Dissolution of Unirradiated Mechanically Blended Mixed Oxide Fast Reactor Fuel (a)

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ABSTRACT
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A statistical, fractional factorial experiment was performed to determine the effect of six fuel fabrication variables on the dissolubility of unirradiated, mechanically blended PuO₂-UO₂ fuels. The six fabrication variables were source of PuO₂, PuO₂ content, sintering temperature, sintering time, rate of temperature rise during sintering, and pellet pressing pressure. All six fabrication variables had some effect on pellet dissolubility with source of PuO₂, PuO₂ content, and sintering temperature having major effects.
DISSOLUTION OF UNIRRADIATED MECHANICALLY
BLENDED MIXED OXIDE FAST REACTOR FUEL

INTRODUCTION

This paper describes experiments that have been performed at HEDL and
PNL to measure the dissolubility* of unirradiated, mechanically blended
mixed oxide fast reactor fuel. Two significant histories of fast reactor
fuels are recognized as having an important effect on the dissolution prop­
erties of the fuel. They are the fabrication history and the reactor
history. Investigations of the effect of fabrication parameters on dis­
solution are important to establish what operations influence dissolubil­
ity and dissolution rate of the fuel and to what extent. Dissolubility data on
unirradiated fuels provide the necessary base from which to assess the
effects of irradiation time, temperature, and reactor flux upon dissolubil­
ity and dissolution rate of irradiated fast reactor fuel. The information
will also be helpful for recycle of scrap materials from the fabrication
process and for optimizing the overall fuel cycle.

In order to investigate the effect of fabrication variables on the
dissolubility of unirradiated mechanically blended PuO$_2$-UO$_2$ fuels, a sta­
tistical, fractional-factorial experiment was performed.$^{1}$ The six
independent variables shown in Table 1 were investigated. The dependent
variable in the experiment was the weight percent of pellet that didn't
dissolve.

EXPERIMENTAL

On the basis of previous work on dissolution of mixed oxide fuel, all
six variables shown in Table 1 could have an effect on the dissolution
properties of the fuel. In particular, temperature and time$^{(2)}$ have been
previously shown to strongly affect dissolution of mechanically blended
15 wt% PuO$_2$ fuel. In setting the levels of the six variables, primary
consideration was given to procedures already in use or under consideration
for FFTF fuels. Only two levels of investigation were chosen for variables
$X_4$, $X_5$ and $X_6$ to facilitate fabrication of the pellets. The levels were
sufficiently separated so that any effects due to the particular variable

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* As used here, dissolubility refers simply to the fraction of plutonium and
uranium which dissolves when the oxide fuel is exposed to boiling nitric
acid for a short time [$<24$ hours].
were easily identified, however, whether or not the effect was linear
or curvilinear could not be determined.

The following fabrication parameters were held constant:
1. PuO$_2$ calcination temperature: 700°C
2. PuO$_2$ particle size: -325 mesh
3. UO$_2$: Eldorado ceramic grade
4. Blending conditions: wet process to provide good homogeneity
   for the small bath sizes
5. Ball milling: -325 mesh
6. Binder type and percentage: 3% carbowax
7. Drying conditions: 4-5 hours at 70°C
8. Screening of mixed oxide: agglomeration for press feed to -20 mesh
9. Pre-slug conditions: no pre-slug
10. Sintering atmosphere: Dry Ar-8% H$_2$; less than 5 ppm H$_2$O

A complete discussion of the choice of variables and their particular
effects on the physical properties of mixed oxide pellets has been published.$^{1,3}$

The statistical design of the experiment was a 1/3 replicate (i.e.,
it locked at 1/3 of all possible interactions) of a full factorial experiment
for three variables at three levels and three variables at two levels. The
design, defined to be the combination of values of the six variables for
each run, resulted in 72 treatment conditions or "cells", each cell
representing a different combination of fuel pellet fabrication variables.
About 12 pellets were fabricated for each particular cell.

The dissolubility procedure used in this experiment consisted of treating
single fuel pellets (ca. 0.2 in. in diameter x 1/4 in. long; 1.5 g) with boiling
12M nitric acid for one or, if necessary, two 6-hour periods as shown in
Figure 1. After the second 6-hour treatment any residue remaining was
separated from the solution, dried, and weighed. The residue was then dissoled
in fresh 12M HNO$_3$-0.05M HF and the resulting solution analyzed for
plutonium and uranium. Two or three pellets were dissolved for each cell
and the mean of the weight percent undissolved was used in the statistical analysis.

