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REPORT OF THE WORKING COMMITTEE TO THE
FUEL ELEMENT DEVELOPMENT COMMITTEE FOR
ACTIVITIES ASSOCIATED WITH THE SMALL
AND K HANFORD REACTORS

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DOUGLAS UNITED NUCLEAR, INC.

REPORT OF THE WORKING COMMITTEE TO THE FUEL ELEMENT

DEVELOPMENT COMMITTEE FOR ACTIVITIES ASSOCIATED

Classification Cancelled and Changed To WITH THE SMALL AND K HANFORD REACTORS

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REPORT OF THE WORKING COMMITTEE TO THE FUEL ELEMENT
DEVELOPMENT COMMITTEE FOR ACTIVITIES ASSOCIATED
WITH THE SMALL AND K HANFORD REACTORS

PRODUCTION REVIEW

Current and Forecasted Mode of Reactor Operation

Currently the B, C, KE and KW reactors are in operation. The B and C reactors are operating to produce 12% Pu-240 in both the E metal (0.95) and natural loadings. Some columns will be taken to 15% Pu 240 on a test basis. Production of 12% Pu-240 will result in exposures of 1400 MWD/ton on natural metal and 2,000 MWD/To on E metal. Currently exposures are such that $10\frac{1}{2}\%$ Pu-240 has been achieved. Some control problems and temperature cycling have been encountered with this mode of operation.

Both K reactors are on enriched-depleted loads. The depleted metal is scheduled for irradiation to produce 27-30% Pu-240 (about 3,000 MWD/Ton; 15 months residence) and is currently at 30% of goal. The E metal drivers (0.95) will be discharged at weapons grade Pu exposures. The relatively small quantity of natural uranium in the K reactors will be exposed to produce 12% Pu-240.

For the near term future the B and C reactor will continue to operate to produce 12-15% Pu-240. By November 1, it is expected that C reactor will be equipped with a 44 tube, one-inch overbore block which will be fueled with a rod-and-tube geometry. The K reactors, after the present E-D loads are completed, may either continue with more E-D loadings or revert to regular natural-enriched (0.95) loadings to produce 12% Pu-240. It is expected that $\frac{1}{4}$ of one K reactor will be used to demonstrate the capability of E-N operation using 2.1 enriched drivers.

The modes of operation described above are the expected production modes of operation and do not include many test loadings currently being irradiated and planned for irradiation as part of the Hanford Mission Program.

The above described modes of reactor operation have affected the fuels business. With large quantities of fuel going to higher exposures, the quantity of uranium processed in the fuels plant is low. At the present time, the AlSi canning facility is operating four lines on a one-shift basis with a throughput of about 90,000 pieces per month (300 tons/month). In addition, the longer exposures and residence times for the fuels in the reactors increase the severity of service to which the fuels are exposed. In particular, the ability of the aluminum jackets to withstand longer exposure to the corrosive environment is of concern. Also, there is some evidence that the standard alloy (200 ppm Fe and Si) is not optimum for the current and proposed high exposure service.

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Uranium Technology

Grain Size

The average grain size of uranium heat treated by salt bath heating-water quench and induction heating-oil quench received during the period December 1, 1966 through July 31, 1967 was unchanged and ranged between A-6 and A-7.

Inspection of Bare Cores

The combined visual and grain size reject rate was 0.23 percent for the period January 1, 1967 through June 30, 1967.

Hot-Die-Size Heat Treatment Variables Test

A test, PT-023 was conducted in B Reactor using normal metal of O3AN (hot-die size) dimensions, to evaluate differences in dimensional stability, if any, between hot-die-size cores oil quenched in hollow form vs. solid form.

The test was composed of a total of 620 Hot-Die-Sized O3AN elements, 155 elements of each of the following heat treatments:

<u>Heating Method</u>	<u>Core Blank Form</u>	<u>Quenching Medium</u>
Induction	Solid	Oil
Induction	Hollow	Oil
Salt Bath	Solid	Oil
Salt Bath	Hollow	Oil

X-ray diffraction data on transverse sections on each of the four heat treatments show no major differences among their growth-index coefficients. On the basis of the longitudinal growth index coefficients, the predicted length change while under irradiation on the four heat treatments would be only minor. The x-ray data are summarized in Table I below:

Table I
Longitudinal Coefficients on the HDS
Heat Treat Variables Test

<u>Longitudinal Growth Index Coefficients</u>			
<u>Heat Treatment</u>	<u>OD</u>	<u>Mid</u>	<u>ID</u>
Hollow-Induction	+0.060	+0.059	+0.050
Solid-Induction	+0.065	+0.059	+0.030
Solid-Salt Bath	+0.059	+0.065	+0.034
Hollow-Salt Bath	+0.038	+0.043	+0.042

UT-2 peak voltage, which is related to grain size was recorded on the cores for each heat treatment. The grain size varied according to whether the core blanks were solid or hollow prior to heat treatment and whether the core blanks were salt bath or induction heat treated. The data show cores heat treated in solid blank form have a larger but a more uniform grain size, whereas cores heat treated in hollow blank form have a shallow layer next to the ID and OD

which is fine grain size but the grain size becomes larger toward the mid wall of the core. In both cases, the induction heat treated cores were a finer grain size than the salt bath heat treated cores.

