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# STATISTICAL TECHNIQUE FOR DETERMINING SOURCES OF VARIATION IN $(U, Pu)O_2$ OXYGEN-TO-METAL RATIO

W. J. Lackey, R. A. Bradley, W. H. Pechin, and T. L. Hebble

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# STATISTICAL TECHNIQUE FOR DETERMINING SOURCES OF VARIATION IN $(U,Pu)O_2$ OXYGEN-TO-METAL RATIO

W. J. Lackey, R. A. Bradley, W. H. Pechin, and T. L. Hebble

JUNE 1971

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# STATISTICAL TECHNIQUE FOR DETERMINING SOURCES OF VARIATION IN $(U,Pu)O_2$ OXYGEN-TO-METAL RATIO

W. J. Lackey, R. A. Bradley, W. H. Pechin, and T. L. Hebble[1]

## ABSTRACT

With a statistically designed experiment we demonstrated that gravimetric determinations of the oxygen-to-metal ratio of $(U,Pu)O_2$ depend strongly on the day of analysis, slightly on the particular sintering run, and not detectably on position in either the sintering furnace or the analysis furnace. We can determine the average oxygen-to-metal ratio of a single sintering run to a precision of ±0.005 at the 95% confidence level.

## INTRODUCTION

Since the characterization of ceramic processes and products frequently requires numerous analyses, the use of statistics to facilitate the design of sampling plans and the analysis of experimental data is advantageous. As an example, we used a statistically designed experiment to isolate the sources of variation in the oxygen-to-metal ratio found in $(U,Pu)O_{2\pm x}$. The example is pertinent since $(U,Pu)O_{2\pm x}$ is an important reactor fuel whose performance depends upon its oxygen-to-metal ratio.

Our purpose was to determine the precision to which slightly hypostoichiometric fuel can be fabricated and analyzed and to isolate and measure the sources of variation associated with fabrication and analysis. In particular, we wished to:

1. determine how much, if any, the oxygen-to-metal ratio varied between nominally identical sintering runs;
2. determine if the positions in the tube furnace used for sintering were a source of variation in the oxygen-to-metal ratio;

[1]Mathematics Division.

3. determine if positions in the tube furnace used for oxygen-to-metal ratio determination were a source of variation in the analytical results;
4. determine if the results of the analysis depended on the day of analysis; and
5. determine the precision of the analytical technique.

## EXPERIMENTAL

The stoichiometry was determined by a widely accepted gravimetric technique.[2] The oxygen-to-metal ratio is calculated from the weight before and after an oxidation-reduction treatment that is assumed to yield stoichiometric $(U,Pu)O_2$. The exact gas composition, temperature, and time required to yield precisely stoichiometric $(U,Pu)O_2$ are subject to debate;[3,4] the treatment thought to yield stoichiometric material could actually produce an oxygen-to-metal ratio in the range $2.000 \pm 0.005$. However, this potential error was not the subject of this investigation; we were concerned with the precision of the analytical technique rather than its accuracy.

Two pellets each from the inlet gas end, the middle, and the effluent gas end of the sintering furnace were analyzed for each of six different sintering runs. The objectives were accomplished by basing the selection and order of analysis of samples on a modification of a statistical

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[2]W. L. Lyon, The Measurement of Oxygen to Metal Ratio in Solid Solutions of Uranium and Plutonium Dioxides, GEAP-4271 (May 31, 1963).

[3]C. E. McNeilly and R. E. Woodley, pp. 2.17–2.19 in Quarterly Progress Report October, November, December 1969 Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1279 (February 1970).

