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ATOMIC ENERGY ACT
OF 1954

WANL-TNR-092
February 26, 1963

DEVELOPMENT OF BATCH
FORMULATION EQUATIONS
FOR GRAPHITE FUEL ELEMENTS
(Title Unclassified)

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DEVELOPMENT OF BATCH FORMULATION

(Title L)

SUMMARY

The development of equations used for determining starting weights of the individual mix batch constituents is presented. Experimental data resulting from batches compounded in this manner are shown for uranium loading, carbon density, and composite density of as graphitized natural uranium loaded fuel elements.

INTRODUCTION

Of all the individual process steps which can affect the final uranium content of a finished fuel element, certainly the initial compounding and mixing of the individual constituents represents the area of greatest possible error. It was recognized that a need for accurately determining starting weights of all batch constituents existed.

In addition, it was determined that variations in uranium content and particle density of the PyCUC₂ beads could cause variations in uranium loading and carbon density which would exceed design specifications.

DISCUSSION OF DATA

The batch formulation employed by LASL at the time that WANL entered the area of uranium bearing graphite fuel elements was based on the C plus UO₂ system. The following equations were used for calculating the starting batch ingredient quantities:

$$1. \text{ Weight \% UO}_2 = \frac{\text{Wt. UO}_2}{\text{Wt. DCI} + \text{Wt. UO}_2} \times 100$$

$$2. \text{ Weight of Catalyzed Binder} = X (\text{Wt. DCI} + .127 \times \text{Wt. of UO}_2)$$

where X = the desired weight fraction of binder to be added to the batch.

A loading curve was experimentally determined by processing a number of batches calculated as shown above. The procedure was to start with a known quantity of UO₂, dry carbon ingredients (DCI), and binder, process the resulting extrusion through graphitizing and determine the final U/cc by wet analysis. In this way, a curve could be drawn showing Weight % U versus mg/cc bading. This curve would then be used to calculate batches of any loading within the range of the loading curve.

With the introduction of coated UC₂ beads as fuel, it was our intention to devise a method for compounding the necessary batch ingredients that would be flexible enough to factor

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ELEMENTS

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in variations in starting materials thus reducing the number of batches required to establish a "loading curve". A second objective was to devise a method which would produce loadings in the "as graphitized" condition within +2%, -3% of the desired loading.

At the outset of the process familiarization work at AFF, limited data was available concerning PyCUC₂ bead loaded fuel elements. It was known that final fuel loading was determined by the following method.

1. $\text{Weight \% U} \times \text{Composite Density} = \text{Loading in mg/cc.}$

The carbon density was then determined by subtracting the uranium loading from the composite density.

In order to compound the initial fuel bearing batches, it was necessary to estimate the carbon density for a given fuel loading. Using the estimated "carbon density" the following equations were developed.

- I. To determine the weight fraction of uranium in the "as graphitized" condition, we assumed a loading (A) (g/cc) and a final "carbon density" B (g/cc).

Therefore; (Eq. I)
$$\frac{A}{A + B} = C = \text{weight fraction of uranium in the "as graphitized" condition}$$

- II. To determine the weight fraction of pyrocoated uranium dicarbide required to yield a loading "A" (Equation I), we assumed a weight fraction of uranium in pyrocoated uranium dicarbide equal to D

Therefore; (Eq. II)
$$\frac{C}{D} = E = \text{weight fraction of pyrocoated uranium dicarbide in the "as graphitized" element}$$

Combining Equations I & II

(Eq. III)
$$E = \frac{A}{D (A + B)}$$

- III. To determine the total batch weight (carbon ingredients and pyrocoated uranium dicarbide), we assumed a pyrocoated uranium dicarbide starting charge weight equal to F. The assumption was that any changes which might occur would have little effect on the chemical characteristics of the starting batch constituents, i.e., DCI and pyrocoated uranium dicarbide and therefore, would not disturb the carbon to uranium ratio.

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Therefore; (Eq. IV) $\frac{F}{E} = G = \text{Total carbon and uranium dicarbide weight.}$

Substituting the value for E in Equation III, we could then define the total carbon plus uranium dicarbide weight by the expression -

$$\text{(Eq. V)} \quad G = \frac{(F) (D) (A+B)}{A}$$

- IV. To determine the amount of carbon which must be added in the form of DCI plus the carbon contribution made by the binder, we subtracted the starting charge weight of pyrocoated uranium dicarbide F from the total carbon plus uranium dicarbide weight as calculated in Equation V.

