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REPORT TO THE WORKING COMMITTEE
FROM THE GENERAL ELECTRIC COMPANY, HANFORD

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January 7, 1963

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PRESENT REACTOR FUEL DEVELOPMENTUranium Core Production Data**DECLASSIFIED**Quality of Uranium Cores

The visual inspection reject rate for all virgin cores continues to be low (less than 0.2%) during the last four month period as shown in Table I. The percentage of virgin cores being rejected by the UT-2 test for fine grain difference, large grain, and surface defects has been less than 2% except for the cores tested in October.

The average total hydrogen values obtained from one core per lot for each core model sampled from August through July are shown in Table II. No cores were found that exceeded 3.0 ppm hydrogen.

During this reporting period, the following 12 ingot cores were found by HAP0 chemical analyses audit of one core per incoming lot to be out of specification:

<u>No. of Cores</u>	<u>Type Metal</u>	<u>Out of Specification Analysis</u>	<u>Maximum Allowable</u>
2	Natural Ingot	762 to 885 ppm C	750 ppm C
1	Natural Ingot	192 ppm Fe	185 ppm Fe
7	Enriched Ingot	775 to 961 ppm C	750 ppm C
3	Enriched Ingot	198 to 200 ppm Fe	185 ppm Fe

Three natural ingots were found that had reactivity values below the minimum acceptable limit. It is not yet known whether the isotopic level is low or whether some unusual impurity is present.

Uranium Grain Size

The trend in average ingot core grain size is shown in Figure 1. There appears to be a decrease in grain size (increase in FEDC grain size number) coincident with when the new heat treating facilities in Plant 9 and 6 were put into production use. The grain size has decreased to such a value in enriched cores in October and in natural cores in November (when iron and silicon began to be added to natural ingots), that NLO is experiencing unusually high reject rates for fine grains (peak voltage). Upon examining some of the "peak" rejects at HAP0, it was found that most of these rejects at NLO were acceptable on the HAP0 UT-2 testers. Therefore, new "peak" standards were calibrated at HAP0 and shipped to NLO to bring the NLO testers in closer agreement with the HAP0 testers.

TABLE I
UT-2 AND METAL QUALITY REJECTS

	<u>August</u>		<u>September</u>		<u>October</u>		<u>November</u>	
	<u>Virgin</u>	<u>Recovered</u>	<u>Virgin</u>	<u>Recovered</u>	<u>Virgin</u>	<u>Recovered</u>	<u>Virgin</u>	<u>Recovered</u>
<u>NATURAL INGOT U</u>								
No. of pieces processed	137 676	20 198	95 518	22 682	143 867	14 941	123 863	26 253
% UT-2 rejects*	1.2	1.0	1.6	-	2.1	0.9	0.8	-
% visual rejects	0.05	0.5	0.06	0.7	0.1	0.6	0.09	0.7
<u>NATURAL DINGOT U</u>								
No. of pieces processed	2 226	0	0	0	3 248	0	0	0
% UT-2 rejects*	1.9				6.5			
% visual rejects	0				0.03			
<u>ENRICHED INGOT U</u>								
No. of pieces processed	39 338	6 754	38 526	8 216	59 053	12 434	47 516	2 721
% UT-2 rejects*	0.9	-	0.7	0.5	2.9	-	1.1	0.9
% visual rejects	0.18	0.8	0.09	0.2	0.07	2.3	0.06	1.9

*Includes large grain, difference, fine grain, and surface rejects.

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TABLE II

TOTAL HYDROGEN
VALUES OF AS-RECEIVED CORES

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<u>Core Model</u>	<u>Lot Type</u>	<u>Type Metal</u>	<u>Avg. Total H₂ ppm</u>	<u>Range, ppm</u>	<u>Total No. Samples</u>
CIIN	KJ	Natural Ingot	1.3	1.1 - 1.5	16
CVN	KW	Natural Ingot	1.1	0.9 - 1.2	6
KIVN	KF	Natural Ingot	1.2	0.8 - 1.7	82
OIIIN	KZ	Natural Ingot	1.2	0.9 - 1.6	100
KIVW	KB	Natural Ingot	1.2	0.9 - 1.6	22
OIIIW	KD	Natural Ingot	1.6	-	1
KIVE	KK	Enriched Ingot	1.4	0.8 - 2.4	51
OIIIE	KY	Enriched Ingot	1.3	0.8 - 2.1	70
CIIE	KC	Enriched Ingot	1.3	1.0 - 2.3	24

NOTE 1. Start of Fe and Si additions to enriched ingots.