The difference in grain size between induction and salt bath heat treat cores is presumed to be a function of the cooling rate on the core. The core heat treated in the salt bath would have a very shallow surface layer of salt on it when transferred from the bath to the quench tank which would change the heat transfer rate from the core. The core heat treated in the induction units would not have this shallow surface layer of salt and when quenched the heat transfer rate from the core to the bath would be greater.

The reactor test was charged to eliminate any tube-to-tube and position-to-position effect and irradiated to an average exposure of 800 MWD/T. After discharge, the fuels were measured to determine dimensional changes. The average dimensional changes are presented in Figure 1. The dimensional data show that the hollow induction-oil heat treated cores exhibited greater OD growth than the other three heat treatments. However, the solid induction-oil and salt-oil heat treated cores exhibited greater ID growth. The warp experienced on each of the four heat treatments was approximately the same. Although small differences in the dimensional stability of the cores during irradiation were noted for the four heat treatments, the difference in performance is considered negligible under current reactor operating conditions.

Limited quantities (approximately 20 tons/month) of Model K5N cores which have been induction heat treated in solid blank form will be charged into the K reactors beginning September.

Fuel Performance

Status of Upstream Fuel Program

In the upstream fuel program Class II fuel cores manufactured at Fernald and certain substandard fuel elements assembled at Hanford are irradiated as a special charge in the first five upstream positions in any of the Hanford reactors. Upstream fuel categorized as follows is irradiated in these low severity positions with negligible increase in failure risk:

Fernald Class II Category

1. Low-Yield Ingots - Non-reject cores from ingots with greater than 25% but less than 70% reject rate will be accepted.
2. Fine grained core rejects - Cores with a UT-2 peak voltage between 5.2 and 5.8 volts which corresponds to an A-9 grain size.
3. Defective radius rejects.
4. Chemical Composition Rejects - The chemical composition limits of Class II cores may be 20% above or below the allowable specified limits.

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5. Undersized dimension rejects - Class II cores may vary five mils under the minimum OD and five mils over the maximum ID and ten mils under the minimum length.
6. Process Specifications Deviation Rejects - Ingots out of specification on the casting cycle and/or on the rolling cycle within reasonable limits may be used for Class II cores.

Hanford Upstream Categories

1. Facing rejects where the length is less than .100 inch above the present lower limit.
2. Marred surfaces less than .010 inch deep.
3. Fuel elements rejected by the bond tester provided that the total external count is less than 300 on enriched and 500 on natural uranium elements.
4. Fail-prone fuel lots if considered acceptable upstream quality.

During CY 1966 and through July 1967, 131,000 Class II and substandard fuel elements were accepted for the upstream fuel program. To date, no failures have occurred.

Fuel Performance

Five normal and six enriched fuel elements failed during the period January 1, 1967 through July 31, 1967. Four normal fuel element failures were watermix elements and all four failures resulted from mechanical damage during charging operations. Currently, studies are being conducted trying to reproduce the defect observed on the watermix elements and determine why these elements are susceptible to mechanical damage.

After one year without any core split failures, four such failures have occurred within a four-month period. The core split failures in B and D reactors were normal production cores while the core split failures in KW occurred in a demonstration load, PT-040, for the high-power-density program. It is noteworthy that only three fuel element failures have been caused by manufacturing defects and that no failures have occurred from side hot spots or groove corrosion.

Table II
Fuel Element Failures
January 1, 1967 - July 31, 1967

<u>Fuel Element Type</u>	<u>Reactor</u>	<u>Exposure % of Goal</u>	<u>Classification</u>	<u>Failure Date</u>
Natural-Watermix	B	68	Mechanical Damage	1/18/67
Natural	B	109	Core Split	3/6/67
Enriched Self-Support	KW	32	Manufacturing Defect	3/20/67
Enriched	D	24	Manufacturing Defect	3/27/67
Enriched	D	39	Core Split	3/29/67
Enriched	D	20	Manufacturing Defect	5/1/67
Natural-Watermix	B	60	Mechanical Damage	5/15/67
Natural-Watermix	D	55	Mechanical Damage	5/29/67
Enriched 125 Metal 1)	KW	42	Core Split	7/9/67
Natural-Watermix	B	79	Mechanical Damage	7/10/67
Enriched 125 Metal 1)	KW	63	Core Split	7/23/67

1) Production Test Material.

Thoria Program

Thoria Irradiation Summary

Since the initial thoria loadings in CY 1964, 198,000 thoria elements have been irradiated in DUN reactors and 27 in-reactor failures have occurred to date. Twenty failures occurred in 57,800 irradiated elements during the period October 1964 to April 1, 1965 and seven failures occurred in 139,200 irradiated elements for the period April 1, 1965 to June 30, 1967. On April 1, 1965 several process and control procedures* were incorporated into the thoria canning process to reduce the high failure rate experienced in the first campaign. These procedures and their effectiveness are vividly illustrated by the reduction in failure rates.

*Process changes incorporated which improved outgoing target quality are helium leak tester installed, use of heat sink chucks on the welder and radiographing the weld of each target element.