Therefore; $G - F = H = \text{Total carbon added to the batch in the form of DCI plus binder carbon.}$

Substituting the value for G in Equation V, we defined the total carbon addition weight as

$$\text{(Eq. VI)} \quad \frac{(F) (D) (A+B)}{A} - F = H$$

- V. The total carbon addition to the batch exclusive of carbon supplied by the fuel beads, was made up of DCI and binder carbon.

Therefore; the value of H (Eq. VI) could be written:

$$H = K + M \left(K + .127 (F) \right)$$

where K would represent the DCI weight and M $\left(K + .127 (F) \right)$ represented the LASL equation for calculating the catalyzed binder required for a DCI weight of K and a fuel weight of F. However, only a part of the total binder charge remains as carbon in the "as graphitized" element, making it necessary to express the binder carbon contribution as

$$(L) (M) \left(K + .127 (F) \right)$$

where L represents the weight fraction of the starting binder weight which remains as carbon in the "as graphitized" element.

The expression for H could then be written as

$$H = K + (L) (M) \left(K + .127 (F) \right)$$

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Substituting Equation VI and solving for K the batch make up was expressed as Eq. VII below:

$$\frac{\left(\frac{(F)(D)(A+B)}{A} - F \right) - 0.127 (L)(M)(F)}{1 + (L)(M)} = K = \text{Starting DCI wt.}$$

- VI. To determine the starting catalyzed binder weight we assumed that the binder carbon was equal to the total batch carbon (exclusive of carbon supplied by the pyrocoated uranium dicarbide) minus the carbon weight represented by the DCI divided by the weight fraction of the binder which remains as carbon in the "as graphitized" condition. This is expressed as

$$\frac{H - K}{L} = P = \text{Starting binder weight}$$

Substituting Eq. VI for H, Eq. VIII was written,

$$\frac{\left(\frac{(F)(D)(A+B)}{A} - F \right) - K}{L} = P = \text{Starting binder weight.}$$

It should be noted that the value of "B" in Equation VII and VIII was an estimated value for the "carbon density" in the "as graphitized" element and as such represented the greatest source of error in batch calculations. As more data became available, a curve similar to Figure 1 was developed making it possible to select a value for "B" for any loading. However, this curve constituted a need which could be considered as similar to a "loading curve" and in essence was contrary to the objective to find a method which would be flexible enough to mathematically compensate for starting material variations.

A total of eleven production sized fuel bearing batches were calculated using Equations VII and VIII and were processed through graphitizing. The results are shown in Table I. As these data indicate, the deviation between "aim" loading and actual loading ranged from - 1.6% to 1.57% for all batches except Batch 158 which showed a deviation of + 3.04%.

Prior to the time that Batch 158 was produced, all batches had been made using PyCUC₂ beads supplied by 3M. These beads had ranged from 4.401 g/cc to 4.584 g/cc and 69.05 to 71.39% Uranium. As we have since found, this variation in density and chemistry was not large enough to produce any great variation from batch to batch with respect to "aim" and actual loading. However, with the introduction of beaded particles supplied by NUMEC, we found that batches calculated and processed

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in an identical manner to the batches containing 3M material showed a considerable difference between "AIM" and actual loading and "AIM" and actual composite densities. The only obvious difference between batches made from 3M material and batches made from NUMEC material was the difference in PyCUC₂ particle density. The average 3M material density was 4.493 g/cc while the NUMEC material density was 5.362 g/cc. This indicated that where previously the PyCUC₂ particle density was not considered, a correction factor was needed for calculation of the batch make-up.