NOTE 2. Start-up of Plant 9 enriched heat treating facility.

NOTE 3. Start-up of new Ansler-Morton heat treating facility in Plant 6.

FIGURE 1 - AVERAGE GRAIN SIZE OF I&E URANIUM CORES MEASURED AT HAFO VERSUS SHIPPING DATE FROM FEED SITES

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A production test to determine the dimensional stability of uranium fuel cores classified by the UT-2 tester has been completed and evaluated. This test graded ingot and dingot OIIIN cores into different grain size levels as categorized by peak voltage, difference voltage, and large grain voltage. In addition, Sonic Orientation Resonance Tester (SORT) values were taken for each core. The following conclusions could be drawn from this test:

- 1) The only irradiation effects which could be attributed to UT-2 grain size values were surface bumping and length change. Grain size showed no effect on OD₁, OD₂, OD₃, ID, warp, or tube filling capacity.
- 2) Cores with a UT-2 "peak" reading of 4.5 had an average length change of 0.020 inch less than those cores with 2.0 volts.
- 3) The larger grain size material (smaller UT-2 readings) showed significantly more incidence of "bumping" than the smaller grain material. Also, more dingot than ingot elements showed "bumping."
- 4) There is a very high correlation between the UT-2 peak, average, difference, and large grain readings. Therefore, in almost all cases, if one reading is known, the other three can be calculated to within the accuracy of the UT-2 instruments.
- 5) As has been shown in the past, the dingot material has a greater diametric growth (as shown by OD₂) than the ingot elements. This test showed that the dingot elements also have a greater reduction in length during irradiation than the ingot material.
- 6) Although the SORT values were definitely different in the ingot and dingot elements, no relationship between SORT and irradiation effects could be determined.

Oil Quenched Ingot Cores

Approximately 22 tons of hollow core blank, heat treated oil quenched natural uranium ingot fuel elements were discharged as of December 1 at goal exposure and 25 tons were being irradiated. About 75 additional tons of oil quenched cores are to be irradiated under the existing production test. Preliminary evaluation of 14 columns of alternate oil and water quenched cores exposed to 800 MWD/T indicates that oil quenched cores are dimensionally more stable. A complete evaluation of this data is expected to be completed in

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January, 1963. Evaluation of oil quench natural ingot cores will continue in 1963 with 400 additional tons authorized at a maximum rate of 90 tons per month. Approximately 700 oil and 700 water quenched enriched cores machined alternately from alpha rolled rods are currently being fabricated by NLO for reactor testing.

Dingot Uranium

In April, it was concluded that the irradiation performance of alloyed dingot metal was two-fold worse than ingot metal after discharging approximately 1200 tons of OIIIN dingot cores. Considerable effort was expended by MCW and HAP0 in April through October on dingot uranium in developing methods for eliminating ID quench cracks, improving the core surface chemistry, and improving the canned quality of dingot fuel elements. However, the decision was made in October to discontinue the use of dingot cores at HAP0 based on the marginal performance of dingot fuel elements and the absence of incentives, economic and technical, to continue further with the dingot program. This decision also halted other programs that used dingot uranium such as overbore cores, alpha extruded rods and tubes, gamma extruded rods and tubes, and cores for alternate canning processes.

As of December 1, alloyed dingot OIIIN fuel elements undergoing irradiation and discharged at goal exposure were as follows:

	<u>In-Reactor</u> <u>(Tons)</u>	<u>Discharged</u> <u>(Tons)</u>	<u>Total Number</u> <u>of Failures</u>
Old Reactors	15	1490	29
K Reactors	3	296	0

Eight-Inch Diameter Ingots

NLO has completed an evaluation of casting and rolling 8-inch diameter ingots with favorable results. X-ray diffraction data on cores from 8-inch diameter ingots at HAP0 revealed no unusual diffraction patterns. Grain size, chemistry and inclusion results at NLO were satisfactory. The major advantages of casting 8-inch diameter ingots are:

- 1) A 10% increase in throughput capacity without any equipment changes.
- 2) A larger diameter to height ratio which aids in removal of impurities during solidification.
- 3) An annual savings of about \$100,000 due to generation of less scrap.

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A waiver is being issued to accept the 9 tons of 8-inch ingot material now on hand at NLO and specification changes will be made to permit the use of 8-inch diameter ingots on a production basis.

