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INVESTIGATION OF MIXED OXIDE ($U_{.75} Pu_{.25}$) $_2$ PROCESSING
USING A NINE VARIABLE FRACTIONAL FACTORIAL EXPERIMENT

By

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ABSTRACT

A process development program was undertaken to support production of high quality mixed oxide (25% PuO_2 -75% UO_2) fast reactor fuel pellets. In addition to an evaluation of variables associated with each unit operation, a fractional factorial experiment was designed to show variable effects and interactions that occur in the process as a whole. This report discusses the experiment involving nine key process variables including: calcination temperature of PuO_2 , preslugging pressure, pressing pressure, binder type, binder percentage, percent addition of sintered recycle powder, particle size of sintered recycle powder, sintering time, and sintering temperature. The measured dependent variables were green density, sintered density, sorbed gas, percent shrinkage and uniformity of diameter and sintered density.

INTRODUCTION

Large numbers of consistent high quality mixed oxide fuel pellets will be needed to meet the core requirements of the Fast Test Reactor. Ceramic fuel pellets can be produced using several basically different fabrication processes, i.e., dry press, extrusion, hot press and slip cast. Each basic process has its own sequence of unit operations and corresponding key process variables. The dry-press process was selected as the reference FTR pellet fabrication process. This decision was based on 1) industrial experience and familiarity with the process, 2) the inherent capability of this process to produce a quality fuel pellet from the raw materials available, and 3) the relatively high level of process and end product control that can be obtained. The reference raw materials will be oxalate source PuO_2 mechanically blended with ceramic grade UO_2 . The initial raw material source and blending technique were selected on the basis of the quality and quantity of PuO_2 expected to be available when production quantities of oxide are needed, and the extent of industrial experience with mechanical blending of ceramic materials. The reference materials and process will be used as a base from which to develop raw material specifications, fuel pellet specifications, and quality assurance procedures.

SUMMARY

The major effects and interactions between nine key process variables were evaluated in a fractional factorial experiment. Sintered pellet properties such as density, density variation, percent diameter shrinkage, and off-gas quantity are expressed by equations and response curves. Relationships between seven process variables and green pellet density are also expressed by equations and derived response curves. The factorial approach to process development has supplied data indicating the relative importance of process variables within a complete fabrication process.

Data obtained indicates the following:

- Variables having a significant effect on green density include press pressure, binder type, calcination temperature of PuO_2 , preslugging pressure and percent addition of sintered recycle powder.
Variables having a significant affect on sintered density include binder percentage, binder type, preslugging pressure, sintering temperature and percent addition of sintered recycle powder.
- The particle size of sintered recycle powder within the sizes evaluated had a small but sianificant effect on sintered pellet density
- Variables affecting diametral variation between sintered pellets include pressing pressure, binder percentage, preslugging pressure and percent addition of sintered recycle powder

TECHNICAL APPROACH

All ceramic processes involve a series of unit operations. Each unit operation has several associated variables and could be extensively studied by itself. This fractional factorial design does not attempt to optimize or simplify any operation; however, the variables selected were those which would affect one or more pellet characteristics. Optimization of individual unit operations and evaluation of variables not included in this experiment will be accomplished in small complete factorials or additional fractional factorials as appropriate.

This fractional factorial experiment evaluated the direct effects of, curvilinearity, and many interactions between nine key process and material variables. The factorial was designed to show relationships and relative importance of variables when individual unit operations are interconnected into a manufacturing process.

The variable limits were selected after evaluating the results of some preliminary process experiments. These small experiments involved raw materials and processing techniques similar to those used in the fractional factorial. The initial blending procedure used during formation of process groups within the factorial was designed to promote good homogeneity with a minimum of milling. Extensive milling would affect the PuO_2 and sintered recycle particulate properties, thus confusing the analysis of results. The lack of milling lowered the maximum sintered density from 94% to about 90% of theoretical.

The nine variables considered in the fractional factorial and a brief discussion of each follows.

1, Raw Material Characteristics as Determined by the Changes in Surface Area of PuO_2

To obtain three different starting material conditions the calcine temperature of oxalate precipitated PuO_2 was varied. The three levels selected were:

- a. As-received PuO_2 - calcined at 425°C by the supplier, surface area of 38 meters²/gm
- b. 700°C calcined PuO_2 from same lot as above, surface area of 9.2 meters²/gm
- c. 1000°C calcined PuO_2 from same lot as (a), surface area of 1.4 meters²/gm

By varying the calcine temperature, the surface activity and impurity levels were also changed. It is impossible to change an individual particle characteristic without affecting others; therefore, although only one or two particle parameters are determined, the change in those few parameters is representative of a change in the overall material condition. For the purposes of this experiment, the UO_2 source was constant.

2. Press Pressure or Change in Green Density

To determine the effect of changes in press pressure on shrinkage and density, three pressure levels were used, all within the range where the greatest effect of pressure change should be noticed in the percent shrinkage. The low pressure level was selected close to the area where pressure changes affect density. The center and upper pressure should also be safely below the point at which laminations occur. It is not intended to determine the maximum feasible pressing pressure.

3. Sintering Temperature

Temperature is one of the more important variables considered in this experiment. The ultimate density and, to a lesser degree, grain size and pore structure of the pellet is strongly influenced by the sintering temperature. In this experiment three temperatures were used, separated sufficiently to bracket the optimum sintering temperature for all powder conditions used in this study,

4. Sintering Time

Time and temperature must both be considered in the same experiment, as they are in effect inseparable. The time at temperature will influence grain growth and to a lesser degree density. In this experiment three sintering times were used. The largest effect of a time difference should be observed at the lower temperatures. Long sintering times at the highest temperature (more of an academic value than practical), were included to determine the nature (linear or curvilinear) of the relationship.