This was further evidenced by the fact that a batch compounded to give a 348 mg/cc loading would result in "carbon densities" of 1.791 g/cc for 3M material of 4.584 g/cc density and 71.39% U as opposed to 1.825 g/cc for NUMEC material of 5.362 g/cc density and 70.59% U. (Batches 145 and 158 respectively - Table I)

A correction factor which included the PyCUC₂ bead density was devised as follows:

1. Assume a unit volume = 1 cc
2. Assume a fuel loading = A g/cc
3. Assume the chemical analysis of PyCUC₂ = B % Uranium
4. Assume a measured density of PyCUC₂ = D g/cc
5. Assume the density of an unfueled piece processed in the same manner as a fueled piece as 1.86 g/cc. (This was established experimentally.)
6. Assume that no reaction occurs between the PyCUC₂ beads and the matrix materials (DCI and binder) which would change the chemistry of any of the mix constituents. (It has been determined that the binder charge in the mix will lose 51 weight per cent as volatiles during thermal treatment.)

Therefore:

$$\text{Eq. IX} \quad \frac{A}{B} = \text{Weight of PyCUC}_2 \text{ beads expressed as grams in one cc of a graphitized fuel element of loading "A".}$$

$$\text{Eq. X} \quad \frac{A}{B} \div D = \frac{A}{(B)(D)} = \text{Volume occupied by the PyCUC}_2 \text{ beads in one cc of graphitized fuel element.}$$

$$\text{Eq. XI} \quad 1 - \frac{A}{(B)(D)} = \text{Volume of carbon (DCI + binder carbon) in one cc of graphitized fuel element.}$$

$$\text{Eq. XII} \quad 1.86 \left(1 - \frac{A}{(B)(D)} \right) = \text{Weight of carbon (DCI + binder carbon) in one cc of graphitized fuel element.}$$

$$\text{Eq. XIII} \quad 1.86 \left(1 - \frac{A}{(B)(D)} \right) + \frac{A}{B} = \text{Weight of carbon (DCI + binder carbon) plus the weight of PyCUC}_2 \text{ in one cc of graphitized fuel element. This is the composite density of a graphitized fuel element of loading A.}$$

$$\text{Eq. XIV} \quad \left(1.86 \left(1 - \frac{A}{(B)(D)} \right) + \frac{A}{B} \right) - A = \text{Carbon Density}$$

The expression for composite density shown as Eq. XIII above was substituted for the quantity (A + B) in Eq. VII and VIII. The equations were simplified to give the following:

$$\text{Eq. XV} \quad \frac{F \left(1.86 \left(\frac{B}{A} - \frac{1}{D} \right) - (L)(M)(N) \right)}{1 + (L)(M)}$$

$$\text{Eq. XVI} \quad \frac{F \left(1.86 \left(\frac{B}{A} - \frac{1}{D} \right) \right) - K}{L} = P$$

Where:

- A = Loading in g/cc required in the final as graphitized fuel element.
- B = Weight fraction uranium in the PyCUC₂ beads
- D = Density of PyCUC₂ beads
- F = Starting weight of PyCUC₂ beads assigned on the basis of charge size desired.
- L = Weight fraction of binder remaining as carbon in the graphitized fuel element.
- M = Weight fraction of starting binder.
- N = Constant = 0.127
- K = Starting weight of DCI blend
- P = Starting weight of catalyzed binder
- 1.86 = Density of unfueled graphite graphitized at 1900°C - 9-hour hold.

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Equations XV and XVI were used to calculate sixteen production sized fuel batches in which PyCUC₂ beads from four vendors were used. The beads varied in uranium content from 64.92% to 73.79% with a density range of 4.288 to 5.7059 g/cc.

Table II shows the data which resulted from processing these sixteen batches through graphitizing. As these data indicate, the actual loading varied from the "aim" loading from + 1.22% to -0.98%. In addition, the data indicates that we are able to predict the composite density (i.e., carbon density + uranium loading) within $\pm 0.70\%$.

A significant point which was brought out by processing the fueled batches shown on Table I and II was the need for a specified range of fuel particle density. From the standpoint of core loading design, obviously the most desirable condition is one in which the spread in carbon density from element to element is minimized. Figure 2 is a plot of carbon density versus PyCUC₂ density for a constant PyCUC₂ uranium content of 70.0% as calculated from Eq. XIV. As this plot indicates, the carbon density for a 400 mg-U/cc loading could vary from 1.766 g/cc to 1.854 g/cc if PyCUC₂ containing 70% U varied in particle density from 4.0 g/cc to 6.0 g/cc.