[4]J. W. Dahlby, G. R. Waterbury, and C. F. Metz, Los Alamos Scientific Laboratory, "The Determination of Oxygen to Metal Atom Ratios in Sintered Oxides," paper presented at the 13th Conference on Analytical Chemistry in Nuclear Technology, September 30–October 2, 1969, Gatlinburg, Tennessee. Also reported in Quarterly Status Report on the Advanced Plutonium Fuels Program July 1 to September 30, 1969, LA-4307-MS, p. 63 (limited distribution).

experimental design called a Graeco-Latin Square.[5] The modified design is known as a Finney or F-Square.[6–8] The resulting design is best understood by reference to Table 1. Each row in the randomized 6 × 6 array represents a position in the tube furnace used in the oxygen-to-metal

[5]R. L. Wine, Statistics for Scientists and Engineers, pp. 434–444, Prentice-Hall, Englewood Cliffs, New Jersey, 1964.

[6]D. J. Finney, "Orthogonal Partitions of the 6 × 6 Latin Squares," Ann. Eugenics 13, 184–196 (1946).

[7]A. Hedayat and E. Seiden, "F-Square and Orthogonal F-Squares Design: A Generalization of Latin Square and Orthogonal Latin Square Design," Ann. Math. Statist. 41(6), 2035–2044 (1970).

[8]R. F. White, Randomization Analysis of the General Experiment, ARL-70-0239 (October 1970).

Table 1. Modified Graeco-Latin Square Design and Oxygen-to-Metal Ratios[a]

| Analysis Furnace Position | Ratio Determined and Experimental Design for Each Day of Analysis | | | | | | Row Average |
|---|---|---|---|---|---|---|---|
| | Day 1 | Day 2 | Day 3 | Day 4 | Day 5 | Day 6 | |
| 1 | 1.975<br>Cγ | 1.982<br>Aα | 1.980<br>Fγ | 1.981<br>Dα | 1.957<br>Bβ | 1.972<br>Eβ | 1.9745 |
| 2 | 1.981<br>Bβ | 1.989<br>Fγ | 1.960<br>Eβ | 1.975<br>Cγ | 1.964<br>Aα | 1.975<br>Dα | 1.9740 |
| 3 | 1.990<br>Dγ | 1.983<br>Bα | 1.981<br>Aγ | 1.978<br>Eα | 1.950<br>Cβ | 1.988<br>Fβ | 1.9783 |
| 4 | 1.982<br>Eα | 1.982<br>Cβ | 1.974<br>Bα | 1.981<br>Fβ | 1.945<br>Dγ | 1.984<br>Aγ | 1.9747 |
| 5 | 1.986<br>Aβ | 1.972<br>Eγ | 1.982<br>Dβ | 1.976<br>Bγ | 1.951<br>Fα | 1.984<br>Cα | 1.9752 |
| 6 | 1.985<br>Fα | 1.978<br>Dβ | 1.977<br>Cα | 1.984<br>Aβ | 1.951<br>Eγ | 1.975<br>Bγ | 1.9750 |
| Column Average | 1.9832 | 1.9810 | 1.9757 | 1.9792 | 1.9530 | 1.9797 | |

[a]Letters A through F denote the six sintering runs. The Greek letters α, β, and γ denote sintering furnace positions. Mean oxygen-to-metal ratio for each sintering run and sintering furnace position:

| A | B | C | D | E | F | α | β | γ |
|---|---|---|---|---|---|---|---|---|
| 1.980 | 1.974 | 1.974 | 1.975 | 1.969 | 1.979 | 1.976 | 1.975 | 1.974 |

analysis, and each column represents a different day's analysis. The six sintering runs are represented by the letters A through F, and the Greek letters $\alpha$, $\beta$, and $\gamma$ designate respectively the inlet gas end, the middle, and the effluent gas end of the sintering furnace. Note that each row or column contains one pellet from each sintering run. Further, each sintering furnace position (Greek letter) occurs twice in each row, twice in each column, and twice with each sintering run.