5. Binder Percentage

An effective method of controlling density is by the addition of a "pore forming" binder. Assuming reasonable control over raw materials and processing, changes in density should be directly related to the percent addition of the "pore former". An additional consideration is that a lubricating material of some sort is a useful aid when pressing a powder into pellet form. Pressing pressures must be distributed throughout the pellet to aid green strength and to minimize laminations, cracks, and diameter taper. The binders evaluated were added in three evenly spaced percentages, and the effect on sintered pellet density and taper was observed. Binder removal or burnout was accomplished during the sintering operation through the use of a flowing atmosphere.

6. Binder Type

Two binders were used for this variable as opposed to three levels of all other variables. The binders (methyl-cellulose and Carbowax) were selected on the basis of their wide industrial use, availability, and the results of some preliminary experiments. Methyl-cellulose is considered a pore former and also improves the green strength of pellets. Carbowax is generally not considered a pore former and therefore has little effect on density when used in percentages below about 4 weight percent. Carbowax, being highly water soluble, can easily be uniformly dispersed throughout a powder permitting the more equitable distribution of pressure.

7. Preslug Pressure

A technique potentially useful in producing porosity in sintered pellets is to preslug (prepress) a powder, granulate, and then press to a lower pressure than was used to preslug. The reproducibility and actual effect of this technique are not quantitatively known. Two preslug pressures have been selected with the higher pressure being greater than the highest pressing pressure. To determine the effect of pressing without preslugging, a zero preslug pressure was included.

8. Addition of Sintered Recycle Material

In any ceramic process many rejects will occur for minor reasons such as chips, laminations, or improper grinding. Some of this material can be granulated and recycled into new batches of powder with no detrimental effect on pellet quality. The amount of sintered material suitable for recycle is expected to be less than 15% to 20%. To determine the effect of recycle material on sinterability, microstructure, and shrinkage of fuel pellets; granulated and screened recycle material was added in two percentage levels, 7.53 and 15%.

9. Particle Size of Recycle Material

The particle sizes used will be determined by screen separation and will be much coarser than the base raw material. This was done for two reasons:-

- a. Accurately defining, separating, and **reproducing** fractions of ground powder with an average particle size less than 44 microns (-325 mesh) is difficult and equipment capable of doing this (air classifiers) were not available.
- b. There is no information available specifically stating that recycle powders must be the same as, or close to, the particle size of starting raw materials. (In this case, approximately 0.1 to 10 microns.) The finest particle size used in this experiment is classified only as -325 mesh.

The fractional factorial test design showing effects and interactions is shown in Table 1. Specific variables and their limits are shown in Table 2. Data obtained from the fractional factorial experiment are presented in the results section.

TABLE 1
EFFECTS WHICH CAN BE ESTIMATED IN THE
FRACTIONAL FACTORIAL EXPERIMENT

<u>Effects</u>	<u>Variables</u>								
Main (Linear)	x_1	x_2	x_3	x_4	x_5	x_6	x_7	x_8	x_9
Quadratic	x_1^2	x_2^2	x_3^2	x_4^2	x_5^2	x_6^2	x_7^2	x_8^2	x_9^2
Interactions	x_1x_2	x_1x_3	x_1x_4	x_1x_5	x_1x_6		x_1x_8	x_1x_9	
		x_2x_3	x_2x_4	x_2x_5	x_2x_6	x_2x_7		x_2x_9	
			x_3x_4	x_3x_5	x_3x_6		x_3x_8	x_3x_9	
				x_4x_5	x_4x_6			x_4x_9	
					x_5x_6			x_5x_9	
						x_6x_7		x_6x_9	
								x_8x_9	

x_1 Surface Area (PuO_2)

x_2 Green Density (Press Pressure)

x_3 Sintering Temperature

x_4 Sintering Time

x_5 Binder Percentage

x_6 Preslug Pressure

x_7 Binder Types (2)

x_8 Particle Size of Recycle

x_9 Percent Recycle Additive

When x_9 is 0, x_8 is meaningless

TABLE 2
LIMITS FOR FRACTIONAL FACTORIAL

<u>Variable</u>	<u>Description</u>	<u>Code</u>	<u>Limits</u>
x_1	Surface Area Vary calcination of PuO_2	-1 = As-received PuO_2 0 = 700° calcine PuO_2 1 = 1000° calcine PuO_2	--- 38 meters ² /g --- 9.2 meters ² /g --- 1.4 meters ² /g
x_2	Green Density Vary Press Pressure	-1 = 15,000 psi 0 = 27,500 psi 1 = 40,000 psi	
x_3	Sintering Temp.	-1 = 1525°C 0 = 1590°C 1 = 1660°C	
x_4	Sintering Time	-1 = 2 hours 0 = 5 hours 1 = 8 hours	
x_5	Binder Percent	-1 = 2 Wt% 0 = 3 1/4 Wt% 1 = 4 1/2 Wt%	
x_6	Preslug Pressure	-1 = None 0 = 25,000 psi 1 = 50,000 psi	
x_7	Binder Type	-1 = Carbowax 1 = Methyl Cellulose	
x_8	Particle Size of Recycle Material	-1 = -325 Mesh - 44 Micron 0 = -200 +325 Mesh 74 to 44 Micron 1 = -100 +200 Mesh 147 to 74 Micron	
x_9	Percent Addition of Recycle Material	-1 = None added 0 = 7 1/2% 1 = 15%	

DISCUSSION OF RESULTS

A. Powder Characterization and Processing History

The raw materials used in this experiment were Eldorado normal UO_2 and oxalate source PuO_2 that had been calcined to various temperatures. The UO_2 was wet slurry ball milled for four hours before blending with PuO_2 . Some impurity from the milling media in the form of tungsten carbide was introduced into the powder; however, the effect on curves or data presented in this document is considered negligible.

The surface area of the UO_2 powder after milling was $9.3 \text{ m}^2/\text{g}$ with particle size and distribution of the milled powder shown in Figure 1. The asymmetric curve is typical of curves obtained from material containing a large number of particles smaller than an instrument is capable of measuring. A scanning electron micrograph (Figure 2) shows the UO_2 "particles" to actually be agglomerates of extremely fine submicron crystallites.