RESULTS

A full quadratic model, that is, one that considers all possible two factor interactions (e.g., $X_1^2$, $X_1X_2$, $X_1X_3$, etc.) of all six variables, was used in the first attempt at estimating the effect of each independent variable on dissolubility. After deleting the terms from the full quadratic model which were statistically insignificant and again estimating the parameters, the equation shown in Table 2 resulted.

In the equation, Y is the weight percent undissolved and the X's are the statistical designations of the particular variable and are defined to run from -1 to +1. By inserting the appropriate values for the various X's, this equation can be used to give the predicted or expected weight percent undissolved. A negative percent undissolved would indicate that the pellet is completely soluble. The equation can also be used to calculate response curves which show the effect of going from one level to another within a given variable.

The relative dissolubility of a pellet can be estimated from fabrication variables within the range of those studied (but not exactly the same as used in this study) by substituting a fractional value into equation 1 for the particular variable of interest. The fractional value would be directly proportional to the spacing within the variable and would have a value between -1 and +1. (For sintering temperatures of 1500°C and 1600°C, for example, the values of -0.33 and +0.33, respectively, would be substituted for $X_3$ in equation 1.)

The relative effect of each particular fabrication variable on dissolubility can be roughly estimated by comparing the coefficients in the equation. The relative order of decreasing effect was sintering temperature, source of PuO$_2$, PuO$_2$ content, rate of temperature rise during sintering, sintering time, and press pressure.

Since the equation as it stands is of limited direct use, a computer was used to calculate the predicted weight percent undissolved for a number of combinations of the X's. The series of curves given in the following sections was plotted from those calculated values and should be useful in depicting
some of the relationships that exist. They do not represent all possible
curves that could be drawn but rather represent selected curves that are
useful in depicting trends that exist within a particular fabrication
variable.

Source of PuO₂

The source of PuO₂ had a major effect on the dissolubility of mecha­
ically blended, mixed oxide fuel as shown in Figure 2 for 15 wt% PuO₂ pellets. Mixed oxide pellets made from burned metal PuO₂ were more soluble than pellets made from either calcined oxalate or calcined nitrate PuO₂ at all three
levels of sintering temperature and PuO₂ content investigated. Similar re­
lationships were noted for dissolution of 20 and 25 wt% PuO₂ pellets as shown in Figures 3 and 4. The primary difference was that the curves were
shifted toward lower dissolubilities (i.e., higher weight percent undis­
solved material) as the PuO₂ content increased. Source of PuO₂ produced the largest difference in dissolubility for the 15 wt% PuO₂ pellets.

PuO₂ Content

The PuO₂ content of the mixed oxide fuel also had a major influence on
the dissolubility of the fuel as shown in Figure 5 for burned metal PuO₂. The curves shown in Figure 5 are the same as the lower curves shown in
Figures 2, 3, and 4. In general, as the weight percent of PuO₂ increased,
the dissolubility decreased for all three sintering temperatures and all
three sources of PuO₂ investigated. The PuO₂ content produced the largest
difference in dissolubility of burned metal PuO₂-UO₂ pellets. The dis­
solubility relationships were similar for calcined oxalate and calcined
nitrate derived PuO₂.

Sintering Temperature

A third variable having a major effect on the dissolubility of the
mixed oxide fuel was the sintering temperature. As the sintering tempera­
ture increased, the dissolubility of the pellets increased for all three
sources of PuO₂ and for all three PuO₂ contents investigated. Typical
dissolubility curves are shown in Figure 6 for calcined nitrate PuO₂. In
general, the dissolubility of mixed oxide fuel increased by 3 to 5 percent
when the sintering temperature was raised from 1400°C to 1700°C. The
largest increase in dissolubility with an increase in sintering temperature
was noted for the UO₂-25 wt% PuO₂ fuel pellets. This indicates that a high sintering temperature (e.g., 1700°C) is necessary to obtain good mixed crystal formation (i.e., solid solution) and a high degree of dissolubility in the 25 wt% PuO₂ fuel.

Sintering Time

Sintering time (i.e., the time at temperature or "soak time") was also found to have an effect on dissolubility but to a lesser degree than the three variables discussed previously. An increase in sintering time produced an increase in dissolubility except for the 15 wt% PuO₂ pellets. The combined effects of sintering time and sintering temperature are shown in Figure 7 for burned metal PuO₂-UO₂ fuel. (Note the reverse effect of sintering time for the 15 wt% PuO₂ fuel.) The magnitude of the effect of soak time is essentially the same at all three temperatures investigated. The largest effect of soak time on dissolubility occurs with the 25 wt% PuO₂ fuel. This is probably due to the relative degree of solid solution taking place during sintering and is consistent with results of earlier work at ORNL(4) where researchers found that as the concentration of PuO₂ increased, the time (and/or temperature) required for solid solution formation also increased. Longer soak times than 6 hours would undoubtedly increase the dissolubility of 20 and 25 wt% PuO₂ fuel even further.