## RESULTS AND DISCUSSION

The results of the 36 oxygen-to-metal determinations are given in Table 1. Based on a statistical analysis of the data, which is presented in the Appendix, we can say at the 95% level of significance that the day of analysis and the sintering run influenced the reported oxygen-to-metal ratio; the analysis day effect was much larger than that of the sintering run. Further statistical analysis of these data revealed that only day 5 differed significantly from the other days. Although we do not know with certainty, we expect that a systematic weighing error, such as an erroneous tare weight, caused all the oxygen-to-metal ratios measured on the fifth day to be low by about 0.027. We are currently weighing the same platinum wire each time an analysis is performed in an attempt to detect such errors. The standard deviation associated with the variation between sintering runs was 0.0032 in the oxygen-to-metal ratio and was predominantly the result of using two sintering furnaces. We used one furnace for runs A, D, and F and another for B, C, and E. There was no evidence that analysis furnace position or sintering furnace position influenced the oxygen-to-metal ratio. After subtracting the variation associated with the day of analysis and with the sintering run, the standard error of the gravimetric technique[9] was 0.0055.

The importance of using a statistically designed experiment in this case is evident if one considers the usual manner of sampling these six sintering runs. If the design had not been used, all the samples from a

[9]Two recent statistical analyses showed that this standard error was reduced to 0.004 by the use of a more precise analytical balance.

given sintering run probably would have been analyzed on the same day. In that case the day of bad analysis would not have been detected and the low oxygen-to-metal ratio would have been attributed to some sintering run effect.

To determine the most suitable sampling scheme for future analyses, confidence intervals for the mean oxygen-to-metal ratio of future sintering runs were calculated for a variety of sampling schemes. The results of these calculations are listed in Table 2. The confidence intervals are unacceptably wide for those cases where undetected days of bad analysis are assumed to exist. Analysis of more than one sample per day for a given sintering run is not particularly efficient, especially for the case where days of bad analysis are not detected and does

Table 2. Confidence Intervals[a] About the Mean Oxygen-to-Metal Ratio

| Number of Sintering Runs | Number of Days | Number of Analyses Per Day Per Sintering Run | 95% Confidence Interval About the Mean | |
|---|---|---|---|---|
| | | | Days of Bad Analysis Detected | Days of Bad Analysis Not Detected |
| 1 | 1 | 1 | 0.012 | 0.029 |
| 1 | 1 | 2 | 0.009 | 0.029 |
| 1 | 1 | 3 | 0.008 | 0.029 |
| 1 | 2 | 1 | 0.009 | 0.021 |
| 1 | 2 | 2 | 0.007 | 0.020 |
| 1 | 3 | 1 | 0.007 | 0.017 |
| 1 | 3 | 2 | 0.005 | 0.016 |
| 3[b] | 2 | 1 | 0.007 | 0.021 |
| 3 | 2 | 2 | 0.007 | 0.021 |
| 3 | 3 | 1 | 0.006 | 0.017 |

[a]The confidence interval would be constructed by subtracting and adding the tabulated uncertainty to the mean value of the oxygen-to-metal ratio.

[b]The confidence interval is for the mean oxygen-to-metal ratio of the three sintering runs.

not improve one's ability to detect such days. To detect such a day and to obtain the maximum information per analysis, we have adopted an analysis scheme that consists of analyzing one sample from each sintering run on each of three different days. This results in an uncertainty[10] of 0.007 at the 95% confidence level in the mean oxygen-to-metal ratio of a sintering run. If it becomes necessary to reduce this uncertainty, the number of days of analysis should be increased.

We believe this statistical design, as well as the many others available,[11,12] could be beneficially used in many types of ceramic investigations. Details of the calculations for our modified Graeco-Latin Square design are given in the Appendix.

## ACKNOWLEDGMENT

The authors thank A. L. Lotts for suggesting this investigation and J. L. Cooper and W. F. Vaughan for conducting the oxygen-to-metal determinations.

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[10]Based on the lower standard error now being obtained with the more precise balance, this uncertainty is 0.005 at the 95% confidence level.

[11]R. L. Wine, Statistics for Scientists and Engineers, pp. 311–451, Prentice-Hall, Englewood Cliffs, New Jersey, 1964.

[12]W. G. Cochran and G. M. Cox, Experimental Designs, Wiley, New York, 1957.