Particle size and distribution curves of oxalate source PuO_2 after calcination to 450, 700, and 1000°C are shown in Figure 3. The curves in Figure 3 would indicate that calcination temperature did not affect particle size or distribution to a significant degree. A scanning electron micrograph of oxalate source PuO_2 calcined to 450°C is shown in Figure 4. The scanning micrograph confirms the particle size data and shows the particles to be relatively firm agglomerates of angular platelets or crystallites.

The fractional factorial was composed of 82 pellet groups with the PuO_2 type, binder type, binder percent and pressing pressure controlled by the experimental design. The UO_2 and PuO_2 powders were initially blended in a twin shell blender and further blended in a rubber lined ball mill (wet slurry) containing five 3/4 inch diameter tungsten carbide cylinders. As binder type and quantity were variables, binders were added in a separate

operation. All pellets were pressed by hand with the die being wiped with Butyl-Stearate lubricant to aid ejection. Sintering was accomplished in an Al_2O_3 tube furnace in flowing argon-8% hydrogen atmosphere.

An example of pellet homogeneity produced by the aforementioned mixing process may be seen in Figures 5 and 6. Figure 5 is an alpha-autoradiograph of another pellet from the same pellet group. In both autoradiographs PuO_2 rich areas and artifacts are dark with PuO_2 deficient areas and voids or pores appearing light. It should be noted that pellet group 56 contained 7.5% of -200 +325 mesh sintered recycle material.

FIGURE 1. WET BALL MILLED 4 HOURS AND DISPERSED ULTRASONICALLY FOR 2 HOURS USING SODIUM- LIGNIN SULFONATE

FIGURE 2. MILLED ELDORADO UO_2

S-684

FIGURE 3 . CALCINED OXALATE SOURCE PuO_2 DISPERSED FOR 15 MINUTES USING SODIUM LIGMIN SULFONATE

FIGURE 4. OXALATE SOURCE PuO_2 CALCINED TO 450°C

#1370

GREEN PELLETS
10X ALPHA-AUTORADIOGRAPH
30 SECOND EXPOSURE
#8278A

FIGURE 5. PELLET GROUP 56

SINTERED PELLET
15X ALPHA-AUTORADIOGRAPH
#4953A

FIGURE 6- PELLET GROUP 56

E. Green Pellet Data

1. Experimental Design

The 82 pellet groups comprising the entire fractional factorial design were divided into three parts. The principle portion consisted of 62 groups which involved the variables X_1 , X_2 , X_3 , X_4 , X_5 , X_6 and X_7 , each at three levels, and X_8 at two levels. The variable X_8 for these 62 groups was either undefined (which occurs when X_9 takes on its -1 value) or else was held fixed at the 3 level. The 62 groups consisted of a fractional replication of a 3^7 design blocked on X_7 - the binder type; i.e., half of the groups contained the -1 level of X_7 and half the +1 level.

Since this particular design does not permit estimation of interactions with X_7 (binder type) the 62 groups were supplemented by 8 additional groups. These 8 groups were a simple 2^3 design involving X_6 , X_7 , and X_8 , as it was felt that if interactions with X_7 existed, they would involve the two variables X_6 (press pressure) and X_8 (preslun pressure).

The final 12 groups in the experimental design were used to estimate the effects of X_9 - the particle size of the recycle material. These 12 groups included the variables X_1 , X_3 , and X_9 and consisted of a half replicate of a 3×2^3 experiment. For these groups, X_1 , X_3 , and X_9 were each investigated at two levels rather than three.

2. Analysis of Green Pellet Data

The data submitted for analysis consisted of measurements made on the green pellets. For each of the 82 pellet groups, about 12 pellets were measured. Measurements submitted for analysis included diameter, length, weight and percent theoretical density for each pellet.

The first step in the analysis consisted of editing the raw data to detect errors. This was accomplished by keypunching the data and processing them in the computer. Calculations performed on the computer consisted of a recomputation of the percent density based on the given diameter, length, and weight. percent density was computed as being 0.7 times the weight divided by the diameter squared times the length. The other function of the editing task was to detect any outliers in the data, that is, to pick out those one or two pellets in a group which gave results inconsistent with the remaining pellets. Several such pellets were detected, but in no case were there more than two pellets in a given group. Such data were excluded from the analysis and the average theoretical density was recomputed based on the remaining data.

The dependent variable of primary interest is the average percent theoretical density which will be expressed as a function of the process variables. Each average is based on 10 to 12 pellets.

The model employed in the first attempt at estimation was the full quadratic model. This was based on pellet groups 1 through 62 and 75 through 82. i.e., it excluded those 12 pellet groups in which x_8 , the particle size of recycle, was varied. Therefore, the first model does not have x_8 appearing as a variable. It should be kept in mind that when referring to x_9 at its -1 level (no recycle), x_8 as a variable is undefined. When x_9 takes on the value 0 or 1, x_8 is defined to take on its 0 value of -200 +325 mesh size. Variables x_3 and x_4 being sintering variables do not appear in this model.

The full quadratic model consists of 24 terms. This includes the overall average, 7 linear effects, 5 quadratic effects, and 12 two factor interactions which are estimable from these data. The first attempt at fitting the data with the full model indicated that most interactions could be deleted from the model. The dominant effects are the main effects of each of the variables involved. However, two significant interactions were retained for the next estimation step. Both of these interactions included X_2 (press pressure) as one of the variables. Specifically interaction terms retained in the model were X_2X_6 , and X_2X_9 .

Upon deleting the remaining interaction terms from the model and re-estimating the parameters the following equation resulted.

$$Y = 52.525 + 1.644X_1 + 2.768X_2 - 0.061X_5 + 1.557X_6 - 2.113X_7 + 1.255X_9 \\ - 0.788X_1^2 - 0.647X_2^2 + 0.314X_5^2 - 0.702X_6^2 + 0.399X_9^2 - 0.788X_2X_6 - 0.748X_2X_9$$

where Y is the percent theoretical green density.

