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YANKEE ATOMIC ELECTRIC COMPANY
RESEARCH AND DEVELOPMENT PROGRAM

PROGRESS IN THE DEVELOPMENT OF A
PROCESS FOR PRODUCING UO_2 PELLETS

R & D SUBCONTRACT No. 1 under
USAEC-YAEC CONTRACT AT(30-3)-222

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DECEMBER, 1958

WESTINGHOUSE ELECTRIC CORPORATION
ATOMIC POWER DEPARTMENT
PITTSBURGH, 30 P. O. BOX 355 PENNSYLVANIA

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Yankee Atomic Electric Company
Research And Development Program

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PROCESS FOR PRODUCING UO_2 PELLETS

Prepared by
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Nuclear Core Engineering

December, 1958

For The Yankee Atomic Electric Company
Under Research and Development Subcontract
No. 1 of USAEC-YAEC Contract AT(30-3)-222

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I. ABSTRACT

Progress in the development of manufacturing procedures and techniques for uranium dioxide fuel pellets is described. Specifications for both the uranium oxide powder and the final fuel pellet are given. A process is described, for making pellets with closely controlled density, dimensions, purity, and freedom from structural defects, from ADU (ammonium diuranate process) uranium dioxide powder. A formula relating linear shrinkage to green and fired densities has been derived. This formula has proven a useful tool in control of fired, or sintered, pellet dimensions and densities. An example of the use of this formula is given. Processes described were utilized in the manufacture of pellets for use in critical experiments. Improvements in processing continue to be developed.

II. INTRODUCTION

The uranium dioxide research and development program for the Yankee reactor is intended to develop: 1) a simplified and therefore more economical pelletization process for commercially available uranium dioxide (ADU), and 2) specifications for powder and pellets such that the fabrication process developed will yield uniform high quality uranium dioxide fuel pellets.

The program has the following categories:

1. The evaluation of various obtainable UO_2 powders to determine which can be fabricated to specified density and dimension with a minimum loss of material (i.e., from grinding, chipping, cracking, and laminating) and to prepare specifications for the powder that will provide such pellets uniformly.
2. The investigation of pelletization procedures and techniques to provide maximum quality pellets at minimum cost.
3. The examination of the UO_2 pellets so produced to ascertain their properties and behavior to enable the specification of pellets which will insure dependability of operating fuel elements.

This report indicates the progress so far made in attaining the objectives indicated above. Specifications for both the uranium dioxide powder and the final fuel pellet are attached (Section VIII). A process using ADU powder meeting the above powder specification, was successfully employed to manufacture approximately 450,000 pellets for Yankee criticality experiments. The investigation of pellet fabrication procedures to provide a simpler and less expensive process is continuing, and will be reported in more detail at a later date.

III. CONCLUSIONS

1. Powder and pellet specifications have been developed and are included as Section VIII in this report. These specifications proved satisfactory for controlling the quality of pellets now undergoing critical tests in the Yankee Critical Experiment.
2. A process for fabricating pellets for the ADU uranium oxide powder has been developed and is reported. Use of this process, in conjunction with the above powder and pellet specifications, enables the manufacture of pellets from ADU powder to closely controlled dimensional and density tolerances.
3. A formula has been developed relating shrinkage upon sintering to green and fired density. This has been proven to closely approximate observed behavior of fabricated pellets. The relationship may be expressed:

$$\% \text{ linear shrinkage (fired basis)} = \left[\left(\frac{D_f}{D_g} \right)^{1/3} - 1 \right] 100$$

where D_f = fired density

D_g = green density

IV. RAW MATERIAL

In early commercial production, UO_2 powder was largely made by the hydrogen reduction of UO_3 which, in turn was a decomposition product of uranyl hexahydrate produced from uranium ore concentrates. These hydrogen-reduced powders were made from natural uranium (i.e., containing a U^{235} content of about 0.7%) whereas the design of the Yankee reactor required enriched UO_2 .

As a result of diffusion methods of isotopic separation, enriched uranium in the form of uranium hexafluoride (UF_6) became available from the U. S. Atomic Energy Commission. Commercial producers of UO_2 have found to date that the conversion of UF_6 to UO_2 can be most economically effected by the "Ammonium Diurante Process" (ADU).

Although this process yields a UO_2 powder of high purity which can be processed into pellets satisfactory for use as nuclear fuel, problems arose (and have continued) relative to reproducibility in fabricating pellets from different batches of powder made by the ADU process.