Dry-Blend Enriched Cores

In September, HAP0 began receiving production quantities of "dry-blend" recycled enriched ingot cores. With "dry blend" ingots, NLO prepares constant weight packages of UF_4 prepared from HAP0 UO_2 (322.8 lb.) and Paducah UF_6 (102.2 lb.) which is blended in the dry state with 76 lb. of Mg and charged into a reduction bomb. These weights are based on isotopic analyses of 0.851% from HAP0 and 1.25% from Paducah. With this method of blending of small batches of UF_4 , with a possibility of non-constant isotopic UO_2 from HAP0, it was believed that a likely potential existed for producing isotopically out-of-specification enriched metal. Therefore, one core from each "dry-blend" enriched ingot is being tested in the Hanford Test Reactor (HTR). To prevent the number of tests in the HTR from becoming excessive, composite diH limits were established for testing three enriched cores per stringer. If the diH value falls outside of the composite limit, the three cores are tested individually and all of the cores from an ingot whose HTR sample falls outside of the individual acceptance limit are rejected and returned to NLO.

Vacuum Degassing Enriched Uranium

Considerable effort has been devoted to improving the quality of enriched fuel elements. One phase of this program is to improve the quality of the enriched cores received from NLO by reducing the surface hydrogen content. Surface hydrogen may be evolved during the canning operation resulting in porous braze layers and poor quality welds. In November, 1962, NLO was authorized to vacuum degas enriched uranium in hollow core blank form following beta heat treatment. The conditions specified for vacuum degassing are as follows:

- 1) Temperature - $950 \pm 5^\circ F$ ($482 - 538^\circ C$)
- 2) Time - 8 hours, minimum
- 3) Pressure - 0.4 mm of Hg, maximum

At the present time NLO does not have the capacity for degassing all of the enriched cores and until sufficient degassing equipment is available, HAP0 has requested that as much of the enriched core blanks as possible be degassed, especially the OIIIE model.

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Ingot Charge Composition

In October, 1961, an agreement was made with NLO to group the normal and enriched ingots into identifiable lots according to the briquette content for shipment to HAPO as follows:

- Class X - 23 through 27% briquettes
- Class Y - Less than 23% briquettes
- Class Z - Greater than 27% briquettes
- Class W - Unidentifiable cores, such as bar ends, regrinds, Class II OD, Class III H₂, and waived material.

By mid-September, 1962, approximately 1-1/4 million cores with known charge make-up had been shipped to HAPO and it was agreed to segregate future cores into only two categories (X and W) until irradiation results could be analyzed. Since only 20 columns of segregated cores, all Class X, have been discharged and evaluated, it will be several months before any irradiation results can be compared between charge compositions. As shown in Table III, there is very little difference in average grain size for the three classifications; however, there is a difference in UT-2 reject rates. Most of the UT-2 rejects in normal ingot cores are for large grains, whereas, in enriched ingot cores most of the rejects are difference and peak rejects, which usually increase as the grain size decreases. Thus, the trends in UT-2 reject rates would be expected if the impurity content increased with briquette content.

Fuel Performance

Production Rupture Summary

Twenty-one (21) natural and twenty (20) enriched production fuel elements failed in the Hanford reactors during the period from August 1, 1962 through December 31, 1962. Irradiation and rupture classification data on these failures are summarized in Table IV.

Rupture Performance Trends

Rupture resistance (q) of standard non-bumper I&E enriched and natural fuel versus time is shown in Figure 2. These data indicate that the rupture resistance of the standard natural fuel has declined somewhat during the last six to nine months. During the same period enriched rupture resistance has generally continued to improve, to the extent that the two fuel types are essentially equivalent.