In this equation the X's are defined to run from -1 to +1. By inserting the appropriate values for the various X's (extrapolated from the limits in Table 2) this equation can be used to give the predicted or expected theoretical density at any combination of the X's in question.

The equation gives rise to a multi-dimensional response surface which is difficult to visualize. The series of curves (Figures 7 through 14) were plotted to depict the relationship. Press Pressure (X_2) appears as a variable with each set of curves since X_2 is the one variable which interacted with the others.

The curves, labeled Figures 7 through 14, depict the following situations:

Figure
Number

Description

7	Green Density versus PuO_2 calcine temperature (x_1) with Carbowax binder (x_7) for three levels of press pressure (x_2) at $x_5 = x_6 = x_8 = x_9 = 0$.
8	Same as Figure 7 with IT-Cell binder
9	Green Density versus binder percentage (x_5) with Carbowax binder (x_7) for three levels of press pressure (x_2) at $x_1 = x_6 = x_8 = x_9 = 0$.
10	Same as Figure 9 with M-Cell binder.
11	Green density versus preslug pressure (x_6) with Carbowax binder (x_7) for three levels of press pressure (x_2) at $x_1 = x_5 = x_8 = x_9 = 0$.
12	Same as Figure 11 with M-Cell binder.
13	Green density versus percent recycle (x_9) with Carbowax binder (x_7) for three levels of press pressure (x_2) at $x_1 = x_5 = x_6 = 0$. $x_8 = 0$ for $x_9 > 0\%$, and is undefined at $x_9 = 0\%$.
14	Same as Figure 13 with M-Cell binder.

Figure 15 shows the observed density for the 70 groups as a function of the density predicted by the model. In general the observed density is very close to that predicted. The standard deviation which expresses the scatter of these points about this line is 0.67.

The standard deviation between pellets within groups was estimated to be 0.31 (density percent) which indicates the amount of variation that will exist among the pellets produced at one set of conditions. If experimental control were perfect, we would then expect the standard deviation between group averages to be about $0.31 / \sqrt{11}$ or 0.093. In

actual fact , this was observed to be 0.67, the difference reflecting the combined effect of time-associated-undefined variables, possible inadequacies in the model, and possible difficulties in attaining the exact levels of the specified independent variables. Variation between pellets once groups conditions are fixed is not considered a major source of uncertainty. This is not unexpected and one implication is; that in future experimentation of this sort, it would not be necessary to measure as many pellets in each group.

An evaluation of x_8 , the particle size of recycle material , was based on analysis of groups 63 to 74. In these groups the variables were x_1 and x_9 in addition to x_8 . For these 12 groups x_2 , x_5 , and x_6 were all held at their 0 level. The binder type (x_7) was at the +1 level, M-Cell.

To evaluate the effect of the particle size of recycle material the equation developed on the basis of the other 70 observations was adjusted for the 12 observations containing the values of x_1 and x_9 present. Calculations are shown in Table 3.

TABLE 3
EFFECT OF SINTERED RECYCLE PARTICLE SIZE
ON PERCENT THEORETICAL GREEN DENSITY

x_1	x_8	x_9	y	y'
-1	-1	0	47.30	-0.68
-1	-1	1	49.49	-0.14
1	-1	1	52.05	-0.87
1	-1	0	49.57	-1.70
1	0	1	52.91	-0.01
1	0	0	50.76	-0.51
-1	0	0	47.59	-0.39
-1	0	1	48.67	-0.96
-1	1	0	46.25	-1.73
-1	1	1	47.63	-2.00
1	1	1	50.62	-2.30
1	1	0	49.38	-1.89

Where: Y is the percent theoretical green density

Y^1 is the residual

If X_8 had no effect we would expect the Y^1 values in the last column to all be near 0, since these values represent the densities corrected for the values of X_1 , X_2 , X_5 , X_6 , X_7 and X_9 present. Also, since the previous model was based on $X_8 = 0$ we would expect for consistency that the Y^1 values corresponding to these four pellet groups be not different from 0. The four values in question average out to -0.47 which is not significantly different from 0.

It is of interest to compare the first four values in this table with the next four and with the last four, since the first four values are at the -1 level of X_8 , the middle four are at the 0 value, and the last four are at the +1 value. The averages are respectively -0.85, -0.47, and -1.98. The first two of these averages are not significantly different from one another but the average corresponding to the **largest** particles is significantly smaller. This indicates that with the **larger** particles the green theoretical density is about 1.3% lower than with the **smaller** particles. The standard deviation between the pellets for a fixed level of X_8 is about 0.5, indicating that X_8 does not interact with either X_1 or X_9 (surface area of PuO_2 and percent addition of sintered recycle).