Early in this program some ADU powders were found to sinter to over 92% of theoretical density at only 1400°C while others, supposedly identical, did not attain as much as 90% of theoretical density at 1700°C . Subsequent experience has indicated that a considerable number of powder variables may be significant in the behavior of this powder during the pellet fabrication process. The relative influence of such properties as particle size and distribution, surface area and possible activation - as by ball milling - or powder history, on such pellet parameters as green and fired density, shrinkage, or sinterability is not fully understood. As was indicated earlier, one of the objectives of this program is to develop an improved pelletization procedure. While these efforts are continuing, and are outlined in Section VII, "Progress in Development of UO_2 Pelletization", of this report, these efforts have been hindered by apparent variability in successive batches of ADU powder. While this situation continues, a better understanding of the relative significance of powder variables is necessary in order to develop a dependable improved pelletization procedure.

Insufficient information correlating properties of present ADU powders with sintering behavior exists to enable the specification of physical properties of powders which will press and sinter satisfactorily, but experience has taught various UO_2 suppliers how to produce a product which will sinter to at least 92% of theoretical density in two hours at no more than 1675°C . The present UO_2 powder specification meets this minimum sinterability problem with a performance requirement.

The problem of variability is of prime concern to pellet manufacturing, because this variability means each lot must be treated like new material. Trials must be conducted to determine:

1. The applicability of the method for preparing press feed material.
2. The pressure required to produce the desired green density.
3. The sintering conditions yielding the proper fired density.

The differences in UO_2 from lot to lot make dimensional control difficult. Further, these differences have impeded the development of an abbreviated material preparation procedure. Simple procedures which suffice for several lots of UO_2 often fail on other lots.

The problem of lot-to-lot variability has been minimized, in the specifications, by requiring larger lots so that fewer pilot runs are necessary to establish forming parameters.

V. FABRICATION OF UO_2 FUEL PELLETS

Previous investigators* had developed a process for dry pressing cylindrical pellets from hydrogen reduced UO_2 . From this procedure, a process was developed for the dry pressing and sintering of pellets from UO_2 made by the ammonium diuranate process. This process is given in Table I.

Some 450,000 pellets were made according to this process for the Yankee Critical Experiment. Fabrication information of the product variables are available from statistical analyses performed by the pellet vendor and from receiving inspection data collected at ^(W) Atomic Power Department. Summaries of the collected data are included in Section VI of this report.

* Glatzer, J., Hoge, H. R., & Schaner, B., "Fabrication of Dense Uranium Dioxide Cylindrical Compacts by Cold Pressing and Sintering", WAPD-126, October, 1955.

TABLE I

Pelletization Process

1. Ball mill as received powder for two hours in 50 weight percent water.
2. Dry the mill slurry at 80°C.
3. Granulate the mill cake to 20 mesh.
4. Mix in 1% of 100 mesh polyvinyl alcohol (DuPont 51-05, "Elvanol") in a twin shell blender or P. K. Blender (The Patterson-Kelly Co., Inc., E. Stroudsburg, Pa.).
5. Slowly add about 9% water - Aerosol solution (0.025 g. Aerosol OT per 1000 ml H₂O).
6. Partially dry.
7. Granulate to 8 mesh.
8. Dry thoroughly, 80°C.
9. Granulate to 20 mesh.
10. Blend with 0.2% of -100 mesh Stero-tex Z (A lubricant: powdered vegetable stearine, Capital City Products Co., Columbus, Ohio).
11. Press to constant green density, usually 7.2 g/cc (but see discussion in Section VI (of this report)).
12. Presinter at 800°C for 2 hours in dissociated ammonia atmosphere to volatilize organics. Place compacts on molybdenum trays.
13. Sinter in hydrogen atmosphere at 1450°C to 1700°C; 2 to 8 hours, depending upon powder properties. Pellet density 91.5 to 94.5% of theoretical density (10.03 to 10.36 gm/cc).
14. Centerless grind to 0.300 ± 0.0005 inches diameter.
15. Sample and inspect.

Material accountability was carefully controlled throughout the process. Table II shows the distribution of material processed to make the pellets for the Yankee CRX fuel rods. One goal of future investigations would be to increase the percentage of acceptable product.

Various control points set up during the manufacturing run allowed for adjustments in the process to compensate for the lot-to-lot variation encountered. A study of the problems encountered presented many areas which needed further investigation. These areas are the basis of the research and development program currently underway.

VI. DISCUSSION OF DIMENSIONAL CONTROL

The largest loss of material in the pelletizing process occurs during the centerless grinding operation (Table II), and consequently good dimensional control is required to minimize this loss. Measures used to establish stability in the process were; 1) green density 2) fired density, 3) diameter, and 4) length. An additional problem of shape deformity (hour glassing) compounded the above measures and efforts to minimize this deformity are being considered.