TABLE IV

PRODUCTION RUPTURES
I&E FUEL ELEMENTS

Date	Reactor	Type of Fuel Element	Exposure		Tube Power At Failure (kw)	Rupture Classification
			(MWD/T)	(% Goal)		
8-13-62	B	Natural Non-Bumper	404	(75)	1134	Side Hot Spot
8-15-62	H	Enriched Bumper	1193	(98)	429	Hole
8-17-62	DR	Enriched Bumper	948	(115)	1134	Hole
8-22-62	B	Enriched Bumper	1024	(99)	1090	Unclassified
8-27-62	D	Natural Bumper	902	(111)	1122	Hole
9-5-62	B	Natural Non-Bumper	588	(103)	1157	Side Hot Spot
9-8-62	B	Natural Non-Bumper	385	(62)	1018	Side Hot Spot
9-12-62	H	Natural Bumper	685	(112)	982	Side Other
9-16-62	DR	Enriched Bumper	45	(5)	1038	Mech. Damage
9-28-62	DR	Enriched Bumper	698	(83)	1037	Hole
9-29-62	B	Enriched Bumper	810	(103)	1183	Side Other
10-2-62	DR	Enriched Bumper	874	(92)	1016	Unclassified
10-5-62	C	Enriched Non-Bumper	316	(33)	1117	Side Other
10-7-62	DR	Enriched Bumper	791	(96)	1056	Hole
10-10-62	B	Enriched Bumper	858	(93)	1181	Side Other
10-12-62	KW	Enriched Non-Bumper	268	(34)	1687	Side Hot Spot
10-13-62	C	Enriched Non-Bumper	413	(67)	1364	Side Other
10-15-62	D	Natural Bumper	877	(106)	1169	Hole
10-19-62	D	Natural Bumper	868	(104)	1182	Hole
10-20-62	C	Enriched Non-Bumper	659	(92)	1257	Unclassified
10-24-62	F	Natural Non-Bumper	416	(74)	1081	Side Hot Spot
10-27-62	C	Natural Non-Bumper	404	(72)	1209	Unclassified
10-27-62	D	Natural Bumper	779	(90)	1172	Hole
11-2-62	D	Natural Bumper	786	(93)	1153	Hole
11-3-62	F	Enriched Non-Bumper	593	(73)	998	Side Other

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TABLE IV (Continued)

PRODUCTION RUPTURES
I&E FUEL ELEMENTS

<u>Date</u>	<u>Reactor</u>	<u>Type of Fuel Element</u>	<u>Exposure</u>		<u>Tube Power At Failure (kw)</u>	<u>Rupture Classification</u>
			<u>(MWD/T)</u>	<u>(% Goal)</u>		
11-4-62	F	*Enriched Non-Bumper	605	(96)	1373	Side Hot Spot
11-11-62	KW	Natural Non-Bumper	416	(57)	1662	Side Hot Spot
11-13-62	DR	Enriched Bumper	950	(106)	1137	Hole
11-14-62	C	**Natural Non-Bumper	523	(89)	1296	Unclassified
11-14-62	F	Natural Non-Bumper	539	(93)	1241	Side Other
11-22-62	F	Natural Non-Bumper	591	(101)	1085	Side Other
11-26-62	C	*Enriched Non-Bumper	778	(100)	1262	Side Hot Spot
11-27-62	D	Natural Bumper	783	(100)	1214	Hole
12-7-62	D	Enriched Bumper	736	(96)	1261	Hole
12-7-62	F	Enriched Bumper	910	(109)	1130	Side Other
12-13-62	B	Natural Non-Bumper	591	(105)	1281	Side Hot Spot
12-22-62	D	Natural Bumper	836	(102)	1279	Hole
12-23-62	B	Natural Non-Bumper	459	(74)	1134	Unknown
12-27-62	D	Natural Bumper	657	(80)	1240	Side Other
12-30-62	F	*Enriched Bumper	1180	(111)	1020	Side Other

*Second failure in lot.

**Water-mix piece failed.

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Figure 2

The incidence of standard bumper enriched and natural fuel element failures increased markedly during the past six months. Available data indicate that groove and/or ledge-type corrosion (both are extremely non-uniform types of attack noted on HAPO fuel elements) may have caused the great majority of the bumper fuel failures during the past two quarters. This mechanism has caused failures on both the external and internal fuel element surfaces, producing ruptures classified as side-other and hole failures. Spire (hole) failures that are believed to be corrosion induced, characteristically have been incurred at higher exposures (greater than 750 MWD/T). Annular (side other) failures that are believed to be corrosion induced begin appearing at somewhat lower exposures (550 MWD/T). The annular type of failure has been incurred in both bumper and non-bumper fuel. To date, all of the failures of this type, however, have been in charges irradiated in relatively new process tubes not containing water mixing pieces.

Previous theoretical studies have indicated that the current mathematical rupture model would probably not adequately describe the effects of power, temperature, and exposure on the probability of failure for "non hot-spot" corrosion mechanisms. This work indicated that the failure rate would probably be more highly sensitive to temperature and exposure. The rather extreme sensitivity to temperature (for constant residence time failure probability might increase an order of magnitude or more for a 10°C increase in temperature as opposed to two-fold under the current model) is not at variance with the observed reactor experience. This being the case, the reactor performance of fuel elements failing due to corrosion could be strongly affected by traditional measures of fuel quality (i.e., dimensional stability and bond integrity, in that both influence the true fuel surface temperature) as well as by reactor oriented factors such as coolant corrosivity, operating temperatures, etc.