Rate of Temperature Rise During Sintering

The rate of temperature rise during sintering was also found to have an effect on the dissolubility of mixed oxide fuels but to a lesser degree than source of PuO₂, PuO₂ content, and sintering temperature. In general, a slower rate of temperature rise during sintering favored increased dissolubility at all three sintering temperatures investigated and for all three sources of PuO₂ as shown in Figure 8 for fuel sintered at 1700°C. The relationships are similar for fuel sintered at 1550°C and 1400°C with the dissolubility curves being shifted to lower dissolubility. In general, the magnitude of the increase in dissolubility was between 1 and 2 wt% when the rate of temperature rise during sintering was lowered from 250°C/hr to 100°C/hr.

Pressing Pressure

The final variable that was evaluated in this experiment was the
pressure used to press the blended oxide into green fuel pellets. The press pressure was found to have very little effect on dissolubility. In general, dissolubility of mixed oxide pellets was increased <0.5 wt% when the pellet pressing pressure was increased from 25,000 psi to 50,000 psi.

**DISCUSSION**

**Accuracy of Statistical Model**

To measure the accuracy of the model, a "goodness of fit" was prepared to compare the observed weight percent undissolved for the 72 observations as a function of the weight percent undissolved predicted by the model. Figure 9 shows the observed weight percent undissolved to be very close to that predicted. The standard deviation which expresses the scatter of these points about the line is ±0.53. Another way to express the adequacy of the model is in terms of the amount of the total variation of weight percent undissolved explained by the model. In this experiment, the multiple correlation coefficient was 0.976. Thus, the model explained approximately 95% of the total variation (calculated by multiplying 100 times the square of the multiple correlation coefficient) which indicates that the model was very effective in accounting for the overall variation.

The standard deviation between pellets within cells was calculated to be 0.11 (weight percent undissolved). If experimental control were perfect, the standard deviation between cell averages would be about 0.11/√2 or 0.08. In actual fact, the residual standard deviation, that is, the standard deviation expressing the scatter of data points about the predicted line, was observed to be 0.53. The difference reflects 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. A residual standard deviation of 0.53 is considered to be very good in this type of experiment.

**Density vs Dissolubility**

A plot of sintered density vs dissolubility is shown in Figure 10 for all of the samples investigated in this dissolubility experiment. The data were evaluated by regression analysis to determine the relationship between weight percent undissolved and sintered density of the fuel pellets. The
The model used in the analysis was of the form \(Y = b_0 + b_1 X\) where \(Y\) is the pellet density and \(X\) is the weight percent undissolved, \(b_0\) is the intersect of the line on the \(Y\) axis and \(b_1\) is the slope of the line. The estimates of \(b_0\) and \(b_1\) were 92.19 ±0.51 and -0.562 ±0.123, respectively. The estimate of the slope \(b_1\) is significantly different from zero at the 0.05 level of significance and thus indicates that there is some correlation between sintered density and dissolubility.

Sintered density was also added as an independent variable to the quadratic model used in the original evaluation of the dissolubility data (see equation 1). The addition of sintered density as a variable, however, did not improve the model in the sense of reducing the residual standard error.

Analysis of Residues

The residues remaining after dissolution were dissolved in 12M HNO₃ - 0.05M HF and the resulting solutions were analyzed for uranium and plutonium by X-ray fluorescence. Preferential dissolution of uranium was found to occur in every case. The ratio of plutonium oxide to uranium oxide in the final residues ranged from 3.9 to 14.0. The relationship between \(\text{PuO}_2/\text{UO}_2\) ratio in the residues and pellet "weight percent undissolved" is shown in Figure 11 for 25 wt% \(\text{PuO}_2\) pellets. The degree of preferential dissolution taking place was related directly to the amount of residue remaining in the sample.

The relationship between pellet weight percent undissolved and amount of starting \(\text{PuO}_2\) undissolved is shown in Figure 12. In the worst case, nearly 40 weight percent of the original plutonium oxide in the pellet was still undissolved after 12 hours in boiling 12M nitric acid. Greater than 99 wt% of the original uranium dioxide dissolved in all but one sample. All data in Figure 12 were fit to the lines by the method of least squares. The relative positions of the three lines are exactly as expected and are very near to the positions of a calculated line if only plutonium oxide remained in the residues.