The control of final pellet dimension requires the following information regarding the powder performance when processed: 1) pressing pressure versus green (unfired) density, and 2) fired density under various sintering treatments. Figure 1 shows the relationship between pressing pressure and green density for one powder. Figure 2 shows the relationship of green density vs. sintered density for different sintering temperatures. The green density should be as high as practicable, but not excessive (laminae occur during sintering of pellets of too high green densities). High (not excessive) green density correlates with minimum shrinkage, and warpage, and enhances densification.

Experience has indicated that green density increases with increasing pressing pressure but varies in amounts from lot-to-lot of powder.

Green Bulk Density

The bulk density of the UO_2 was determined on a lot basis using a Scott Paint Volumeter. Table III shows the bulk density measurements on the as-received powder (Yankee CRX) and the reported vendor measurements. A bulk density test was also taken on each prepared batch prior to pressing. Prepared materials with a bulk density below 3.0 grams/cc were rejected because they would not fill the die cavity of the press and gave erratic pellet lengths. The bulk density average for each lot of prepared material is given in Table III.

TABLE II

Distribution of UO₂ Material for Yankee CRX

1. Recoverable scrap loss		
a. operations prior to pressing	0.43%	
b. pressing	0.43%	
c. presintering	0.16%	
d. sintering	0.37%	
e. centerless grinding	5.50%	
	<hr/>	6.89%
2. Unrecoverable loss		
a. unaccountable loss (portions of this material were later recovered from ductwork and dust collector)	1.80%	
	<hr/>	1.80%
3. Acceptable material	91.31%	
	<hr/>	91.31%
Total		<hr/> 100.00%

PRESSING PRESSURE VS GREEN DENSITY

FIGURE 1

GREEN DENSITY VS SINTERED DENSITY

FIGURE 2

TABLE III

Bulk Density of As-Received and Compounded UO₂ Powder

Lot	Bulk Density Received grams/cc	Mallinckrodt Chemical Works Bulk Density grams/cc	Bulk Density Before Pressing grams/cc
15	2.5	2.8	3.2
16	2.5	2.5	3.3
17	2.5	2.6	3.2
18	2.5	2.6	3.2
19	1.8*	1.9	**
20	2.4*	2.6	**
21	1.9*	1.8	**
22	2.3*	2.5	**
28	2.4	2.6	3.2
46	2.4	2.6	3.0
64	2.3	2.5	3.1
76	2.4	2.5	3.1
77	2.3	2.4	3.0
78	2.3	2.4	3.0
79	2.7	2.4	3.4
80	2.3	2.4	3.0

*Material returned for blending into Lot 78

**Returned to vendor - not processed

Table IV shows the comparison of the desired green density and the actual average green density for 18 batches* representing 4 lots of ADU UO₂ powder processed for the Yankee CRX. Within a batch, the green densities were constant, but the variance from batch-to-batch and lot-to-lot was significant. The existence of these variables presents difficulties in predicting final dimensions. An attempt was made to establish a quantitative relationship between: 1) green and fired densities, and 2) green and fired pellet diameters.

Figure 3 shows a plot of the relationship found between green density and fired pellet diameter. This relationship may be expressed by the following formula:

A. Diameters

$$\frac{dg}{df} = \left(\frac{Df}{Dg}\right)^{1/3} \quad \text{where}$$

dg = green dimension

df = fired dimension

Df = fired density

Dg = green density

B. Percent of linear shrinkage (on fired basis)

$$\left(\frac{dg - df}{df}\right) 100 = \left[\left(\frac{Df}{Dg}\right)^{1/3} - 1\right] 100$$

A more exact correlation with experimental data may be obtained by applying a correction factor for loss in weight or mass during sintering. In the preceding formulae an assumption is made that mass does not change. In actual practice, however, volatilization of organics and other weight changes do occur to an extent that the use of a correction factor of 0.98 times green density insures a closer relationship. Also, in certain powders, anisotropic shrinkage on sintering has been observed.

* A lot is defined as that material designated by the powder supplier as being processed at one time. A batch is a subdivision of the lot of such amount to be processed at the pellet manufacturers in the available equipment and is sized with safety considerations in mind.