Post-Irradiation Warp and Dimensional Changes

In mid-1961, the method for computing fuel element distortion was revised in order to detect phenomena other than simple bending of the fuel elements.⁽¹⁾ In programming this method for the computer, an error was committed which, in some specific situations, caused the warp of individual fuel elements to be underestimated. This in turn caused the average warp for groups of fuel elements to be underestimated. This error was discovered shortly after the last

(1) HW-70643, "Describing Fuel Element Distortion using Orthogonal Polynomials," J. L. Jaech, August 4, 1961 (Unclassified)

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working committee report was prepared. The programming error has since been rectified, and the affected data have been corrected. A portion of the revised curve of fuel element warp versus canning date is shown in Figure 3. Essentially the portion of the curve from approximately mid-1960 canning date forward was adjusted upward by $0.25\sqrt{\text{mils}}$. Thus, the reduction in warp beginning about that time, while highly significant, was not as large as had been previously reported.

Also shown on Figure 3 are trends in TFC and ΔOD_2 versus canning date. The aforementioned programming error had no effect on these measurements.

External Bond Quality

Figure 4 shows trends in external total bond count versus time for standard enriched and natural I&E material. Strong cyclic tendencies are indicated in the case of the natural material. Both the enriched and natural material passed through minimums in early 1962 canning. As noted previously, the relationship between enriched and natural (q's) (see Figure 1) continue being consistent with the relationship between the enriched and natural bond count data. This can be readily seen from inspection of the data.

Bumper Fuel Program

Approximately 410,000 natural and 225,000 enriched bumper fuel elements have been discharged at variable goal exposure (650-1150 MWD/T), incurring 55 ruptures. None of these ruptures have been of the accelerated external fuel cladding corrosion (hot spot) type bumper fuel was designed to prevent. As a result of an increased incidence of "hole" and "side other" ruptures, the performance of bumper fuel declined in October, 1962. The average ratio of rupture resistance "q" for non-bumper fuel to that for bumper fuel indicates the present performance of bumper fuel is about 1.8 times better than non-bumper fuel. Adjusting for differences in severity of service, the resistance of bumper fuel to ruptures other than the "hot spot" type is essentially equivalent to non-bumper fuel.

Bumper fuel loadings currently comprise the D, DR and H reactors, and the enriched fringe of B and F reactors. Because the charge-discharge rates for bumper fuel are about 50 percent less than for non-bumper fuel, use of natural bumper fuel in the control zone of DR reactor has not proven to be economically attractive and is being discontinued. Pending necessary authorization, a second H reactor E-N (enriched uranium bumper fuel and Li-Al alloy pieces) demonstration loading is scheduled early in CY 1963.

FIGURE 3 - DIMENSIONAL STABILITY TRENDS WITH TIME

FIGURE 4 - EXTERNAL BOND QUALITY TRENDS WITH TIME

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Based on favorable preliminary irradiation test results and the lower hydraulic resistance offered by an elliptical bumper rail over the present rectangular bumper rail design, production transition to the elliptical rail design is planned by July, 1963. Testing and debugging of a prototype automatic feeder to feed elliptical rails into the ultrasonic welders in pre-stamped strip form is complete except for minor adjustments.

Self-Supported Fuel Program

The irradiation of self-supported fuel elements (CIVNS and CVNS) in the ribless zircaloy, 69-tube, C reactor test facility progressed without incident during this period. About 15,000 fuel elements have been discharged to date at variable goal exposure (850 MWD/T), sustaining one external cladding corrosion failure as a result of broken and damaged rails of a former design.

Planned replacement of 5200 existing K reactor ribbed aluminum tubes with ribless zircaloy tubes is currently scheduled to begin by mid-February, 1963 and be completed on or about year end. Assembly of the natural (KVNS) and enriched (KVES) fuel models required for this program has been initiated. In order to provide projection rails with a higher strength on an interim basis until more optimum rail designs are developed, the thickness of the rails has been increased from 45 mils to 50 mils. A three-dimple collapsible bridge rail design will be used initially on the KVN fuel element. Selection of a final rail design for the KVE fuel element, which requires a shorter rail, is awaiting completion of a test to compare the relative compressive strength of a two-dimple collapsible bridge rail and a single-dimple arch-type rail. Post-sizing techniques after rail welding have been developed and are being demonstrated on a production basis as a means of controlling maximum rail projected diameter and minimum height within the specified limits for in-reactor temperature and flow balance.