Table III
Listing of Thoria Target Element Failures*

<u>Failure</u>	<u>Date</u>	<u>Residence Time-Days</u>	<u>%Goal Exp.</u>	<u>Failure Classification</u>
1	1/19/65	51	80	EM
2	1/19/65	51	80	SM
3	1/19/65	UN	UN	SM
4	2/18/65	49	24	EM
5	2/22/65	30	63	SM
6	2/22/65	30	63	SM
7	2/22/65	30	63	UN
8	3/1/65	60	29	UN
9	3/25/65	91	52	SM
10	3/29/65	65	115	SM
11	3/29/65	65	115	EM
12	3/31/65	90	45	ED
13	5/3/65	64	75	EM
14	5/3/65	64	75	EM
15	5/3/65	64	75	-
16	5/12/65	139	87	EM
17	5/12/65	139	87	EM
18	5/14/65	141	88	EM
19	6/23/65	181	106	EM
20	6/26/65	54	92	EM
21	7/27/65	216	114	EM
22	9/13/65	34	17	UN
23	10/21/65	42	75	EM
24	12/30/65	37	46	ED
25	4/13/66	50	13	UN
26	7/7/66	135	72	ED
27	10/17/66	-	51	UN

EM - End defect, (pinholes, weld porosity)

SM - Side defect (manufacturing defect)

ED - Charging machine defect

UN - Unknown

*Summarizes all thoria failures to date.

Thoria Failure Modes

Thoria target elements have been observed to fail by one or more of three distinct modes:

1. Expansion due to internal gas pressure evidenced by bulging of the can at its weakest point and consequent coolant flow restriction.
2. Weld porosity allowing entry of water into the element and escape of fission products, often accompanied by fusing and uniform swelling of the thoria producing flow restrictions and further fission product release.
3. Mechanical damage caused by improper handling and/or charging, producing surface damage severe enough to allow penetration of the cladding and thus fission product release and water entry prior to attainment of goal exposures.

The tendencies of thorium oxide to fuse during irradiation appear to be best described as a function of firing temperature of the gel and the presence of water vapor in the fuel element. The fused thoria, found in elements exhibiting overall swelling after irradiation, is characterized by the occurrence of color changes to the thoria. These colors occur in circular bands running parallel to the longitudinal axis of the element, with natural yellow colored thoria, dark blue, and pale blue bands occurring from the outer diameter region to the center of the element (Fig. 2 and 3). These color bands have been associated with definite grain structures and also are dependent on adsorbed gases present at elevated temperatures.

Thoria Separation

A total of 192 tons of thorium has been processed through the separation plant. The average dissolver charging rate for the entire campaign was 3.7 tons of thorium per day with a peak rate of 5.3 tons per day.

The uranium 233 product quality has generally been within all established goals. The only quality problem of significance resulted from the pick-up of residual U-238 by the process streams. Thus, the first approximately 9.6 kilograms of U-233 product contained greater than the goal limit of 3.9% U-238. During the production of approximately the next 110 kilograms, the U-238 content of all U-233 product was within goal limit. The next batch progressively decreased to about 0.7 percent, the "background" level expected on the basis of U-238 content of the thoria originally charged to the reactors.

Thoria Characterization Studies

Past requirements for dense thoria powder for reactor loadings to protect the biological shield at Hanford have been met by using standard dense thoria prepared by the sol-gel process. Some consideration is now being given to using a sintered "light-oxide" instead of dense thoria for future reactor fringe loadings.

On this basis, denitrated "light oxide" of thorium which had been irradiated and separated, and virgin thorium powder were compared. Also, a study was made on the virgin and recycled light oxides after they were sintered at 1200°C. The data for this study are summarized in Table IV.

Table IV
Characterization Studies
In Virgin and Recycled ThO₂

	<u>Virgin</u>		<u>Recycled</u>	
	<u>Non Sintered</u>	<u>Sintered</u>	<u>Non Sintered</u>	<u>Sintered</u>
Compaction Density gram/cc	4.77	5.66	4.50	6.87
Offgas Analysis cc/g	1.56	0.064	1.09	0.059
Surface Area Units	37.4	.440	42.2	.434
Dissolution %	100%	100%	100%	100%
U (ppm)	29.3	--	153	--
LOI (grams)	0.69	--	1.10	--

The offgas analyses for the unsintered light oxides show such a large volume release of non-condensable gases, charging of such oxide in the reactor would be undesirable.

Twelve reactor grade thorium elements have been produced from sintered recycle "light oxide". The "light oxide" powder was fired to 1100°C to remove the excess water and absorbed gases. The "light oxide" was compacted into aluminum cans to a density of 5.6 g/cc. Such compacted target elements did not present any special canning problems except at the helium backflow station where low density and very fine particle size of the "light oxide" made it necessary to evacuate the element very slowly.

Thorium Canning Development

Continued use of a 3/8 inch thick thorium wafer plus chilling the upper part of the assembly during welding has resulted in fewer weld rejects caused by gas expansion (blow outs). The welding reject rate improvement for the last five campaigns is shown in Figure 4. During the fifth campaign, the copper heat sink was replaced with a stationary water cooled collet and the welding torch was rotated rather than rotating the thorium assembly under a stationary torch.

The use of the thorium wafer resulted in an overall yield increase of about 10% compared to that experienced with the copper heat sink only. A breakdown of various reject categories before and after the use of the thorium wafer is shown in Figure 5.

Coproduct Mission (Tritium)

A 2.1 E-N demonstration load was charged in KW reactor during May to demonstrate the feasibility of large scale production of tritium using 2.1% enriched fuel and lithium bearing target elements. The test consisted of 17 columns of AlSi canned 2.1 fuel elements and four columns of lithium bearing target elements. The 2.1% enriched fuel columns will operate at about 130% of the average natural

uranium column power or about 2,200 kilowatts. Three of the lithium bearing columns contain lithium-aluminum alloy cores and the other column has elements made with both solid and hollow lithium aluminate (LiAlO_2) pellets.