TABLE IV

Desired Green Density vs. Average Actual Green Density

Lot	Batch	Sintering Temperature (°C)	Desired Green Density (gm./cc)	Actual Average (gm./cc)
64	12	1610	7.4	7.39
	14	1610	7.4	7.45
	15	1610	7.4	7.42
	16	1610	7.4	7.40
	18	1610	7.4	7.36
	19	1610	7.4	7.46
76	2	1610	7.4	7.42
	3	1610	7.4	7.46
	4	1610	7.4	7.51
	5	1610	7.4	7.44
	8	1625	7.2	7.29
77	1	1645	7.2	7.24
	2	1645	7.2	7.18
	12	1635	7.2	7.21
	13	1645	7.2	7.19
	14	1645	7.2	7.25
78	1	1600	7.2	7.19
	2	1600	7.2	7.17

GREEN DENSITY VS FIRED DIAMETER

FIGURE 3

The diameter formula (1) may be used to select the proper die to be used to press pellets to a particular diameter (the green density is chosen, sintered density is specified, and the fired diameter is established). For example: a 0.3" diameter pellet 0.6" long having a fired density of 10.19 gm/cc (93% of the theoretical) is required. The powder is known to behave like that in Figure 1 and 2. A green density of 8.0 g/cc can be selected and would be attained by pressing to 85 TSI (Figure 1). The resulting pellets could be sintered at 1610°C for three hours to attain the specified density (Figure 2). Several additional factors should be then taken into account before final selection of the proper die size. A maximum of 0.0050" of hour glassing* occurs. Therefore a first approximation would be for a 0.3050" diameter at the pellet ends. Taking into account that the minimum diameter occurring would be approximately 0.0020" less than the average found, the final fired diameter of the pellet (worst case) desired is equal to 0.3070". However, this would result in a larger loss due to grinding the pellet to final size. Therefore, a compromise is effected by the selection of an average sintered diameter as measured at the pellet ends of 0.3060".

Substituting in formula (1)

$$\frac{dg}{df} = \left[\frac{Df}{Dg \cdot 0.98} \right]^{1/3}$$

$$dg = 0.3060 \left[\frac{10.19}{8 \times 0.98} \right]^{1/3}$$

$$dg = 0.3340"$$

It is known from experience that pellets expand approximately 0.0040" upon release from the die; consequently, the die size may be reduced by this amount. The final die diameter then selected would be 0.330" so that green pellets having a diameter of 0.334" will be obtained, in order to produce sintered pellets with a 0.300" diameter after grinding.

*Because the centers of pellets press to lower green density than the ends, the center of the pellet shrinks more than the ends in sintering. The difference between the diameter at the ends and that at the center is known as hour-glassing.

Density Variation

Table V presents fired density data collected during the manufacture of Yankee CRX pellets. Randomly selected pellets were weighed and measured from each of three sintering trays* from 18 random process batches taken from four lots. Densities were calculated and averaged for each tray. The overall average was 93% of the theoretical, with individual pellets ranging between 90.00% and 95.00% and 95% of the values within $93 \pm 1.5\%$ of the theoretical density.

Three methods of determining densities were studied. Randomly selected pellets were measured and weighed. A nomograph was constructed to give direct readings of densities from the measurements. The data were used to calculate the densities of the same pellets. Table VI is a comparison of the results of the two methods of determining density.

A method to determine the density by the displacement of water was used. The values obtained are compared to calculated values for the same pellets (Table VII).

Figure 4 is the distribution curve for the data collected for three methods of determining densities during Yankee CRX pellet inspection.

Length Variation

Information relative to length variation of sintered pellets is available from: 1) statistical analysis of the vendors' data and 2) (W) Atomic Power Department's receiving inspection reports. Finished pellets were received packaged in about 500 pellet "batches", each of which was samples. Length measurements for 6783 pellets from approximately 450,000 received are summarized in Table VIII.

Figure 5 is the distribution curve for length measurements made during receiving inspection.

Pellet Distortion (Hour-Glassing)

Four lots of UO_2 processed for the Yankee CRX core revealed that the overall average of hour-glassing was 0.0026". The range of averages was from 0.0023" to 0.0028". The range of measurements for 95% of pellets samples was 0.0003" to 0.0049".

Diameter Control

Pellets were ground on a Cincinnati No. 2 centerless grinder using two passes to insure a smooth surface finish. The grinding wheel used was a type 6-C-80-H6-UP from the Norton Company.

* Pellets are sintered in molybdenum trays, 1000 to 1500 per tray.