Overbore Program

Irradiation testing of self-supported (CVINS) overbore fuel elements is continuing in 62 ribless zircaloy tubes in C reactor. Approximately 4,300 fuel elements have been discharged to date at exposures ranging from 200 to 1,000 MWD/T, with the majority of the tubes in the range of 600 - 900 MWD/T. Single ruptures were sustained in a fourth and fifth overbore column early in September, 1962, at exposures of 710 MWD/T and 661 MWD/T, respectively. One was classified as "side other" and resulted from broken rails of a former design. The other rupture was the second core split in a dingot lot. Because dingot uranium fuel elements have proven to be generally more rupture prone in the Hanford reactors, the exposure of dingot overbore fuel has been reduced to 75 percent of variable goal (800

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MWD/T) until the present inventory (~ 20 tons) of alpha extruded dingot overbore cores is exhausted. Alpha rolled ingot cores are being obtained to meet future overbore fuel requirements.

A new overbore fuel model (CVIIINS) has been proposed, and it is planned to assemble about 200 of these fuel elements for irradiation testing on or before August, 1963. This model achieves a 9 percent lower elastic core stress with only a 2 percent loss in reactivity and no appreciable loss in conversion ratio. The nominal external dimensions are essentially unchanged from the present CVINS fuel design; however, the core ID and spire wall have been increased as shown below.

<u>Fuel Model</u>	<u>Finished Fuel Dimensions</u>		
	<u>O.D.</u>	<u>I.D.</u>	<u>Length</u>
CVINS	1.986	0.339	8.865
CVIIINS	1.986	0.039	8.865

<u>Fuel Model</u>	<u>Core Dimensions</u>		
	<u>O.D.</u>	<u>I.D.</u>	<u>Length</u>
CVINS	1.883	0.452	8.325
CVIIINS	1.833	0.563	8.325

AlSi Process Development

Dingot Process Optimization

Major development effort in the AlSi process over the past six months has been directed toward developing canning conditions for dingot uranium which would produce fuel elements that had bonding characteristics similar to ingot uranium fuel elements. It is believed that at least part of the poorer performance of dingot fuel as compared to ingot fuel could be attributed to the fact that the bond between dingot uranium and AlSi does not form as readily as with ingot uranium and that the resulting bond, when it does form, is more brittle. Ingot and dingot metal in the past have both been canned by the same process. Development effort was directed toward modifying (or optimizing) the AlSi process for dingot uranium to produce dingot fuel elements that were the equivalent of ingot fuel elements in those characteristics which could be measured before irradiation and which were believed to be crucial to the performance of the fuel element in the reactor. The amount of UAl_3 compound layer as compared to the amount of more brittle USi_3 compound layer in the canned fuel element and the total bond count were considered to be critical measures of fuel quality.

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A great deal of development effort in both the pilot plant and the production line was carried out and large numbers of fuel elements were destructively examined for bond metallography. It was found in the development program that compound layer structure for dingot could be made equivalent to ingot by reducing the silicon content of AlSi in the duplex furnace from 9.0 to 7.5 percent and by increasing the duplex lead temperature from 616 to 625°C. These conditions were applied to the production process and canning was initiated on some 400 tons of dingot fuel cores on hand at Hanford. However, after two days of canning, processing on the dingot was discontinued because of excessive (up to 25%) reject rates for internal bonding on one line. The critical need to devote development effort to other pressing programs which had been put aside while the dingot process was studied, necessitated setting aside the dingot program and no additional information regarding the cause for poor internal bonding is presently available.

Effect of Dingot Uranium Heat Treatment on Nitric Acid Dissolution

Rates and Compound Layer Formation

Early in the investigation of the differences of ingot and dingot uranium, it was determined that the dingot metal had a slower nitric acid dissolution rate. The etching mechanism is greatly dependent on the impurities in the metal. These impurities are activation sites for the acid attack. Dingot metal has less activation sites than ingot. Mallinckrodt Chemical Works were able to improve the etching rate of dingot by precipitation of two phases in the additive dingot metal. The etch rate depends on the number, size, and location of these particles. Four types of dingot uranium cores were prepared for evaluation and testing at HAPO. The testing involved five groups which were:

- 1) Alpha extruded dingot cores, normal beta heat treatment in hollow form and oil quenched. These cores should contain small amounts of the precipitated phases.
- 2) Alpha extruded dingot cores, long (45 minute) beta soak period to dissolve nearly all of the precipitated phases.
- 3) Alpha extruded dingot cores, normal beta heat treatment in hollow form, oil quenched, vacuum annealed at 950-1000°F for 8 hours to precipitate a maximum of the phases.