High Power Density Program

An initial test was conducted in KW Reactor using 125 metal of K5E fuel dimensions, except 5.54-inch long rather than 6.54, to evaluate various uranium alloys operating at 150 percent of the average specific power of a K reactor, central zone, natural column. The average specific power in the test was approximately 120 kw/ft. The test was irradiated to an average exposure of 1700 MWD/T with calculated exposures on individual elements as high as 2400 MWD/T and calculated uranium temperatures up to 500 C.

The four alloys listed below and two heat treatments, salt-oil and induction-oil were tested:

<u>Alloy</u>	<u>Nominal Composition</u>		
	<u>Fe-ppm</u>	<u>Si-ppm</u>	<u>Al-ppm</u>
I	150	120	-
II	350	-	800
III	350	350	800
IV	-	350	-

The test was charged to eliminate tube-to-tube and position-to-position effects. A total of 640 elements (160 of each alloy, half of which were salt-oil heat treated), were charged into 16 columns. Metallographic examination of unirradiated specimens indicated that the metal quality was good, and there were no significant differences among alloys in this respect. It is significant to note that the charge compositions for the ingots for the above material were essentially all derbies.

Table V
PTA-040 Core Crack
and Length Change Data

<u>Alloy</u>	<u>Heat Treatment¹⁾</u>	<u>No. of Cracked Cores</u>	<u>Average Length Change - Mils</u>
I	Salt	9	1.4
	Induction	0	7.5
II	Salt	4	6.3
	Induction	2	16.3
III	Salt	1	0.0
	Induction	0	7.6
IV	Salt	0	- 3.0
	Induction	0	6.5

1) Refers to method of heating. All cores were quenched in oil.

After discharge the fuels were measured to determine dimensional changes and tested ultrasonically to determine if any core cracking had occurred. The average diameter changes are presented in Figure 6, the average length changes and the core cracking data are presented in Table V. The dimensional data show that Alloy I exhibited the greatest diameter growth on both the OD and ID of the fuels and that the other three alloys were all roughly comparable. The induction-oil heat treated cores showed consistently greater length changes than did the salt-oil heat treatment. The core-crack data indicated Alloys I and II are more prone to cracking than are Alloys III and IV. The tester signals indicated the cracks were $\frac{1}{2}$ to two inches long and were close to the mid-point along the fuel length. The cracks occurred in elements irradiated in the central 15 positions of the fuel columns. Figure 7 shows a cross section of one of the cracked cores.

A second high power density test of 125 metal with K5E fuel dimensions has been partially irradiated. The purpose of the test is to irradiate a total of one-hundred columns of fuel at 150 percent of standard specific power to demonstrate high power density operation. Initially, the cores were to be of composition I but the results of the test described above which indicated composition III or IV would perform better; plus quality problems with the cores received from FMPC, led to the decision to irradiate only twenty columns of Alloy I and eighty columns of Alloy III. The twenty columns of Alloy I were charged into KW Reactor in May and the remaining eighty are scheduled for charging in September or October. Two failures were sustained in the initial load, both attributed to longitudinal core splitting. The first failure occurred at a column exposure of 640 MWD/T and the second at 900 MWD/T. The second failure occurred on July 23, 1967, and the irradiation was terminated at that time. Metallographic examination of unirradiated cores show the 125-metal used in this test to be comparable in stringer content to standard metal routinely irradiated. However, the relatively low exposures at which the failures occurred suggested they were caused by metal quality defects.

Metal quality defects were observed in unirradiated cores from this group (See Figures 8, 9, 10, 11, 12) and it is surmised that these metal defects were a function of the scrap additions to the crucible charge, although it did conform to the FMPC specifications. The charge for these 1.25 enriched ingots was as follows: 600-800 lbs of .947 solid scrap from the zirnlo process; 200-300 lbs of 2.1 solid scrap from sawed sections of NPR primary ingots; and 300-330 lbs of derby metal. The sawed sections of NPR primary ingot (these were ingots cast from briquettes and double melt top crop of the last 2.1 enriched NPR campaign where the ingots had the carbide problem) are believed to be the contributing factor to such defects.

Work is being conducted on quality control samples of other test and standard production cores to determine if such inclusions are actually observed in other samples or if such inclusions are only characteristic of these cores.

Reactor Modernization (Overbored Reactors)

Two charges of one-inch overbore, rod-in-tube, fuel fabricated by the Hot-Die-Size process have been successfully irradiated in the KER Loop. The one-inch overbore fuel consists of a $2\frac{1}{2}$ -inch outer tubular element fitted with a one-inch rod element. The rod element may be either uranium or target material depending

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upon the desired irradiation product. Examination of irradiated fuel has shown no unusual corrosion or problem areas.

One-inch overbore fuel for larger scale testing in C-Reactor has been designed and initial phases of production have started. Forty-four one-half inch overbore tubes located in the central core of C-Reactor have been selected for additional overboring to one inch, 34 for smooth bore Zircaloy-2 and ten for ribbed aluminum tubes. Two models of C-Reactor fuel were designed, one (CM1E) to fit smooth bore Zircaloy-2 and one (CM2E) to fit ribbed aluminum process tubes. Dimensions of the two C-Reactor fuel elements are tabulated in the Table below along with the KER test fuel, Model XMIN, for comparison purposes.