TABLE VCalculated Pellet Densities For Each Tray

<u>Lot</u>	<u>Batch</u>	<u>Tray</u>	<u>Average Density</u> <u>% Theoretical</u>
64	12	1	93.22
		2	91.62
		3	91.62
	14	1	Not Sampled
		2	94.088
		3	93.39
	15	1	93.34
		2	90.89
		3	91.88
	16	1	92.41
		2	92.43
		3	92.73
	18	1	92.71
		2	92.90
		3	92.76
	19	1	93.47
		2	93.38
		3	93.24
76	2	1	91.92
		2	91.49
		3	92.03
	3	1	92.25
		2	92.23
		3	92.45
	4	1	93.12
		2	92.79
		3	92.48
	5	1	93.14
		2	93.87
		3	92.84
	8	1	91.74
		2	91.92
		3	91.77

TABLE V (Continued)

<u>Lot</u>	<u>Batch</u>	<u>Tray</u>	<u>Average Density</u> <u>% Theoretical</u>
77	1	1	92.86
		2	92.01
		3	92.57
	2	1	93.20
		2	92.98
		3	93.49
	12	1	93.90
		2	94.01
		3	93.60
	13	1	92.38
		2	92.55
		3	91.86
	14	1	93.28
		2	93.50
		3	93.17
78	1	1	94.30
		2	94.02
		3	94.43
	2	1	94.53
		2	94.79
		3	94.51

TABLE VI

Nomograph vs. Calculated Pellet Density Data (% Theoretical)

<u>Method</u>	<u>Nomograph</u>	<u>Calculated</u>
Maximum	95.25	95.25
Minimum	90.75	90.25
Range	4.50	5.00
Average	92.97	93.08
Root mean square deviation	1.03	0.83
Sample Size	408 pellets	408 pellets

TABLE VII

Calculated vs. Wet Density Pellet Data (% Theoretical)

<u>Method</u>	<u>Calculated</u>	<u>Wet</u>
Maximum	95.25	94.25
Minimum	91.25	90.75
Range	4.00	3.50
Average	93.02	93.09
Root mean square deviation	1.14	1.64
Sample Size	58 pellets	58 pellets

CRX PELLET INSPECTION

FIGURE 4

Distribution Curve of Pellet Densities

TABLE VIII
Pellet Length Data

Length Requirements = 0.550" to 0.650"	
Maximum	0.6850"
Minimum	0.5250"
Range	0.1600"
Average	0.5989"
Root mean square deviation	0.0034

Distribution Curve of Pellet Lengths

CRX PELLET INSPECTION

FIGURE 5

The unground diameter (center measurements) averaged 0.3078" and ranged from 0.3020" to 0.3136". The pellets were ground to 0.3000" $\pm$ 0.0005". Figure 6 is the distribution curve of diameter measurements from receiver inspection reports for the 450,000 Yankee CRX pellets.

A summary of the results of these diameter measurements is given in Table IX.

VII. PROGRESS IN DEVELOPMENT OF UO₂ PELLETIZATION

As mentioned, previous investigators had developed a process for dry-pressing UO₂ pellets. Initial development adapted this process to ADU UO₂ (i.e., UO₂ made from UF₆ by the ammonium diuranate process), and subsequent efforts have been directed toward reducing pelletization costs by the following methods:

- A. Simplification of the process by reduction in number of material preparation operations and elimination of pre-sintering.
- B. Improvement in unground dimensional control and reduction in losses by improved process control.
- C. Improving pellet specifications.

None of the investigations is complete at this time. The following is a summary of the status of the program.

Under the process simplification program, samples have been prepared using methods that reduce the number of material preparations required. Early efforts indicated that ball milling improved the sinterability of the UO₂. Figure 7 shows sintered densities plotted against sintering temperature for material ball milled and not ball milled.

It is more desirable to sinter at moderate temperatures because furnace maintenance time is reduced. Consequently, the ball milling operation was included in the revised process to lower sintering costs and to provide added assurance that any UO₂ powder could be processed to the required density.

Confirmation has been obtained that ball milling increases sintered density and produces a more uniform fine-grained fracture surface (as opposed to the grainy rough fracture surface apparent on the non-ball milled product). Further, sintered compacts of ball milled material appear to shrink more isotropically, (i. e., to a more uniformly circular cross section).

Distribution Curve of Pellet Diameters
CRX PELLET INSPECTION

FIGURE 6

TABLE IX

Pellet Diameters (inches)

Maximum	0.3005
Minimum	0.2993
Range	0.0012
Average	0.3000
Root mean square deviation.	0.0002
Sample Size	6894

SINTERED DENSITY VS SINTERING TEMPERATURE AND BALL - MILLING

FIGURE 7

Pellets have been or are being processed under the following conditions:

1. no ball milling
2. dry milled
3. wet milled in dilute H_2SO_4
4. milled with different binders and lubricants

Completion of these experiments is expected to result in sufficient data to insure a substantially simplified pelletizing process.