# APPENDIX

## Statistical Analysis

The experimental design used to investigate oxygen-to-metal ratios is a modified 6 × 6 Graeco-Latin Square recently referred to as a Finney or F-Square.[13,14] Although a 6 × 6 Graeco-Latin Square is an impossible configuration, the modification in which three levels instead of six levels are assigned to one variable makes the resulting design readily adaptable to experimental situations For completeness the underlying structure of the design and the observed oxygen-to-metal ratios are included in Table 3.

[13]A. Hedayat and E. Seiden, "F-Square and Orthogonal F-Squares Design: A Generalization of Latin Square and Orthogonal Latin Square Design," Ann. Math. Statist. 41(6), 2035–2044 (1970).

[14]R. F. White, Randomization Analysis of the General Experiment, ARL-70-0239 (October 1970).

Table 3. Modified Graeco-Latin Square Design and Oxygen-to-Metal Ratios[a]

| Analysis Furnace Position | Ratio Determined and Experimental Design for Each Day of Analysis | | | | | | Row Totals |
|---|---|---|---|---|---|---|---|
| | Day 1 | Day 2 | Day 3 | Day 4 | Day 5 | Day 6 | |
| 1 | 1.975<br>Cγ | 1.982<br>Aα | 1.980<br>Fγ | 1.981<br>Dα | 1.957<br>Bβ | 1.972<br>Eβ | 11.847 |
| 2 | 1.981<br>Bβ | 1.989<br>Fγ | 1.960<br>Eβ | 1.975<br>Cγ | 1.964<br>Aα | 1.975<br>Dα | 11.844 |
| 3 | 1.990<br>Dγ | 1.983<br>Bα | 1.981<br>Aγ | 1.978<br>Eα | 1.950<br>Cβ | 1.988<br>Fβ | 11.870 |
| 4 | 1.982<br>Eα | 1.982<br>Cβ | 1.974<br>Bα | 1.981<br>Fβ | 1.945<br>Dγ | 1.984<br>Aγ | 11.848 |
| 5 | 1.986<br>Aβ | 1.972<br>Eγ | 1.982<br>Dβ | 1.976<br>Bγ | 1.951<br>Fα | 1.984<br>Cα | 11.851 |
| 6 | 1.985<br>Fα | 1.978<br>Dβ | 1.977<br>Cα | 1.984<br>Aβ | 1.951<br>Eγ | 1.975<br>Bγ | 11.850 |
| Column Totals | 11.899 | 11.886 | 11.854 | 11.875 | 11.718 | 11.878 | 71.110 |

[a]Letters A through F denote the six sintering runs. The Greek letters α, β, and γ denote sintering furnace positions. Entry totals:

| A | B | C | D | E | F | α | β | γ |
|---|---|---|---|---|---|---|---|---|
| 11.881 | 11.846 | 11.843 | 11.851 | 11.815 | 11.874 | 23.716 | 23.701 | 23.693 |

Notation

Let $X_{(ijk\ell)}$ represent the oxygen-to-metal ratio for the ith day of analysis ($i = 1,2,\ldots,6$), jth analysis furnace position ($j = 1,2,\ldots,6$), kth sintering run ($k = 1,2,\ldots,6$), and $\ell$th sintering furnace position ($\ell = 1,2,3$). Further, let $X_{(\ldots\ldots)}$(71.110) be the sum of all 36 observations and $X_{(i\ldots)}$ be the sum of the oxygen-to-metal ratios for the ith analysis day. The dot notation symbols $X_{(.j..)}$, $X_{(..k.)}$, and $X_{(\ldots\ell)}$ are similarly defined. For example, $X_{(1\ldots)}$(11.899) is the sum of the first column (i.e., first analysis day) and $X_{(\ldots 3)}$(23.693) is the sum of all twelve $\gamma$ entries. A "hat" (^) covering certain quantities denotes an estimate of that quantity (e.g., $\hat{\sigma}^2$ is an estimate of $\sigma^2$).