Table VI
One-Inch Overbore Fuel Dimensions

<u>Dimensions</u>	<u>Fuel Model*</u>		
	<u>XMIN</u>	<u>CM1E</u>	<u>CM2E</u>
Outer Tube: Length, in.	8.870	8.870	8.870
Outer Diameter, in.	2.530	2.451	2.400
Inner Diameter, in.	1.295	1.234	1.180
Clad Thickness, in.	0.050	0.045	0.045
Projected Diameter (max), in.	2.696	2.631	None
Support Height (min), in.	0.075	0.085	None
Core Length, in.	8.400	8.400	8.400
Core Outer Diameter, in.	2.430	2.361	2.310
Core Inner Diameter, in.	1.402	1.324	1.270
Core Weight, lbs	17.73	17.18	16.74
Inner Rod: Length, in.	8.400	8.400	8.400
Diameter, in.	0.996	0.957	0.957
Clad Thickness, in.	0.050	0.045	0.045
Core Length, in.	8.400	8.400	8.400
Core Diameter, in.	0.896	0.867	0.867
Core Weight, lbs	3.61	3.36	3.36

*XMIN - KER Loop

CM1E - C-Reactor smooth bore Zircaloy-2 process tubes

CM2E - C-Reactor ribbed aluminum process tubes.

Process Technology

Status of Hot-Die-Size Program (including 313 Building Production Line)

A supplemental one line production facility in 313 is currently being installed to produce some 400-450 Hot-Die-Size pieces per shift. Planned improvements could later increase this capacity to 600-700 pieces per shift. Operation of this facility has been primarily designed for single shift operation along with the regular AlSi lines. Development Hot-Die-Size facilities in the 306 Building will be used for process development and preparation of most small

quantity Hot-Die-Size test lot material. Material for tests requiring more than a few hundred pieces would probably be prepared on the 313 line. The 313 production facility will also be used to produce the necessary rod-and-tube fuel for the C-Reactor 44-tube overbore block. The 313 facility is expected to be in operation about January 1968.

Routine fabrication of Hot-Die-Size fuel in prototypic equipment is continuing and providing experience in fabrication and irradiation of this process fuel. Thus far in excess of 200 tons of Hot-Die-Size fuel have been irradiated. Differences in localized corrosion behavior between the AlSi process and Hot-Die-Size process which showed up during the initial irradiation have still not been completely resolved. At present, both AlSi and Hot-Die-Size fuels experience about the same severity of ledge-and-groove corrosion in the B and C Reactors. However, based on earlier tests, the severity of groove corrosion on Hot-Die-Size fuel irradiated in the central zones of the K Reactor appears to be somewhat worse than that observed on AlSi fuel. Both the AlSi fuel and the Hot-Die-Size fuel undergo localized corrosion by the grooving and ledging phenomenon.

Currently two tests are awaiting irradiation to determine the effect of core size on the incidence of ledge and groove corrosion (the Hot-Die-Size fuel has an approximately 20 mil larger diameter uranium core than the AlSi), and the possible effect of any graphite lubricant (used during sizing) which may remain imbedded in the finished fuel surface to form an aluminum-graphite couple.

Evaluation of Dichromate Autoclave Films

A test was conducted in KE Reactor to evaluate the effects of dichromate autoclave films on the corrosion of 8001-clad, Hot-Die-Sized fuels. The variables tested are tabulated in Table VII.

Table VII
Special and Standard Autoclave Treatments

Water Cycle:	Deionized water
Dichromate Cycle:	Potassium dichromate, 8-10 wt%, pH 6.6-6.8
Time:	25-30 hours each cycle

<u>Treatment</u>	<u>Cycle Temperature -C</u>	
	<u>Water Cycle</u>	<u>Dichromate Cycle</u>
A	170	170
B	290	170
C	290	230
D	340	230

Standard Cycle: Deionized Water, 40 hours, 170C

The test consisted of 18 columns of fuel with the five treatments (four test conditions plus the standards) charged in an alternating pattern in the downstream ten positions of each column. One column was discharged at an exposure of 315 MWD/T, and the remaining 17 were irradiated to an average exposure of 845 MWD/T at an average tube power of 1900 KW. The results of the test

are summarized in Table VIII. The data show that the special autoclave treatments did not improve the corrosion resistance of the cladding, and treatments C and D appear to be worse with respect to weight loss and ledge corrosion than the standard treatment or treatments A and B.

Table VIII
Summary of Results of PTA-026
Dual-Cycle, Dichromate Autoclave Treatment

<u>Treat-</u> <u>ment</u>	<u>Number</u> <u>Pieces</u> <u>Examined</u>	<u>Avg. Wt.</u> <u>Loss-g</u>	<u>Percent With*</u> <u>Ledge or Groove</u> <u>Corrosion</u>		<u>Percent With</u> <u>Groove</u> <u>Corrosion</u>		<u>Percent With</u> <u>Ledge</u> <u>Corrosion</u>	
			<u>Total</u>	<u>10 Mils**</u>	<u>Total</u>	<u>10 Mils**</u>	<u>Total</u>	<u>10 Mils**</u>
A	10	3.20	50	10	40	0	20	10
B	20	3.25	55	10	50	5	30	5
C	17	5.12	65	41	41	12	53	29
D	18	5.54	78	39	22	11	72	28
Standard	69	3.38	48	19	33	9	39	10

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*Many pieces exhibited both ledge and groove corrosion.