The present pelletization process (Table I) is long, and involves many operations, some of which are repetitious. Samples have been produced by varying the processing schedules. Three of the experimental procedures used are:

- 1a. ball mill wet for 2 hours
- b. dry, granulate
- c. mix with 0.3% Sterotex
- d. press
- 2a. wet ball mill for 2 hours
- b. dry
- c. granulate
- d. rewet with a 2% Carbowax 20 M water solution
- e. granulate
- f. dry
- g. press
- 3a. wet ball mill a 2% solution of carboxymethyl cellulose
- b. dry
- c. granulate
- d. press

These early promising shortened procedures, when run on production equipment failed to show batch-to-batch reproducibility. The following subsequently developed procedure has been successful a number of times on various types of UO_2 powder:

1. ball mill, wet x 2% Carbowax 20 M + a 1% PVA 51-05
2. dry
3. granulate
4. press

Confirmation trials are planned as part of the present program. This sequence eliminates two drying and two granulation operations and the separate additions of the PVA binder and the lubricant.

Under the simplification program early trials have indicated that presintering (to drive off the added organics) may be unnecessary. A systematic study on a production basis has been planned.

VIII. SPECIFICATION OF POWDER AND PELLETS

The compositional portion is based upon experience gained in making pellets for the Yankee criticality experiment. The impurity values should give the pelletizer no difficulty, and yet are low enough to satisfy design requirements. Cations such as Al, C, Cd, Fe, Mo, and Si are likely to be picked up in pelletization, and therefore are checked and specified for control purposes. As, B, and Cd are specified as being poisons which are so likely to be picked up as to justify the expense of their determination. Ca, Si, and Ti are specified to discourage their addition as sintering aids, until their effects have been studied further.

Table X is a comparison of chemical analyses (averages of twelve lots) of UO_2 powder and the pellets made from these lots for the Yankee CRX.

Table XI is a UO_2 powder specification that was developed during this program.

TABLE X

Chemical Analyses:*Comparison of UO₂ Powder With Resultant Pellets

<u>Element</u>	<u>Powders</u>	<u>Pellets</u>
U	87.8%	88.1%
Ag	0.1 ppm	0.1 ppm
Al	4.5	78.0
B	0.3	0.5
Bi	0.5**	0.5**
Ca	0.6	0.4
Cd	0.4	0.5
Co	1.8	2.0
Cr	31.0	63.0
Cu	1.7	3.8
F	40.0	N.D.
Fe	112.0	610.0
In	1.4	1.2
Mg	7.1	6.5
Mn	1.9	5.1
Mo	1.4	1.2
Ni	95.0	166.0
Pb	2.3	0.7
Si	15.0	202.0
Su	1.0	0.8
Ti	5.2	4.3
U	0.8	0.7
W	20.0	20.0
Zn	15.8	13.3
C	N.D.	7.0

*Average of twelve lots

**Less than

N.D. - not determined

TABLE XI

UO₂ POWDER SPECIFICATIONS

1. No change shall be made in the quality of successive shipments of material furnished under this specification without first obtaining the approval of the purchaser.

Manufacture

2. Blending:
 - 2.1 UF₆ from different cylinders may be blended provided the manufacturer furnishes a statement as to the amounts of UF₆ blended and the isotopic content of each portion in the blend.
 - 2.2 The uranium dioxide powder shall be blended; the manufacturer shall furnish a statement of (1) the origin of the material, (2) proportions used, and (3) the isotopic content of each portion in the blend.
 - 2.3 Unless otherwise agreed upon, whenever the quantity ordered permits, the uranium dioxide powder shall be blended into lots of not less than 500 lbs.

3. Yield Guarantee:

Unless otherwise approved or specified by the purchaser, the manufacturer shall furnish to the purchaser, in the form of uranium dioxide powder conforming to this specification, at least 99 percent of the total uranium received in the form of UF₆. Upon completion of the order, the supplier shall furnish the purchaser a statement describing the quantity and conditions of all unshipped uranium bearing material. Based on this statement, the purchaser shall arrange for disposition of this material.