FIGURE 7. GREEN DENSITY VERSUS PuO_2 CALCINE TEMPERATURE WITH CARBOWAX BINDER FOR THREE LEVELS OF PRESS PRESSURE

FIGURE 8. GREEN DENSITY VERSUS PuO₂ CALCINE TEMPERATURE WITH M-CELL BINDER FOR THREE LEVELS OF PRESS PRESSURE

FIGURE 9. GREEN DENSITY VERSUS BINDER PERCENTAGE WITH CARBOWAX BINDER FOR THREE LEVELS OF PRESS PRESSURE

FIGURE 10. GREEN DENSITY VERSUS BINDER PERCENTAGE WITH M-CELL BINDER FOR THREE LEVELS OF PRESS PRESSURE

FIGURE 11. GREEN DENSITY VERSUS PRESUG PRESSURE WITH CARBOWAX BINDER FOR THREE LEVELS OF PRESS PRESSURE

FIGURE 12. GREEN DENSITY VERSUS PRESUG PRESSURE WITH M-CELL BINDER FOR THREE LEVELS OF PRESS PRESSURE

FIGURE 13. GREEN DENSITY VERSUS PERCENT RECYCLE WITH CARBOWAX BINDER FOR THREE LEVELS OF PRESSURE

FIGURE 14. DENSITY VERSUS PERCENT RECYCLE WITH M-CELL BINDER FOR
THREE LEVELS OF PRESS PRESSURE

FIGURE 15. OBSERVED VERSUS PREDICTED GREEN DENSITY

C. Sintered Pellet Data

The sintered pellet characteristics measured included length, weight, diameter, diameter variation, off-gas, and O/M ratio. The effect of the nine variables on those characteristics is discussed below. Length, average diameter, and weight were used to calculate percent theoretical density. For comparative purposes the theoretical density reported is based on the theoretical density of stoichiometric mixed oxide (11.08 grams/cc). As the O/M ratio of the sintered pellets averaged about 1.94, the true theoretical density is approximately 1.2% higher than that reported.

1, Statistical Treatment

As was done with green pellet data, the first step in the analysis involved editing the raw data. The data were very "clean" in the sense that very few computational errors were committed when calculating pellet density as a function of diameter, length, and weight.

Three dependent variables were considered with respect to pellet densities. These were the group average percent theoretical density, the variation between pellet densities within a given group, and the average nonuniformity as measured by the (max-min) pellet OD. It would be desirable to minimize these latter two variables, i.e., one would like a uniform pellet, and one would like all pellets sintered under nominally the same set of conditions to be as near identical as possible.

The model employed in the first attempt at estimation for each independent variable was the full quadratic model. This was based on observations of groups 1 through 62 and 75 through 82. The 12 groups in which X_8 , the particle size of recycle, was varied were excluded. Thus, the first model does not have X_8 appearing as a variable. It should be kept in mind that when referring to X_9 at its -1 level

(no recycle). X_8 as a variable is undefined. When X_9 takes on the value 0 or 1, X_8 is defined to take on its 0 value of -200 +325 mesh size.

2. Analysis for Density

The dependent variable of primary interest is again the average percent theoretical density expressed as a function of the process variables. Each average is based on measurements obtained on 12 pellets.

The first attempt at fitting the data with the full quadratic model indicated that most of the interactions could be deleted from the model. The dominant effects are the main effects of each variable involved with exception of variables X_1 and X_4 . The three interaction terms retained in the model were X_1X_6 , X_2X_6 , and X_3X_5 .

Upon deleting the remaining interaction terms from the model and re-estimating the parameters the following equation resulted.

$$Y = 83.633 + 0.805X_2 + 1.206X_3 - 2.067X_5 - 1.240X_6 - 1.539X_7 - 1.063X_9 \\ + 1.979X_3^2 - 1.209X_9^2 - 1.178X_1X_6 + 0.745X_2X_6 + 1.249X_3X_5$$

where Y = percent theoretical sintered density

In this equation the X's are defined to run from -1 to +1. By inserting the appropriate values for the various X's extrapolated from the limits in Table 2 this equation can be used to give the predicted or expected theoretical density at any combination of X's in question.

The equation gives rise to a multi-dimensional response surface which is difficult to visualize. The series of curves presented in Figures 16 through 23 should help depict the relationships.

Figure
Number

Description

16	Sintered density versus press pressure (x_2) with Carbowax binder (x_7) for three levels of preslug pressure (x_6) at $x_1 = x_3 = x_4 = x_5 = x_9 = 0$
17	Same as Figure 16 with M-Cell binder
18	Sintered density versus binder percentage (x_5) with Carbowax binder (x_7) for three levels of preslug pressure (x_6) at $x_1 = x_2 = x_3 = x_4 = x_9 = 0$
19	Same as Figure 18 with M-Cell binder
20	Sintered density versus binder percentage (x_5) with Carbowax binder (x_7) for three levels of sintering temperature (x_3) at $x_1 = x_2 = x_4 = x_6 = x_9 = 0$
21	Same as Figure 20 with M-Cell binder
22	Sintered density versus calcine temperature (x_1) with Carbowax binder (x_7) for three levels of preslug pressure (x_6) at $x_2 = x_3 = x_4 = x_5 = x_9 = 0$
23	Same as Figure 22 with M-Cell binder

One is interested in knowing just how adequate the model is. A measure of the "goodness of fit" is to plot the observed density for the 70 groups as a function of the density predicted by the model (Figure 24). In general the observed density is close to that predicted. The standard deviation which expresses the scatter of these points about this line is 1.54.

The standard deviation between pellets within groups was estimated to be 0.38 (density percent) which indicates that when process conditions are fixed, this is the amount of variation that will exist among the pellets produced at one set of conditions. If experimental control were perfect, we would then expect the standard deviation between cell averages to be about $0.38/\sqrt{11}$ or 0.114. In actual fact, this was observed to be 1.54, the difference reflecting the combined effect of time-associated-undefined variables, possible inadequacies in the model, and possible difficulties in attaining the exact levels of the independent variables as specified. Variation between pellets, once group conditions are fixed, is not considered a major source of uncertainty.

The effect of X_8 (particle size of recycle material) is estimated based on observations 63 to 74. In these runs the variables were X_1 , X_3 , X_9 , in addition to X_8 . For these 12 observations X_2 , X_4 , X_5 and X_6 were all held at their 0 level and the binder type (X_7) was at the +1 level (M-Cell).

The effect of sintered recycle particle size was evaluated by adjusting the equation developed on the basis of the other 70 observations with the values of X_1 , X_3 , and X_9 obtained from the 12 groups in question. Calculations are shown in Table 4.