** Percent of pieces examined with corrosion greater than 10 mils deep.

Zircaloy-2 Cladding by the Hot-Die-Size Process

A small quantity of fuel has been fabricated by the Hot-Die-Size process using Zircaloy-2 components. No major sizing problems were encountered. Preheating, which was performed in an air atmosphere, caused excessive oxidation of the cladding. In spite of this oxidation, a Zr-Ni-U diffusion bond was realized on most of the lateral surface area (Figure 13). No bonding was experienced on the fuel ends.

The range of sizing parameters employed is shown below:

<u>Temp °C</u>		<u>Ram Speed in/Min.</u>	<u>Upper Ram Press PSI</u>	<u>Lower Ram Press PSI</u>	
<u>Max</u>	<u>Min</u>			<u>Max</u>	<u>Min</u>
600	450	120	350	650	40

A reduction ratio of 15-20% was experienced in the initially 28 mil thick zirconium cladding. This variation is due to non-uniformity in initial cladding thickness.

Optimum sizing temperature and lower ram pressure appear to be about 550°C and 500 PSI respectively. Lower temperature and ram pressure gave no diffusion bonding. A sizing temperature of 600°C and ram pressure of 650 PSI caused severe deformation of the uranium core. Work to further optimize sizing parameters will continue with inert gas and vacuum techniques being applied during the preheating and sizing operations.

No end bonding parameters have yet been established.

Nickel-Plated Fuels

Studies related to improvements in corrosion behavior of Hanford reactor fuels have included the nickel-plating of the fuel elements. Two plating techniques are being evaluated: (1) the electroplating of aluminum components prior to fabrication by the Hot-Die-Sizing process and (2) the electroplating of finished fuels which have been fabricated by either the AlSi process or the Hot-Die-Sizing process. The bonding of the nickel-plate on fuels fabricated from the nickel-plated components is attained by the high temperature and pressure required in the die sizing operation. Bonding of the plate on fuels plated after fabrication depends upon a subsequent heat treatment or autoclave operation which is not as yet completely evaluated. Concurrent with development work on the plating process, evaluation of the corrosion behavior of nickel-plated fuels is under way.

In a preliminary and limited size test of nickel-plated fuels in the KER flow loop, the fuel elements tested in a Zircaloy tube experienced severe localized corrosion after exposure of about one month. The fuels tested had thin nickel coatings which had been applied to the aluminum components prior to the die-sizing operation. The electroplates were thin and were known to be porous. The fuels in the zircaloy tube were severely attacked by a localized corrosion largely but not totally in the vicinity of the self supports. The aluminum supports showed little or no evidence of attack. The fuels in the aluminum tube showed only minor intermittent corrosion very close to the ends of the

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fuel elements in what appeared to be mechanically damaged areas. These results were not considered to be conclusive but gave evidence of a galvanic system which may have caused the severe and specific attack of the cladding. Additional test fuels were installed in the test loop, including fuels which had been nickel-plated after fabrication.

After exposure of about one month in the K Reactor flow loop in which standard reactor water composition, temperature, and flow rates are duplicated, examination of nickel-plated fuels revealed no evidence of localized corrosion on fuels which had been nickel-plated after fabrication. The prior test in the same system showed severe pitting corrosion attack of fuels in which the aluminum components had been nickel-plated prior to fabrication by the Hot-Die-Sizing process. Known differences between the two types include a greater amount of working of the surface nickel and the presence of particles of graphite from the lubricant used on the fuels which were more severely attacked. The fuels which were nickel-plated after fabrication were returned to the test loop. Purposely defected nickel-plates on similar fuel elements were added to the test.

DEVELOPMENT REVIEW

Nickel-Plating Studies

Work being done on the electrodeposition of nickel on uranium has led to a correlation between anion size in the plating solution and plate stress. Nickel-plating solutions containing larger anions (sulfamate) produced a lower tensile stress than those made from smaller anions (chloride). It was found that nickel-plates having zero stress or stress in compression were brightened. The advantages for a plating solution which will produce a stress-free deposit and also act as a brightener are: (1) better adherence may result when unstressed nickel is deposited on uranium; perhaps acceptable adherence could be obtained on a smooth uranium surface, thus reducing the porosity of the resulting plate, (2) since brighteners are generally used in particulate plating to prevent uneven growth of the deposit, a brightener which is not substantially occluded in the resulting plating is necessary to assure a contamination-free plate.

To evaluate the effects of anion size, nickel salts of water soluble sulfonic acids were prepared. Each solution had 0.1M NiBr_2 added to minimize oxidation of the organic material at the anode, the temperature was held at $49.4 \text{ C} \pm .3 \text{ C}$, the pH was adjusted to optimize the visual quality of the nickel-plate, the current density was 30 mA/cm^2 . The results are tabulated in Table IX in decreasing quality and comments are made on the appearance of the nickel deposit.