Data Analysis

The data analysis is an extension of the Latin Square analysis and has been discussed by several authors including Finney[15] and White.[14] The analysis can be summarized by an analysis-of-variance scheme shown in Table 4. The expected mean square column is the mathematical expectation of the mean square. Note that in partitioning the sum of squares, there is no source representing interactions between the variables. In fact, the interactions are assumed to be zero.

The numerical entries for the analysis of variance are shown in Table 5. The mean squares due to days of analysis and sintering runs are significant at the 0.05 level since the calculated F value exceeds the tabulated or critical F value, as can be seen in Table 5.

Two different furnaces were used for the sintering runs: one for runs A, D, and F and the other for B, C, and E. Table 5 shows an orthogonal partition of the overall sum of squares due to sintering runs into the sum of squares due to furnaces (A+D+F vs B+C+E) and the sum of squares due to sintering runs (less furnaces). The F test indicates that the variation due to furnaces is significant but that due solely to sintering runs is not. These points should be kept in mind when the variance discussed later is selected.

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[15] D. J. Finney, "Orthogonal Partitions of the 6 × 6 Latin Squares," Ann. Eugenics 13, 184–196 (1946).

Table 4. Formulas for Analysis of Variance

| Source | Degrees of Freedom | Sum of Squares | Mean Square | Calculated F Value | Expected Mean Square |
|---|---|---|---|---|---|
| Days of analysis | 5 | $S_1 = \frac{\sum_{i=1}^{6} X^2_{(i...)}}{6} - \frac{X^2_{(....)}}{36}$ | $\frac{S_1}{5}$ | $\frac{18S_1}{5S_e}$ | $\sigma^2 + 6\sigma_1^2$ |
| Analysis furnace positions | 5 | $S_2 = \frac{\sum_{j=1}^{6} X^2_{(.j..)}}{6} - \frac{X^2_{(....)}}{36}$ | $\frac{S_2}{5}$ | $\frac{18S_2}{5S_e}$ | $\sigma^2 + 6\sigma_2^2$ |
| Sintering runs | 5 | $S_3 = \frac{\sum_{k=1}^{6} X^2_{(..k.)}}{6} - \frac{X^2_{(....)}}{36}$ | $\frac{S_3}{5}$ | $\frac{18S_3}{5S_e}$ | $\sigma^2 + 6\sigma_3^2$ |
| Sintering furnace positions | 2 | $S_4 = \frac{\sum_{\ell=1}^{3} X^2_{(...\ell)}}{12} - \frac{X^2_{(....)}}{36}$ | $\frac{S_4}{2}$ | $\frac{18S_4}{2S_e}$ | $\sigma^2 + 12\sigma_4^2$ |
| Residual | 18 | $S_e$ = difference | $\frac{S_e}{18}$ | | $\sigma^2$ |
| TOTAL | 35 | $\sum_{i=1}^{6} \sum_{j=1}^{6} X^2_{(ijk\ell)} - \frac{X^2_{(....)}}{36}$ | | | |

Table 5. Numerical Values for Analysis of Variance

| Source | Degrees of Freedom | Sum of Squares | Mean Square | F Value | |
|---|---|---|---|---|---|
| | | | | Calculated | 95% Critical |
| Days of analysis | 5 | 0.00375489 | 0.00075098 | 24.54 | 2.77 |
| Analysis furnace positions | 5 | 0.00007222 | 0.00001444 | 0.47 | 2.77 |
| Sintering runs | 5 | 0.00046856 | 0.00009371 | 3.06 | 2.77 |
| Furnaces (A+D+F vs B+C+E) | 1 | 0.00028951 | 0.00028951 | 9.46 | 4.41 |
| Sintering runs (less furnaces) | 4 | 0.00017905 | 0.00004476 | 1.46 | 2.93 |
| Sintering furnace positions | 2 | 0.00002272 | 0.00001136 | 0.37 | 3.55 |
| Residual | 18 | 0.00055084 | 0.00003060 | | |
| TOTAL | 35 | 0.00486923 | | | |

If the observations for the detected bad day of analysis (day 5) are omitted, the sum of squares due to analysis days is 0.0001816. The corresponding mean square is 0.00004540 with 4 degrees of freedom. This mean square is not significant at the 0.05 level since the calculated F(1.48) does not exceed the tabular F(2.93).

