor and ensure that the factor is ordered -1, 0, 1. I then hit 'OK' in the create factors dialogue box and then I hit 'OK' in the 'Bayesian Model Specification' dialogue box.

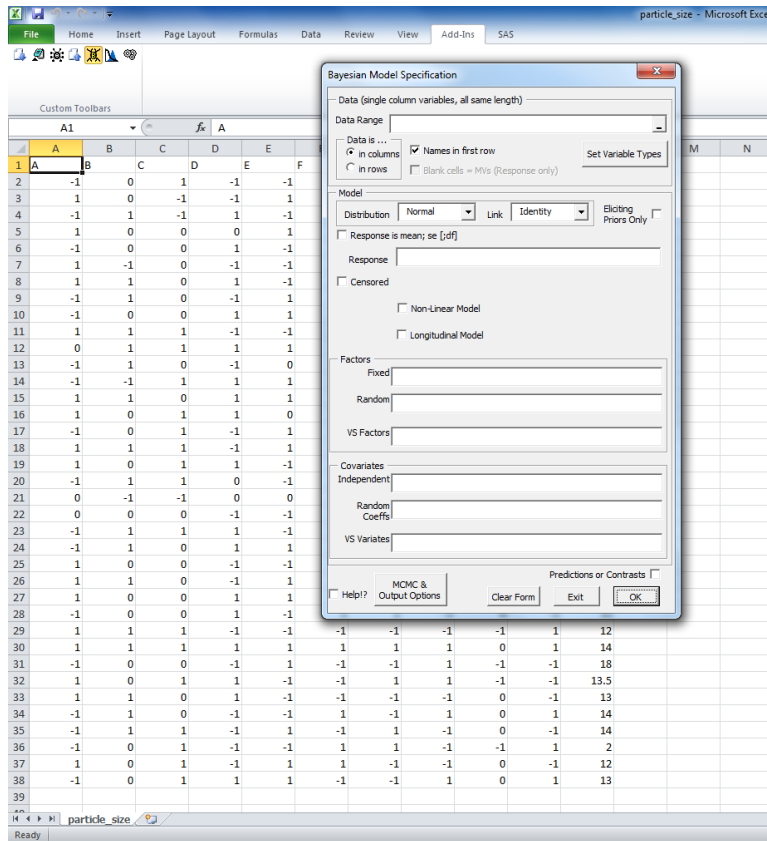


Figure D.1: Initial form.

Now a ‘Prior Distributions’ dialog box appears. I leave everything default and navigate to the ‘VS Terms’ tab in the dialog box. In the bottom left of this box I uncheck strong heredity. This adjusts the priors as discussed in the literature review in order to take weak heredity into consideration. This was done for each response variable using the priors $p_{11} = 1$, $p_{10} = 0.5$, $p_{00} = 0.1$. The final state of the dialog box is shown in Figure D.3. Once ‘Run WinBUGS’ is executed, the algorithm is run.

Once the algorithm finishes, results are imported from WinBUGS into Excel. Among these results are the posterior distributions for each effect. These were illustrated in Figure 3.8.

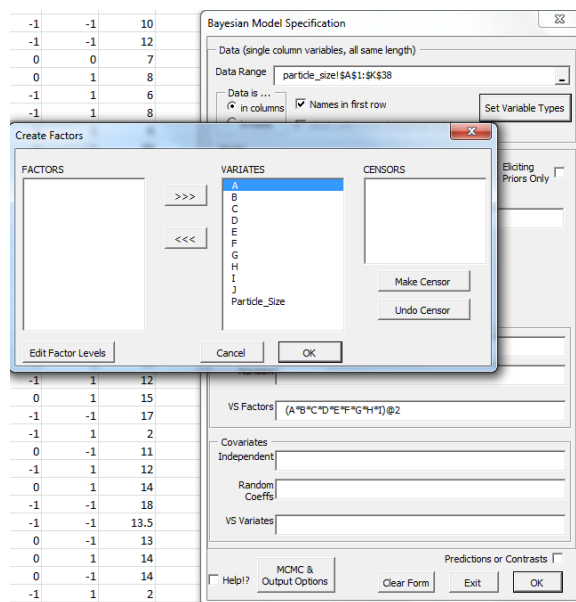


Figure D.2: This box is where I ensure correct variable coding.

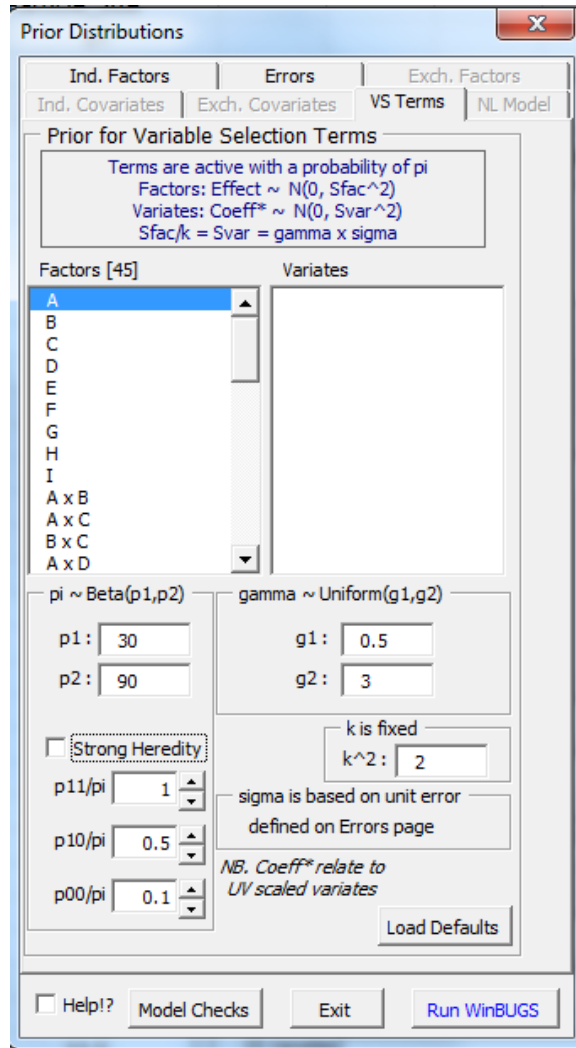


Figure D.3: This is where priors for weak heredity are specified.