Table IX

<u>Sulfonic Acids (Nickel Salt)</u>	<u>pH</u>	<u>Total Nickel Conc. M</u>	<u>Stress (p.s.i.)</u>	<u>Current Efficiency%</u>	<u>Grade</u>	<u>Remarks</u>
7 amino 1,3 naphthalenedisulfonic acid	4.0	0.3	-25,700	69	1A	Bright
3 amino 2,7 naphthalenedisulfonic acid	3.5	0.7	-13,100	76	1B	Bright
6 amino 1,3 naphthalenedisulfonic acid	4.0	0.8	-18,300	64	1C	Bright
8 amino 1,3,6 naphthalenetrisulfonic acid	3.4	0.9	- 2,400	95	1D	Semi- Bright
3 amino 1.5 naphthalenedisulfonic acid	3.5	0.6	- 5,500	76	1E	Semi- Bright
2 naphthol 6,8 disulfonic acid	2.1	0.4	- 6,400	49	2A	Semi- Bright
m benzenedisulfonic acid	3.5	0.8	- 6,900	99	3A	Semi- Bright
1 naphthalenesulfonic acid	2.5	0.3	- 500	58	3B	Semi- Bright
benzenesulfonic acid	2.5	0.5	600	53	3C	Semi- Bright
1,5 nap- thalenedisulfonic acid	2.5	0.2	2,200	53	3D	Semi- Bright
2,7 naphthalenedisulfonic acid	2.8	0.7	20,200	73	3E	Semi- Bright
ethanedisulfonic acid	2.7	1.0	30,600	89	4A	Semi- Bright
ethanesulfonic acid	2.6	0.5	74,000	69	4B	Semi- Bright

Preliminary experiments were carried out for occluding silicon particles in nickel-plate electrodeposited on uranium. This work is directed toward reducing plate porosity and providing an impermeable layer which would assure a secondary barrier to penetration of reactor coolant to the core material. A Watts bromide plating solution was used and 0.2 g/ml of silicon (325 mesh) was added. The silicon was kept in suspension by a magnetic stirrer and the cathode was in a vertical position rotating at 750 rpm. Several mils of nickel were deposited, but very little silicon was occluded in the deposit, even when the cathode was stationary. One sample was plated while using 40 Kc/S ultrasonic agitation with no substantial improvement. Since very little occlusion takes place while the particles are moving,

the cathode was then placed in a horizontal position and the silicon particles were suspended by rapid stirring, then the stirring was stopped and the particles were allowed to fall on the surface while plating. Plans are currently under way to evaluate the application of this technique to plating solutions containing suspensions of finely divided charged particles of a ceramic material.

A tin-nickel alloy was considered to have potential as a bonding medium and a corrosion barrier for hot-die-sized fuel since tin has a low neutron cross section and forms an extremely corrosion-resistant alloy with nickel. An electroplating solution with the following composition was prepared to obtain the Sn-Ni plates:

SnCl ₂ .2H ₂ O	50 g/l
NiCl ₂ .6H ₂ O	250 g/l
NH ₄ F	33 g/l
HCl	to pH 2.5
NaF	28 g/l -
Temp.	65° C
Current Density	27 ma/cm ²

It was found, however, that this solution reacted vigorously with the uranium, the reaction proceeding rapidly at plate pore sites even when the uranium was coated with 12 u of nickel. The bromide ion was substituted for chloride ion to reduce the reactivity of the solution toward the uranium. Using the bromide solution, it was possible to put a tin-nickel alloy coating on a uranium specimen which had approximately 12 u of nickel previously deposited on it. The test specimen was subsequently heated to 650° C for eight (8) minutes in air and the tin-nickel alloy turned white, probably forming an oxide in the vicinity of 500° C. An estimate of the Ni/Sn ratio was made by X-ray fluorescence and found to be 1.7 atoms Ni to 1.0 atoms of Sn. Since the tin-nickel alloy reacts with the atmosphere at fairly low temperatures, it is not compatible with the Hot-Die-Sizing process.

Aluminum Alloy Development

A development program was initiated to evaluate several aluminum alloys for use as cladding material to provide more corrosion resistant fuels. Three types of alloys were included in the study; Al-Ni-Fe alloys, age hardenable alloys, and powdered metal alloys. Preliminary evaluation includes the following specific alloys:

	<u>Alloy No.</u>	<u>Ni</u>	<u>Fe</u>	<u>Zr</u>	<u>Si</u>	<u>Mg</u>	<u>Al</u>
Al-Ni-Fe	1	.064	2.10				Bal.
	2	1.20	1.80				Bal.
	3	0.64	2.10	0.10			Bal.
	4	0.93	0.53	0.10			Bal.
	5	2.64	0.34				Bal.
	6	1.38	1.44				Bal.
Age Hardening Alloys	7	1.00	0.15		11.50	0.84	Bal.
	8	1.20	0.30		12.00	0.66	Bal.
<u>Powder Metal</u>	9	7.00	4.80		(O ₂ < 0.2%)		Bal.

The alloys were prepared by melting in a vacuum induction melting furnace using an aluminum coated graphite crucible. The alumina was applied using the Rokide process. The graphite molds were brush coated with a zirconium silicate mold wash. The melting cycle used on two ingots produced to date is as follows:

1. Low power warmup and pump down to < 5 microns.
2. Backfill with argon at 500 C to 20" Hg absolute.
3. Melt down and raise temperature to 950 C hold two hours.
4. Cool to 760 C with low power.
5. Increase power for stirring effect and pour with power on at 780 to 800 C.
6. Add metal to shrink cavity during solidification.

Some segregation of alloy constituents was experienced during the initial alloy casting efforts. The casting equipment is being modified to provide closer control of the cooling rate of the melt.

A study was undertaken to evaluate the use of dual alloy cladding to provide improved corrosion behavior of aluminum clad fuels. The concept involves an outer alloy which is anodic to the basic aluminum cladding alloy. The outer alloy would then serve as a sacrificial anode in a galvanic corrosion system, thus protecting the fuel cladding. Test quantities of dual alloy tubing were prepared having a ten percent silicon-aluminum alloy inner layer and 8001 alloy outer layer. Studies with this material are being confined to evaluation of the working properties of the alloys and the effects of the die sizing operation on the relative thicknesses of the two layers. Corrosion studies of the dual alloy tubing will require the casting of extrusion billets having acceptable second phase dispersion in the alloys which the modified casting equipment will provide.