TABLE 4
EFFECT OF SINTERED RECYCLE PARTICLE SIZE
ON SINTERED DENSITY

X_1	X_3	X_8	X_9	Y	Y^1
-1	-1	-1	0	82.87	1.42
-1	1	-1	1	83.01	0.97
1	-1	-1	1	80.60	3.07
1	1	-1	0	85.28	1.69
1	1	0	1	83.01	0.71
1	-1	0	0	82.87	1.10
-1	1	0	0	85.28	0.60
-1	-1	0	1	80.66	0.00
-1	-1	1	0	82.87	-1.83
-1	1	1	1	83.01	-2.55
1	-1	1	1	80.60	-3.48
1	1	1	0	85.28	0.27

where Y is sintered density

Y^1 is the residual

If X_8 had no effect we would expect Y^1 values in the last column to all be near 0, since these values represent the densities corrected for the values of X_1 , X_2 , X_3 , X_4 , X_5 , X_6 , X_7 and X_9 present. Also, since the previous model was based on $X_8 = 0$ we would expect for consistency that the Y^1 values corresponding to these four groups be not different from 0. The four values in question average out to 0.60 which is not significantly different from zero.

It is of interest to compare the first four values in this table with the next four and with the last four, since the first four values are at the -1 level of X_8 , the middle four are at the 0 value, and the last four are at the +1 value. The averages are respectively 1.79, 0.60, and -1.90. The second average is significantly smaller than the first average and the third

average is significantly smaller than the second average. The conclusion is that with the larger particles the theoretical density is about 2.7% lower than with these smaller particles. The standard deviation between the pellets for a fixed level of X_8 is about 0.99, indicating the X_8 does not interact with X_1 , X_3 , or X_9 .

3, Variation Among Pellets Within Groups

This variable is measured by the variance between pellet densities within each group. The natural logarithm of this quantity is used in the analysis in order to stabilize the variance. The resulting model in this case is:

$$\begin{aligned} Y = & -2.949 - 0.370X_1 + 0.297X_4 + 0.181X_7 + 0.301X_9 + 0.472X_3^2 \\ & + 0.602X_4^2 + 0.825X_5^2 + 0.728X_6^2 - 0.572X_1X_6 - 0.490X_1X_9 \\ & + 0.535X_2X_4 + 0.782X_2X_9 - 0.516X_3X_5 + 0.695X_4X_6 + 0.641X_5X_9 \end{aligned}$$

The X's are defined the same way as they were in the density model.

Figure 25 is the "goodness of fit" plot for the variation data.

With the transformation used, the residual standard deviation should theoretically be 0.41 with a perfect fit. It was estimated to be 0.74 with the model used, and since it was only 1.10 before fitting the model, it is clear that the variables considered were only partially effective in accounting for the overall variation. Other variables, which have not been identified, contribute to the overall variation.

4. Uniformity in Pellet Diameter

This was measured by determining the average (max-min) OD within a group. This ranged from 0 to 55 mils for these data, the average being 1.0 mils. The model, after elimination of insignificant terms is:

FIGURE 16. SINTERED DENSITY VERSUS PRESS PRESSURE WITH CARBOWAX BINDER FOR THREE LEVELS OF PRESUG PRESSURE

FIGURE 17. SINTERED DENSITY VERSUS PRESS PRESSURE WITH M-CELL BINDER FOR THREE LEVELS OF PRESUG PRESSURE

FIGURE 18. SINTERED DENSITY VERSUS BINDER PERCENTAGE WITH CARBOWAX BINDER FOR THREE LEVELS OF PRESUG PRESSURE

FIGURE 19. SINTERED DENSITY VERSUS BINDER PERCENTAGE WITH M-CELL BINDER FOR THREE LEVELS OF PRESUG PRESSURE

FIGURE 20. SINTERED DENSITY VERSUS BINDER PERCENTAGE WITH CARBOWAX BINDER FOR THREE LEVELS OF SINTERING TEMPERATURE

FIGURE 21. SINTERED DENSITY VERSUS BINDER PERCENTAGE WITH M-CELL BINDER FOR THREE LEVELS OF SINTERING TEMPERATURE

FIGURE 22. SINTERED DENSITY VERSUS PuO₂ CALCINE TEMPERATURE WITH CARBOWAX BINDER FOR THREE LEVELS OF PRESUG PRESSURE

FIGURE 23. SINTERED DENSITY VERSUS PuO_2 CALCINE TEMPERATURE WITH M-CELL BINDER FOR THREE LEVELS OF PRESLOG PRESSURE

FIGURE 24. OBSERVED VERSUS PREDICTED SINTERED DENSITY

FIGURE 25. DENSITY VARIATION AMONG PELLETS WITHIN GROUPS

FIGURE 26. DIAMETRAL VARIATION BETWEEN PELLETS

$$Y = 1.672 + 0.422X_2 + 0.453X_5 - 0.806X_6 - 0.679X_9 + 0.541X_9^2 - 0.562X_1X_3 \\ - 0.425X_1X_9 - 0.375X_2X_3 + 0.540X_2X_5 + 0.269X_2X_6 + 0.590X_2X_9$$

Where Y = (max-min) OD expressed in mils

Figure 26 is the "goodness of fit" plot for the (max-min) OD data.

The standard deviation before fitting the model was 1.06 mils, while the residual after fitting the model was 0.56 mils. Thus, fitting the model did reduce the variation. The results indicate that the variation could be reduced further by including other undefined variables in the model which effect the uniformity in pellet diameter.

The variables which have the greatest effect on pellet diameter variation are preslug pressure and percent recycle followed by binder percentage and press pressure. The higher preslug pressure of 50,000 pounds and percent recycle of 15% appear more desirable along with small binder percentage of 2% and press pressure of 15,000 psi.

The evaluation of X_8 the particle size of recycle material indicates that the variable does not have a significant effect on the uniformity in pellet diameter.