- 4) Rolled dingot cores, normal beta heat treatment in solid form, brine quenched, finished machined cores, vacuum annealed at 950-1000°F for 8 hours.
- 5) Normal ingot cores from storage. These elements were for test controls.

Table V is a summary of the nitric acid etch rate tests.

TABLE V

PWR No. 91-48-Effect of Heat
Treatment of Nitric Acid Dissolution Rates

<u>Group No.</u>	<u>Average Weight Loss (gm/min)*</u>	<u>Average OD Change (mils/min)</u>
1	1.27	1.47
2	1.30	1.60
3	1.40	1.83
4	1.77	2.23
5	1.33	1.43

56% HNO₃ at 60°C

*Etch rates were calculated between 3 and 6 minutes each time.

Experimental results were nearly as expected. The etch rate of dingot cores can be increased by precipitation of the U₆Fe and U₃Si phases. Group 2 (long beta soak) should have dissolved most of the precipitant. Group 4 had a high etch rate due to pitting during etch. This pitting was due to an oxidized surface (the cores were vacuum annealed in the finished machined core dimension whereas the cores of Group 3 were machined after this treatment).

The nitric acid dissolution rate of dingot uranium is important only if there is a relationship between chemical activity and the diffusion activity of AlSi and uranium. Table VI is a summary of a test performed at HAPO with the groups of cores of the same heat treatments described above. All cores were etched in the standard nitric acid pickle bath for 4-6 minutes and canned by the standard AlSi process and the modified process for dingot uranium.

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Metallographic Data								
Average Percentage UAl ₃ -Type Structure								
Present Process (9.5% Si)					Modified Process (7.5% Si)			
<u>Present Process (9.5% Si)</u>			<u>Ext. Bond</u>		<u>Modified Process (7.5% Si)</u>		<u>Ext. Bond</u>	
<u>Int. Bond</u>					<u>Int. Bond</u>			
<u>Group</u>	$\bar{X}$	Range	$\bar{X}$	Range	$\bar{X}$	Range	$\bar{X}$	Range
1	88.8	47	54.9	69	98.9	6	87.1	39
2	90.6	43	41.3	80	99.2	4	84.4	61
3	96.5	11	91.5	19	98.9	8	96.6	27
4	95.7	13	97.9	12	99.6	3	99.7	3
5	96.5	12	82.6	41	99.8	1	94.1	26

There is some indication of an effect of the presence of the precipitant on the external bond. Groups 1 and 2 had the least precipitant and the average of the UAl₃-type structure on the external bond was significantly lower and the range was greater for these groups. The internal bond was affected little by the presence of the precipitant. The bonds produced by the present AlSi process were much better than expected for dingot uranium. The bonds produced by the modified process were better for all groups, indicating more nearly optimum conditions for both ingot and dingot cores. Additional testing is necessary to establish true differences.

Alternate Process Development

The evaluation of an alternate manufacturing process for the production of fuel elements for the Hanford production reactors exclusive of the NPR is continuing. The alternate manufacturing processes considered all use an electro-nickel plated uranium core with an aluminum alloy cladding. The four processes that have been investigated are: (1) HAPO hot press, (2) Sylcor hot press, (3) fluid pressure bonding, and (4) hot-die sizing. Studies have been completed on the economics and production feasibility of these processes with hot-die sizing being the most favorable closely followed by fluid pressure bonding. Therefore, the principal effort is being centered around these two processes.

Fluid Pressure Bonding

A test to evaluate the bonding parameters for producing diffusion bonded fuel elements by fluid pressure bonding is in progress. Contract negotiations were completed with the Battelle Memorial Institute during July, 1962, to produce 128 fuel assemblies under various bonding parameters of time, temperature and pressure. The fuel elements were assembled at HAPO during December, 1962. The assemblies will be shipped to BMI in January for bonding and returned to HAPO for evaluation.