## Variance Estimates

An estimate of variance for each source in the table is calculated by equating the observed mean square to the expected mean square and solving for the particular variance. For example, $\hat{\sigma}_1^2$, the estimate of variance for days of analysis, $\sigma_1^2$, is found by equating $S_1/5$ to $\hat{\sigma}^2 + 6\hat{\sigma}_1^2$ and solving for $\hat{\sigma}_1^2$. Thus,

$$S_1/5 = \hat{\sigma}^2 + 6\hat{\sigma}_1^2 ,$$

and

$$\hat{\sigma}_1^2 = \frac{1}{6}\,[(S_1/5) - \hat{\sigma}^2]$$

$$= \frac{1}{6}\,[0.00075098 - 0.00003060] \qquad (1)$$

$$= 0.0001201 \ ,$$

where $\hat{\sigma}^2 = S_e/18 = 0.00003060$. In Eq. (1) we have assumed that days of bad analysis cannot be detected. However, if such days can be detected, $\sigma_1^2$ is estimated by

$$\hat{\sigma}_1^2 = \frac{(S_1'/4) - \hat{\sigma}^2}{5}$$

$$= 0.00000296 \ , \qquad (2)$$

where

$$S_1' = \frac{1}{6}\sum_{i=1}^{5} X_{(i\ldots)}^2 - \frac{1}{750}\left[\sum_{i=1}^{5} X_{(i\ldots)}\right]^2$$

$$= 0.0001815 \ . \qquad (3)$$

Here, it is understood that the six values for the detected bad day are omitted.

The remaining variance estimates are determined in a manner similar to that which generated Eq. (1). Since $\hat{\sigma}_2^2$ and $\hat{\sigma}_4^2$ become negative, set $\hat{\sigma}_2^2 = \hat{\sigma}_4^2 = 0$. The estimate for the variance of sintering runs is given by

$$\hat{\sigma}_3^2 = \frac{(S_3/5) - \hat{\sigma}^2}{6}$$

$$= \frac{1}{6}\,(0.00009371 - 0.00003060) \qquad (4)$$

$$= 0.00001052 \ .$$

## Confidence Intervals

Confidence intervals about the mean oxygen-to-metal ratios of future sintering runs are derived from the analysis-of-variance table. Two cases

are presented: in one the variation due to sintering is included and in the other it is omitted from the expression for variance. If the sintering run variation is excluded, inferences can be drawn only about a single sintering run. Each case is divided into two parts: (1) days of bad analysis can be detected and (2) days of bad analysis cannot be detected.

The general form of the confidence interval about a mean oxygen-to-metal ratio, $\bar{y}$, is given by formula (5):

$$\bar{y} \pm t\sqrt{\mathrm{Var}(\bar{y})} \ , \tag{5}$$

where

$t$ = Student's t for appropriate degrees of freedom and level of significance, and

$\mathrm{Var}(\bar{y})$ = variance of $\bar{y}$.

The formulas for the variance of $\bar{y}$ and the corresponding degrees of freedom are presented below. The degrees of freedom are estimated by Satterthwaite's approximation.[16]

Case I: The Variation Due to Sintering Runs is Omitted

In this case, we are interested in the variation within a single sintering run. Thus, we can only draw conclusions about the mean oxygen-to-metal ratio for that particular run. For n days of analysis and k analyses per day, the variance of the mean oxygen-to-metal ratio is

$$\mathrm{Var}(\bar{y}) = \frac{\hat{\sigma}_1^2}{n} + \frac{\hat{\sigma}^2}{nk} \ , \tag{6}$$

where

$\hat{\sigma}_1^2$ = estimate of variance of days of analysis, and

$\hat{\sigma}^2$ = error mean square

= 0.00003060 .