5. Percent Diameter Shrinkage

The percent shrinkage is calculated for each group in the following way:

$$\text{Percent Shrinkage} = \frac{\text{Avg. Green Dia.} - \text{Avg. Sintered Dia.}}{\text{Avg. Green Dia.}} \times 100$$

The first attempt at fitting the data with the full quadratic model indicated that most of the interactions could be deleted from the model. The dominant effects are the main effects of each variable involved with exception of variables X_4 . The six interaction terms retained in the

model were X_1X_6 , X_2X_5 , X_2X_6 , X_2X_9 , X_3X_4 and X_3X_5 . Upon deleting the remaining interaction terms from the model and re-estimating the parameters the following equation resulted.

$$Y = 16.604 - 0.973X_1 - 1.255X_2 + 0.427X_3 - 0.333X_5 - 1.082X_6 + 0.685X_7 \\ - 1.172X_9 + 0.430X_3^2 - 0.339X_5^2 + 0.309X_6^2 - 0.827X_9^2 - 0.349X_1X_6 \\ + 0.501X_2X_5 + 0.648X_2X_6 + 0.681X_2X_9 - 0.342X_3X_4 + 0.390X_3X_5$$

Where Y = percent diameter shrinkage

In this equation the X's are defined to run from -1 to +1. By inserting the appropriate values for various X's this equation can be used to give the predicted or expected percent shrinkage at any combination of X's in question.

As the equation gives rise to a multi-dimensional response surface, the series of curves given in Figures 27 through 38 should help depict the relationships.

<u>Figure Number</u>	<u>Description</u>
27	Percent shrinkage versus PuO_2 calcine temperature (X_1) with Carbowax binder (X_7) for three levels of preslug pressure (X_6) at $X_2 = X_3 = X_4 = X_5 = X_9 = 0$
28	Same as Figure 27 with M-Cell binder
29	Percent shrinkage versus press pressure (X_2) with Carbowax binder (X_7) for three levels of binder percentage (X_5) at $X_1 = X_3 = X_4 = X_6 = X_9 = 0$
30	Same as Figure 29 with M-Cell binder
31	Percent shrinkage versus press pressure (X_2) with Carbowax binder (X_7) for three levels of preslug pressure (X_6) at $X_1 = X_3 = X_4 = X_5 = X_9 = 0$

<u>Figure Number</u>	<u>Description</u>
32	Same as Figure 31 with M-Cell binder
33	Percent shrinkage versus press pressure (X_2) with Carbowax binder (X_7) for three levels of percent recycle (X_9) at $X_1 = X_3 = X_4 = X_5 = X_6 = 0$
34	Same as Figure 33 with M-Cell binder
35	Percent shrinkage versus sintering temperature (X_3) with Carbowax binder (X_7) for three levels of sintering time (X_4) at $X_1 = X_2 = X_5 = X_6 = X_9 = 0$
36	Same as Figure 35 with M-Cell binder
37	Percent shrinkage versus sintering temperature (X_3) with Carbowax Binder (X_7) for three levels of binder percentage (X_5) at $X_1 = X_2 = X_4 = X_6 = X_9 = 0$
38	Same as Figure 37 with M-Cell binder

The multiple correlation coefficient for the model is 0.96 which indicates that the model explains 92% of the variation in the percent shrinkage data. Another measure of the "goodness of fit" is a plot of the observed percent shrinkage for the 70 groups as a function of the percent shrinkage predicted by the model (Figure 39). The standard deviation which expresses the scatter of these points about this line is 0.54.

To evaluate the effect of X_8 (the particle size of recycle material), the equation developed on the basis of the other 70 observations was adjusted with values of X_1 , X_3 , and X_9 present in the 12 groups in question. For these 12 groups the variables X_2 , X_4 , X_5 and X_6 were all held at their 0 level and binder type (X_7) was at the +1 level (M-Cell). Calculations are shown in Table 5.

TABLE 5
EFFECT OF SINTERED RECYCLE PARTICLE SIZE
ON PERCENT DIAMETER SHRINKAGE

X_1	X_3	X_8	X_9	Y	Y^1
-1	-1	-1	0	18.27	0.62
-1	1	-1	1	17.12	0.72
1	-1	-1	1	14.32	1.92
1	1	-1	0	16.35	2.40
1	1	0	1	15.17	0.73
1	-1	0	0	16.32	0.80
-1	1	0	0	19.12	0.32
-1	-1	0	1	17.31	-0.34
-1	-1	1	0	18.26	0.46
-1	1	1	1	17.12	0.56
1	-1	1	1	14.32	0.62
1	1	1	0	16.83	1.76

Where Y is percent diameter shrinkage

Y^1 is the residual

If X_8 had no effect we would expect Y^1 values in the last column to all be near 0, since these values represent the densities corrected for the values of X_1 , X_2 , X_3 , X_4 , X_5 , X_7 and X_9 present. Also, since the previous model was based on $X_8 = 0$ we would expect for consistency that Y^1 values corresponding to these four groups be not different from 0. The four values in question average out to 0.88 which is not significantly different from 0.

It is of interest to compare the first four values in this table with the next four and with the last four, since the first four values are at -1 level of X_8 , the middle four are at 0 value, and the last four are at the +1 value. The values are 1.42, 0.38, and 0.85 which are not

significantly different from each other. Therefore, the results indicate that the particle size of recycle material has little effect on the percent shrinkage.

6. Off-gas

Five pellets ranging from the lowest to the highest density for both binder types were analyzed for off-gas. The results are shown in Figure 40.