Correlation of Microstructure and Physical Properties to Dissolubility

Micrographs have been (or will be) taken of pellets from all 72 cells at 10X, 250X, and 500X etched. The 500X etched micrographs will be used to
determine grain size within the pellets. The 10X and 250X micrographs will be used to observe differences in porosity (open vs closed, relative distribution) on a qualitative basis. In addition to the micrographs, some α-autoradiographs were taken of representative pellet batches and will be used to indicate homogeneity (non-quantitative) relative to other similar fabrication runs.

Possible correlation of dissolubility to actual physical properties of the pellets will be studied when characterization of all of the pellet batches is complete.

CONCLUSIONS

All six variables investigated in this experiment were shown to have an effect on the dissolubility of mixed oxide fuels. The relative order of decreasing effect was sintering temperature, source of PuO₂, PuO₂ content, rate of temperature rise during sintering, sintering time, and press pressure. The data provide good base line measurements from which to assess the effects, if any, of irradiation time, irradiation temperature and reactor flux upon dissolubility of fast reactor fuels as well as providing useful information concerning possible recycle of unirradiated fast reactor fuel materials.

ACKNOWLEDGMENTS

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REFERENCES


2. W. Baehr and T. Dippel. On the Dissolution of Breeder Fuels Containing PuO₂ in Nitric Acid for Aqueous Reprocessing by the Purex Method, KFK 673, July 1967. (Also available as report EURFNR-471.)


### TABLE 1: VARIABLES FOR DISSOLUTION EXPERIMENT
**MECHANICALLY BLENDED, UO₂-PuO₂ FUEL**

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### TABLE 2: DISSOLUBILITY EQUATION

\[
Y = 4.86 + 0.53X_1 + 0.94X_2 - 2.24X_3 - 0.24X_6 + 0.75X_5 - 0.14X_6 - 1.87X_1^2 - 0.36X_3^2 - 0.37X_1X_2 + 0.15X_1X_5 - 0.51X_2X_3 - 0.35X_2X_4 \tag{1}
\]

**X₁, X₂, X₃, X₄, X₅, X₆**
- Source of PuO₂
- PuO₂ Content
- Sintering Temperature
- Sintering Time
- Rate of Temperature Rise
- Press Pressure
FIGURE 1 Dissolubility Apparatus
FIGURE 2 The Effect of Source of PuO₂ and Sintering Temperature on Dissolubility of 15 wt% PuO₂-UO₂ Fuel
FIGURE 3 The Effect of Source of PuO$_2$ and Sintering Temperature on Dissolubility of 20 wt% PuO$_2$-UO$_2$ Fuel
FIGURE 4 The Effect of Source of PuO$_2$ and Sintering Temperature on Dissolubility of 25 wt% PuO$_2$-UO$_2$ Fuel
FIGURE 5 The Effect of PuO₂ Content and Sintering Temperature on Dissolubility of Burned Metal PuO₂-UO₂ Fuel
FIGURE 6  The Effect of PuO$_2$ Content and Sintering Temperature on Dissolubility of Calcined Nitrate PuO$_2$-UO$_2$ Fuel
FIGURE 7  The Effect of Sintering Time, Sintering Temperature, and PuO$_2$ Content on Dissolubility of Burned Metal PuO$_2$-UO$_2$ Fuel
\[ X_4 = X_6 = -1; X_2 = 0 \]

SINTERING TEMPERATURE: 1700°C

**FIGURE 8.** The Effect of Rate of Temperature Rise During Sintering and Source of PuO₂ on Dissolubility of Fuel Pellets Sintered at 1700°C
FIGURE 9 Observed Weight Percent Undissolved vs Predicted Weight Percent Undissolved
FIGURE 10. Relationship of Dissolubility to Pellet Density
FIGURE 11 Ratio of PuO$_2$/UO$_2$ in Residue Versus Weight Percent Undissolved for 25 wt% PuO$_2$ Pellets.
INITIAL PuO$_2$ CONTENT
- □ 15 wt% PuO$_2$
- ▽ 20 wt% PuO$_2$
- ○ 25 wt% PuO$_2$

*After two 6-hour treatments in boiling 12M HNO$_3$.

FIGURE 12 Total Weight Percent Undissolved Versus Weight Percent of PuO$_2$ Undissolved
# APPENDIX A

## TABLE A-1

**LEVELS OF THE SIX VARIABLES USED IN DISSOLUBILITY EXPERIMENT**

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