The validity of the calculated values for loading, carbon density, and composite density is illustrated by the data shown in Table II. It follows, therefore, that a family of curves which would define required uranium and density characteristics of PyCUC₂ versus carbon density for any given fuel loading can be presented. Figure 3 is such a plot for a 400 mg-U/cc loading. As Figure 3 indicates, in order to produce carbon densities in the range of 1.80 - 1.84 g/cc with PyCUC₂ containing $70.0 \pm 2\%$ U, the fuel beads would have to have densities in the range of 4.83 g/cc to 5.253 g/cc. This type plot could be calculated for each fuel element loading and used as a process control tool.

CONCLUSIONS

- I A series of equations have been developed which can be used to calculate starting batch ingredient quantities. Batches calculated in this manner have exhibited "as graphitized" fuel loadings within a range of +1.75% to -0.98% of nominal. Similarly, the composite "as graphitized" densities have been within a range of $\pm 0.70\%$ of nominal.
- II The starting PyCUC₂ bead chemistry and density can be used to predict the chemistry, carbon, and composite densities of fueled graphite extrusions.

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TABLE I

(A)			(B)		(A+B)			
Batch No.	Aim Load (g/cc)	Actual Load (g/cc)	%Deviation Aim-Actual Loading	Aim Carbon Density (g/cc)	Actual Carbon Density (g/cc)	Aim Composite Density (g/cc)	Actual Composite Density (g/cc)	%Deviation Aim-Actual Composite Density
144	400	393.6	-1.60	1.797	1.779	2.197	2.172	-1.14
145	348	343.4	-1.32	1.807	1.791	2.155	2.134	- .97
146	264	261.5	-0.94	1.822	1.806	2.086	2.066	- .96
155	220	218.9	-0.50	1.820	1.817	2.040	2.036	- .196
156	264	263.7	-0.11	1.810	1.816	2.074	2.080	+0.29
157	304	304.5	+0.16	1.800	1.799	2.104	2.105	+0.05
158	348	358.9	+3.04	1.790	1.825	2.138	2.184	+2.15
193	444	447.9	+0.87	1.770	1.776	2.214	2.224	+0.45
194	444	446.6	+0.58	1.770	1.779	2.214	2.226	+0.54
195	444	450.8	+1.51	1.770	1.782	2.214	2.233	+0.86
196	444	451.1	+1.57	1.770	1.778	2.214	2.229	+0.68

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TABLE II

Batch No.	Aim Load (g/cc)	Actual Load (g/cc)	%Deviation Aim-Actual Loading	Aim Carbon Density (g/cc)*	Actual Carbon Density (g/cc)	Aim Composite Density (g/cc)**	Actual Composite Density (g/cc)	% Deviation Aim-Actual Composite Density
255	179	179.3	+0.16	1.845	1.836	2.024	2.015	-0.44
256	179	180.4	+0.78	1.845	1.835	2.024	2.015	-0.44
257	314	310.9	-0.98	1.833	1.821	2.147	2.132	-0.69
258	417	418.9	+0.45	1.809	1.794	2.226	2.213	-0.58
259	417	421.3	+1.02	1.809	1.795	2.226	2.216	-0.45
286	179	179.6	+0.33	1.846	1.856	2.025	2.036	+0.54
287	179	181.2	+1.22	1.846	1.850	2.025	2.031	+0.29
288	179	180.9	+1.06	1.847	1.847	2.026	2.028	+0.09
289	179	182.1	+1.73	1.847	1.845	2.026	2.027	+0.05
290	179	177.8	-0.67	1.829	1.838	2.008	2.016	+0.40
291	179	180.9	+1.06	1.829	1.839	2.008	2.020	+0.60
292	179	181.0	+1.11	1.829	1.841	2.008	2.022	+0.70
293	179	181.2	+1.22	1.833	1.841	2.012	2.022	+0.50
294	179	180.2	+0.67	1.833	1.829	2.012	2.009	-0.15
295	179	179.2	+0.11	1.833	1.826	2.012	2.005	-0.35
297	179	179.3	+0.16	1.843	1.834	2.022	2.013	-0.45

*Calculated from Eq. XIV

**Calculated from Eq. XIII

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Figure 2

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Figure 3