Off-gas is expressed as a function of density for each binder type. The model equation is:

$$Y = \beta_0 + \beta_1 X + E$$

Where Y = Off-gas in cc/g

β_0 = A constant depending on binder type (0-intercept)

β_1 = A constant also depending on binder type (slope)

X = Percent theoretical density

E = Confidence band related to binder type and factor desired

An example of values obtained for the constants are as follows:

<u>Constant</u>	<u>Average Value</u>
β_0	22.795
β_1	- 0.253
E (95% Level)	~ 0.74

The off-gas values obtained are generally high and reflect the density range of pellet groups within the fractional factorial experiment (75% to 89% theoretical). Additional data from other development experiments indicate that the off-gas-density relationship is not linear, and values from sintered pellets in the 90% to 95% theoretical density range do not fit the curve. In addition, variations in binder addition technique and in density control methods (such as high pressure

FIGURE 27. PERCENT SHRINKAGE VERSUS PuO_2 CALCINE TEMPERATURE WITH CARBOWAX BINDER FOR THREE LEVELS OF PRESUG PRESSURE

FIGURE 28. PERCENT SHRINKAGE VERSUS PuO_2 CALCINE TEMPERATURE WITH M-CELL BINDER FOR THREE LEVELS OF PRESLOG PRESSURE

FIGURE 29. PERCENT SHRINKAGE VERSUS PRESS PRESSURE WITH CARBOWAX BINDER FOR THREE LEVELS OF BINDER PERCENTAGE

FIGURE 30. PERCENT SHRINKAGE VERSUS PRESS PRESSURE WITH M-CELL BINDER FOR THREE LEVELS OF BINDER PERCENTAGE

FIGURE 31. PERCENT SHRINKAGE VERSUS PRESS PRESSURE WITH CARBOWAX BINDER FOR THREE LEVELS OF PRESUG PRESSURE

FIGURE 32. PERCENT SHRINKAGE VERSUS PRESS PRESSURE WITH M-CELL BINDER FOR THREE LEVELS OF PRESLOG PRESSURE

FIGURE 33. PERCENT SHRINKAGE VERSUS PRESS PRESSURE WITH CARBOWAX BINDER FOR THREE LEVELS OF PERCENT RECYCLE

FIGURE 34. PERCENT SHRINKAGE VERSUS PRESS PRESSURE WITH M-CELL BINDER FOR THREE LEVELS OF PERCENT RECYCLE

FIGURE 35. PERCENT SHRINKAGE VERSUS SINTERING TEMPERATURE WITH CARBOWAX BINDER FOR THREE LEVELS OF SINTERING TIME

FIGURE 36. PERCENT SHRINKAGE VERSUS SINTERING TEMPERATURE WITH M-CELL BINDER FOR THREE LEVELS OF SINTERING TIME

FIGURE 37. PERCENT SHRINKAGE VERSUS SINTERING TEMPERATURE WITH CARBOWAX BINDER FOR THREE LEVELS OF BINDER PERCENTAGE

FIGURE 38. PERCENT SHRINKAGE VERSUS SINTERING TEMPERATURE WITH M-CELL BINDER FOR THREE LEVELS OF BINDER PERCENTAGE

FIGURE 39. OBSERVED VERSUS PREDICTED PERCENT SHRINKAGE

FIGURE 40. EFFECTS OF BINDER TYPE AND SINTERED PELLET DENSITY ON OFF-GAS QUANTITY

preslug or presintering) are also expected to affect the equation. To obtain a model equation for pellets in the 85% to 95% TD range, additional data are necessary.

Further analysis indicates that the difference between the off-gas values for the two binder types used (Methyl-Cellulose and Carbowax) was insignificant compared to the density effect. (The M-Cell and Carbowax lines in Figure 40 may be averaged.)

7. O/M Ratio

Two pellets from each of 22 pellet groups was analyzed for O/M ratio. A model equation was obtained; however, the variations observed both before and after fitting the model make the equation suspect. The variables considered were only partially effective in accounting for the overall variation. The small amount of tungsten carbide impurity is also believed to have had an effect on the values observed.

8. Blended Powder Surface Area Effects on Density

The relationship between blended powder surface area and density was investigated for both green and sintered pellets. The analysis was based on data from 22 pellet groups. Correlation coefficients between surface area and green density, and between surface area and sintered density were -0.17 and 0.10 respectively. Neither one of these coefficients is significantly different from zero at the 0.05 level of significance which indicates little, if any, relationship between blended powder surface area and density.

9. Green Density Effect on Sintered Density

The correlation coefficient between green density and sintered density was calculated as 0.26 for 70 pellet groups. While this correlation coefficient is significantly different from zero at the 0.05 level, its size indicates that the correlation between green and sintered density is rather poor.

D. Statistical Significance of Variables

A brief summary of analytical results from the fractional factorial experiment are presented in Table 6. The correlation coefficients indicate that a high percentage of variability observed can be explained by the model equations. This percentage may be calculated by multiplying R^2 in Table 6 by 100.

TABLE 6
SUMMARY OF ANALYTICAL RESULTS FROM
THE FRACTIONAL FACTORIAL EXPERIMENT

Dependent Variable	Multiple Correlation Coefficient		Is the relationship statistically significant at the 95% confidence level?	Independent Variables								
	<u>R</u>	<u>R²</u>		Is the regression coefficient statistically significant at the 95% confidence level for the range tested?								
				<u>X₁</u>	<u>X₂</u>	<u>X₃</u>	<u>X₄</u>	<u>X₅</u>	<u>X₆</u>	<u>X₇</u>	<u>X₈</u>	<u>X₉</u>
Green Density	.9811	.9626	Yes	Yes	Yes	NA	NA	Yes	Yes	Yes	NA	Yes
Sintered Density	.8639	.7463	Yes	No	Yes	Yes	No	Yes	Yes	Yes	Small	Yes
Density Variation	.7358	.5414	Yes	Yes	No	No	Yes	No	No	Yes	No	Yes
Diametral Variation	.8469	.7172	Yes	No	Yes	No	No	Yes	Yes	No	No	Yes
% Diameter Shrinkage	.9559	.9137	Yes	Yes	Yes	Yes	No	Yes	Yes	Yes	Small	No

X₁ - Surface Area of PuO₂

X₆ - Preslug Pressure

X₂ - Press Pressure

X₇ - Binder Type

X₃ - Sintering Temperature

X₈ - Recycle Particle Size

X₄ - Sintering Time

X₉ - % Recycle Addition

X₅ - Binder Percent

NA - - Not analyzed

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