Chemical Properties and Tests

4. Chemical Composition:
 - 4.1 The uranium dioxide powder shall conform to the following requirements as to chemical composition:

TABLE XI (CONTINUED)

UO ₂ POWDER SPECIFICATIONS

Major Constituents:

Uranium, minimum

87.6 Weight Per Cent

Impurities:

<u>Element</u>	<u>Impurity Max, PPM</u>	<u>Element</u>	<u>Impurity Max, PPM</u>
Ag	0.3	Fe	150
Al	20	In	2
B	0.6	Mo	100
Ca	75	Ni	200
Cd	0.5	Si	100
Cr	75	Ti	40
F	500	W	50

- 4.2 Lots containing one or more of the listed impurities in an amount in excess of the specified maxima will be accepted if the total macroscopic thermal neutron absorption cross section of all impurities listed below is less than 0.0007 cm^{-1} and provided the average of lots previously shipped on the same order is 0.0004 cm^{-1} , or lower. The macroscopic cross-section is computed with the following formula, using the cross section values tabulated below.* The tabulation also includes the cross section imparted by the specified level of each impurity, to enable the calculation to be made by the use of a simple ratio.

$$\sum_a = \left[\frac{(10.2) (\text{PPM})}{10^6} \right] \left[\frac{6.02 \times 10^{23}}{A} \right] [\sigma_a] [10^{-24}]$$

Where:

$\sum_a$ - is the total macroscopic thermal neutron absorption cross section in cm^{-1} imparted to a cubic centimeter of UO₂ pellet of 10.2 g/cc density.

* Except for the absorption cross section values for Cd and Ag, which are adjusted for resonance absorption effects, the values are from the "KAPL Chart of the Nuclides" April, 1956.

TABLE XI (CONTINUED)

UO ₂ POWDER SPECIFICATIONS

PPM - is the parts per million of the impurity imparting this $\sum a$.

6.03×10^{-23} - Avogadro's number, atoms per mol.

A - is the atomic weight of the impurity, grams per mol.

σ_a - is the thermal neutron absorption cross section of the impurity, in barns per atom.

Element	PPM	σ_a	Macroscopic Cross Section Imparted by this Level of Impurity, cm^2/cm^3
Ag	0.3	90	0.15×10^{-5}
Al	20	0.23	0.10×10^{-5}
B	0.6	755	25.8×10^{-5}
Ca	75	0.43	0.51×10^{-5}
Cd	0.5	5070	14×10^{-5}
Cr	75	3.1	2.8×10^{-5}
Fe	150	2.5	4.1×10^{-5}
In	2	190	2.0×10^{-5}
Mo	100	2.5	1.6×10^{-5}
Ni	200	4.6	9.6×10^{-5}
Si	100	0.13	0.28×10^{-5}
Ti	40	6	3.1×10^{-5}
W	50	18	3×10^{-5}
			<hr/>
			67.9×10^{-5}

The powder producer will not be held responsible for excess impurities if a corresponding excess of impurities was present in the UF₆ he received. In general, the powder producer is to make no deliberate additions of extraneous material, and his processing may increase no impurity found in the UF₆ by more than 50%. The basis is to be the UF₆ analysis, of available, or the analysis of the first hydrolyzed UF₆ solution.

TABLE XI (CONTINUED)

UO ₂ POWDER SPECIFICATIONS

5. Analysis:

5.1 An analysis of each lot shall be made by the manufacturer to determine conformance with the requirements specified in Section 4.

5.2 The composition thus determined shall be reported to the purchaser or his representative on the same day the powder is shipped.

6. Check Analysis:

Check analysis and cross section calculations may be made by the purchaser on material representing each lot. The composition and cross section thus determined shall conform to the requirements specified in Section 4. Failure of the check analysis or calculations to conform to the requirements in Section 4 shall be cause for rejection of the entire lot from which the sample was taken.

7. Isotopic Assay:

7.1 The weight percentage of total uranium which is to be in the form of isotope U-235 shall be specified on the Purchase Order. For example, an order for 5.4 percent enriched uranium dioxide means that 5.4 percent of the total weight of uranium in the uranium dioxide is to be contributed by isotope U-235.

7.2 The uranium shall not be depleted in conversion; the manufacturer shall be responsible for delivering UO₂ of the same enrichment as the UF₆ received.

7.3 The manufacturer shall furnish the purchaser the isotopic analysis calculated from data on AEC Form 388 for each lot.

Physical Properties and Tests

8. Density:

8.1 The bulk density of the powder shall be 2.2 to 2.7 grams per cubic centimeter.

TABLE XI (CONTINUED)

UO ₂ POWDER SPECIFICATIONS

- 8.2 The density shall be measured with a Scott Paint Volumeter using an 8 mesh screen without tapping, tamping, or jarring.

9. Sinterability:

The sinterability of the powder shall be such that after no more than 2 hours of wet ball milling the powder will press, at a pressure not exceeding 75 tons per square inch, into pellets which will sinter to a density of not less than 10.1 grams per cubic centimeter in not more than 2 hours in hydrogen or dissociated ammonia at a temperature not exceeding 1675°C (3050°F).