Electrodeposition of Zirconium on Uranium

Electron microprobe examination of zirconium-plated uranium cylinders produced by electrolytic deposition from molten salt baths shows that while there is interdiffusion of the uranium and zirconium, the surface of the fine-grained portion of the plate is essentially pure zirconium. The tendency of the zirconium plate to change from a nonporous, fine-grained structure to a deposit composed of large needles or nodules of metal is therefore not attributed to the formation of a uranium-zirconium substrate.

Results of electroplating experiments done in a KF (.55 mol %) - LiF (45 mols %) (m.p. 550° C) system were very encouraging. As in the CsF - LiF system, a smaller fraction of the deposit was in the form of nodules or needles. Adherence of the plate was good and it appeared to be free of voids under low power magnification.

Cell corrosion necessitated shutdown of the unit for repair. During a pre-electrolysis run prior to start-up of the unit a cycling of current and potential was observed which had been noted previously during actual plating. Whether this cycling is associated with changes in the zirconium plate has not as yet been determined. Subsequent work produced excellent results in plating at 10 and 30 mA/cm^2 . Plates were made from less than a mil to 30 mil thick. An abrupt transition from the smooth to granular plate was not noted. None of these plates had loose dendrites; they were all very adherent and compact with no voids noted.

Current plans are to demonstrate a continuous molten salt purification technique for electrolytic systems. Further work on this study will be deferred for more immediate and pressing development needs in electroplating from aqueous systems.

Pu-238 Program

In May 1967, the irradiation of neptunium target elements was initiated in KE Reactor to demonstrate the Pu-238 production capability of Hanford reactors. The program is directed toward the demonstration of the following advantages inherent to Hanford neptunium irradiations:

1. Conversion to Pu-238 of up to 85% of the Np destroyed with a product purity in excess of 95% by the tailoring of the flux pattern and exposure time to minimize Np-238 fissions and neutron capture by Pu-238.
2. Hanford capability for target charging-discharging operations without interference with normal reactor operation, allowing optimum neptunium exposures which are independent of reactor schedules.
3. A minimum out-of-reactor time for the unconverted neptunium in a sustained recycle mode of operation thereby allowing maximum utilization of the scarce target material. A 45-day recycle was demonstrated between discharge of irradiated targets and the recharging of reconstituted Np-237 elements.

The initial charges consisted of nominal 10 w/o Np-Al alloy cores clad in C-64 aluminum alloy cans made according to the flow sheet shown in Figure 14. The cores were prepared by Cryolite reduction of NpO_2 and casting to a master alloy ingot which was recast to the approximate target dimensions. The target castings were machined to final dimensions and enclosed in C-64 aluminum alloy cans which had been annealed. The target elements were subsequently subjected to a nominal 1000 psig hydrostatic pressure to assure contact between the core material and cladding.

Additional targets are being prepared to evaluate the fabrication and irradiation of neptunium oxide in graphite matrices. This phase of the program includes targets containing pre-machined graphite cores impregnated with NpO_2 and targets containing compacted pellets fabricated from a mixture of powdered graphite and finely divided NpO_2 . The impregnated cores were fabricated by immersing the pre-machined cores in a solution of $\text{Np}(\text{NO}_3)_4$. After drying the cores under controlled conditions, high firing ($\sim 1000^\circ \text{C}$) denitration converts the neptunium to oxide. Preliminary work indicates that cold compaction of graphite powder containing no binder produces a compacted pellet having a specific gravity of 2.2.

FIGURE 1
HDS Heat Treatment Variable Test

FIGURE 2

A Cross Sectional View of a Typical Failure in the Aluminum Cladding. Note the Fused Appearance of the Thoria and the Presence of Various Colored Bands Within the Thoria

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FIGURE 3
A Cross Sectional View of a Typical Failure in the Aluminum Cladding

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FIGURE 4
Effects of the Heat Sink and Thoria Wafer Upon Weld Defects Caused by Internal Gas Expansion During Welding

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FIGURE 5
Production Data Showing the Overall Improvement Achieved with the Thoria Wafer

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FIGURE 6
PTA - 040 High Power Test Average Diameter Changes

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FIGURE 7
Post - Irradiation Examination of Cracked Core from 125 High Power
Density Fuel Test

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FIGURE 8
Transverse Cross Section - 250X As Polished Showing Sub-Surface
Defect in Enriched 1.25 Uranium Cores. (Note Porosity Around Defect)

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FIGURE 9
Transverse Cross Section - 250X As Polished Showing Surface Defect
in 1.25 Enriched Uranium Cores

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FIGURE 10
Transverse Cross Section - 250X As Polished Showing Surface Defect
and Extreme Amount of Porosity Around Defect in 1.25 Enriched Core

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FIGURE 11
Longitudinal Cross Section - 250X As Polished Inclusion Observed in
1.25 Enriched Uranium Cores Which are Approximately 1/8" from
Outside Wall of Core

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FIGURE 12
Transverse Cross Section - 250X As Polished Metal Defect Observed in
1.25 Enriched Uranium Core

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FIGURE 13
Zr - Ni - U Diffusion Bond

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NEPTUNIUM - 237 TARGET ELEMENT FABRICATION
PROCESS FLOW DIAGRAM

FIGURE 14