[16]F. E. Satterthwaite, "An approximate distribution of estimates of variance components," Biometrics 2, 110–114 (1946).

Days of Bad Analysis Detected. — If a day of bad analysis can be detected, $\hat{\sigma}_1^2$ is given by Eq. (2). An estimate of the degrees of freedom associated with $S_1'$ is based on Satterthwaite's[16] formula:

$$\text{d.f.} \approx \frac{[\text{Eq. (6) using } \hat{\sigma}_1^2 = 0.00000296]^2}{\frac{1}{4}\left[\left(\frac{1}{5n}\right)\left(\frac{S_1'}{4}\right)\right]^2 + \frac{1}{18}\left[\left(\frac{1}{nk} - \frac{1}{5n}\right)\frac{S_e}{18}\right]^2} . \tag{7}$$

Days of Bad Analysis Not Detected. — If a day of bad analysis cannot be detected, $\hat{\sigma}_1^2$ is given by Eq. (1). The estimate of the number of degrees of freedom is

$$\text{d.f.} \approx \frac{[\text{Eq. (6) using } \hat{\sigma}_1^2 = 0.0001201]^2}{\frac{1}{5}\left(\frac{1}{6n}\,\frac{S_1}{5}\right)^2 + \frac{1}{18}\left[\left(\frac{1}{nk} - \frac{1}{6n}\right)\frac{S_e}{18}\right]^2} . \tag{8}$$

Case II: The Variation Due to Sintering Runs is Included

In this case conclusions can be drawn about the mean oxygen-to-metal ratio of several sintering runs if the two sintering furnaces are employed in future runs. Since we plan to use these furnaces, we include the variation due to furnaces in the variance. For n days of analysis, m sintering runs, and $\ell$ analyses per day per sintering run, the variance of the mean oxygen-to-metal ratio is given by

$$\text{Var}(\bar{y}) = \frac{\hat{\sigma}_1^2}{n} + \frac{\hat{\sigma}_3^2}{m} + \frac{\hat{\sigma}^2}{nm\ell} , \tag{9}$$

where

$\hat{\sigma}_1^2$ = estimate of variance of days of analysis,

$\hat{\sigma}_3^2$ = estimate of variance of sintering runs

= 0.00001052 ,

and

$\hat{\sigma}^2$ = error mean square

= 0.00003060 .

Days of Bad Analysis Detected. — If days of bad analysis can be detected, $\hat{\sigma}_1^2$ is given by Eq. (2), and the degrees of freedom are calculated by

$$\text{d.f.} \approx \frac{[\text{Eq. (6) using } \hat{\sigma}_1^2 = 0.00000296]^2}{\frac{1}{4}\left[\left(\frac{1}{5n}\right)\frac{S'_1}{4}\right]^2 + \frac{1}{5}\left[\left(\frac{1}{6m}\right)\frac{S_3}{5}\right]^2 + \frac{1}{18}\left[\left(\frac{1}{nm\ell} - \frac{1}{5n} - \frac{1}{6m}\right)\frac{S_e}{18}\right]^2} . \tag{10}$$

Days of Bad Analysis Not Detected. — If days of bad analysis cannot be detected, $\hat{\sigma}_1^2$ is given by Eq. (1), and the degrees of freedom are calculated by

$$\text{d.f.} \approx \frac{[\text{Eq. (6) using } \hat{\sigma}_1^2 = 0.0001201]^2}{\frac{1}{5}\left[\left(\frac{1}{6n}\right)\frac{S_1}{5}\right]^2 + \frac{1}{5}\left[\left(\frac{1}{6m}\right)\frac{S_3}{5}\right]^2 + \frac{1}{18}\left[\left(\frac{1}{nm\ell} - \frac{1}{6n} - \frac{1}{6m}\right)\frac{S_e}{18}\right]^2} . \tag{11}$$

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