Dimensions

10. Particle Size:

The particle size shall be such that 100 percent of the powder will pass through a 10 mesh Tyler screen and 95 percent or more will pass through a 20 mesh Tyler screen with no encouragement other than the use of a Ro-Tap or a Syntron Vibrator.

11. Test Reports:

The manufacturer shall furnish 3 copies of a certified test report showing the following:

1. Chemical composition
2. Isotopic assay
3. Blending data when blended lots are furnished
4. Density
5. Sinterability
6. Purchase Order Number
7. P D Spec Number ((W) A. P. D. Purchasing Department Specification Number)
8. Lot number correlating the test report with the lot represented.
9. A statement that the powder conforms to the specification.
10. Following shipment, a full inventory statement of materials retained or lost in the processing by the manufacturer.

Packing and Marking

12. Packing:

The material shall be properly packed to prevent loss and contamination during shipping and to conform with all pertinent regulations. The material and size of the smallest inside container shall be as specified on the Purchase Order.

TABLE XI (CONTINUED)

UO ₂ POWDER SPECIFICATIONS

13. Marking:

Each drum shall be plainly marked as follows: Purchase Order Number; Uranium Dioxide Lot Number; Batch Number; Drum Number; Gross, Tare, and Net Weight, Name of Manufacturer.

Table XII is a UO_2 pellet specification that has been developed during this program.

TABLE XII

UO_2 Pellet Specification

1. Introduction

This specification applies to sintered UO_2 fuel pellets for pressurized water reactors.

2. Composition

- a. Isotopic - The UO_2 must suffer no depletion in pelletization. The isotopic content of pellets will be exactly that of the powder received by the pelletizer from the powder producer.
- b. Chemical - U minimum 87.7%
 U^{+6} maximum 2.0%

	<u>ppm</u>		<u>ppm</u>
Ag	0.3	Cd	1
Al	300	Fe	1000
B	2	Mo	100
C	200	Si	500
Ca	100	Ti	50

Note: No materials are to be added to the UO_2 except small quantities of organic binders and/or lubricant needed for pelletization.

3. Dimensions

As specified on the drawing which will accompany this specification.

4. Density

$93 \pm 1.5\%$ of theoretical

100% of theoretical density is 10.96 g/cc

Percent Theoretical Density

Equivalent grams per cubic centimeter

90	9.86
91	9.97
91.5	10.03
92	10.08
93	10.19
94	10.30
94.5	10.36
95	10.41
96	10.52
97	10.63
98	10.74
99	10.85

5. Appearance

- a. Pellets should be substantially free of chips and cracks, and must be handled and shipped to minimize these defects. Chipped pellets may not have lost more than 10% of the area of either end of the pellet, and the chip may not extend more than 10% of the length of the pellet.

The purchaser will supply a visual standard for chipped cracked pellets to help the vendor distinguish unacceptable from acceptable pellets having this defect.

- b. Squareness of Ends

The squareness of each end of the pellet will not deviate from the axis of the pellet by more than 0.005".

6. Inspection

- a. Pellets will be accepted or rejected on a batch basis.
b. Each batch will be sampled and measured statistically.
c. Each batch of pellets must conform to a 99% certainty that there is no more than 1 defect per 100 diameters and must conform to a 95% certainty that there are no more than 2 defects per 100 for length, weight, density, and squareness.

7. Yield

- a. The vendor will deliver to Westinghouse Atomic Power Department, in the form of acceptable pellets, as much of the UO_2 received as is practical, and guarantees at least 90% yield.
b. Unaccountable losses of UO_2 may not exceed 2.0% of the UO_2 received.

- c. The vendor will account to the purchaser, as soon as is practical, for all of the received UO_2 which has not been delivered to the purchaser in the form of acceptable pellets, as specified in 7, a. above. The purchaser may request that scrap pellets or powder, which might not produce pellets meeting this specification, be made into pellets (at a price to be negotiated) or the purchaser may instruct the pellet manufacturer to ship the scrap and residues to the AEC, a reprocessor, or to the purchaser, with the purchaser paying freight charges.

8. Packaging and Marking

- a. Because UO_2 is brittle, care must be taken in packaging to minimize breaking, chipping, and cracking.
- b. Packaging must conform to pertinent I.J.C. and AEC regulations regarding nuclear materials.
- c. Each container shall be individually and consecutively numbered and shall be labeled to show net weight of pellets, UO_2 lot numbers from which pellets were made, and certification of inspection.

9. Compliance with Regulations

The vendor must have appropriate licenses and clearances indicating that he is aware of and responsible for complying with all applicable regulations of Federal, State, and local regulatory bodies with respect to receiving, accounting for, processing, storing, and shipping of UO_2 powder and pellets.