Hot-Die Sizing

The development of the hot-die sizing process has been centered around designed tests of the bonding parameters. Two test series have been completed on the die sizing step and the end bonding step. Nineteen (19) response surface diagrams have been developed relating the bonding conditions to fuel element quality attributes.

In the die sizing step the process variables of reduction ratio, preheat temperature, preheat time and sizing or ram speed were investigated.

<u>Variable</u>	<u>Range</u>
Preheat Furnace Temperature	650, 680, 710°C
Preheat Furnace Time	8-1/2, 10, 11-1/2 min.
Aluminum Reduction Ratio	17, 45, 63%
Ram Speed	40, 65, 90 inches/min.

The results of the die sizing test permitted the establishment of a process cycle specification as follows:

Preheat Furnace Temperature	= 680 ± 15°C
Preheat Furnace Time	= 10 ± 1/2 min.
Sizing Speed	= 75 ± 5 ipm
Reduction Ratio	= 63%

Two of the more interesting response surfaces showing the effect of time, temperature and reduction ratio are shown in Figures 5 and 6. The results in Figure 5 show that the OD bond strength measured one inch from the cap end of the fuel element varies linearly with the reduction ratio with no apparent effects due to time or temperature. In Figure 6 the bond strength at the center O.D. varies as a function of the reduction ratio but there is a temperature interaction, too.

The end bonding test was divided into two parts. The following table is a summary of the range of investigation:

Part I

<u>Variable</u>	<u>Range</u>
Pressing Time*	30, 105, 180 seconds
Bonding Die Temperature	560, 600, 640°C
Pressure	1.03, 1.38, 1.73 T/in. ²

This time is greatly reduced because the fuel assembly temperature is raised to ~ 400°C by an initial preheat in an auxiliary furnace at 680°C for 5 minutes.

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FIGURE 5 - HOT DIE SIZING TEST - EXTERNAL BOND STRENGTH MEASUREMENTS AT FUEL ELEMENT CAP END

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FIGURE 6 - HOT DIE SIZING TEST - EXTERNAL BOND STRENGTH MEASUREMENTS AT FUEL ELEMENT CENTER

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FIGURE 7 - END BOND TEST WITH PREHEAT CAP END TOTAL BOND COUNT

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FIGURE 8 - END BOND TEST WITHOUT PREHEAT. CAP
END TOTAL BOND COUNT

Part II

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<u>Variable</u>	<u>Range</u>
Pressing Time	5, 7, 9 minutes
Bonding Die Temperature	560, 600, 640°C
Pressure	1.03, 1.38, 1.73 T/in. ²

The end bond test has yielded valuable technical information for determining process specifications. However, the results of the end bonding test are based upon the containment die principal and not the differential expansion die concept. There are three significant process problems in using the containment die concept: (1) the die tends to gall and upset the cladding, (2) the pressing pressure is limited due to a tendency for back extrusion, and (3) the tendency for lateral bond deterioration. The preliminary runs with the advanced differential expansion die indicate that these problems can be eliminated. A more extensive test is planned using the differential expansion method. The results of the first end bond test did yield valuable information in the time, temperature and pressure requirements to successfully form the solid state end bond. Two of the response surfaces are shown in Figures 7 and 8. The relationship shown is the effect on the ultrasonic total bond count (taken on the flat end of the fuel element) as a function of time, temperature and pressure. The results in Figure 8 compare with the results in Figure 7 where a supplementary preheat in a furnace set at 680°C for 5 minutes was used to shorten the end bond press cycle. The results in both cases indicate that the optimum (lowest) bond count was achieved at the maximum time, temperature and pressure conditions. Caution should be used in applying these results directly in that the parameter conditions that minimize or maximize one quality attribute may not apply in all cases. The end bonding cycle conditions have not been established because of the failure to achieve satisfactory fuel element lateral bonding and surface cladding conditions.

Some typical photomicrographs of the diffusion bonds achieved in this test series are shown in Figures 9, 10, and 11.

In Figure 9 the lateral bond cracking that occurs just outside the containment die is illustrated. This cracking at first appeared to be a function of time and temperature in the press; however, closer examination of the processing conditions indicate that it is really a function of the fit between the fuel element and the containment die.

In Figure 10 the diffusion structures on the end of an end bonded piece are shown. The non-uniformity of the diffusion is a result

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Negative No. 6910-C-2

1000X

FIGURE 9 - LATERAL BOND CRACKING IN THE ALUMINUM-NICKEL
DIFFUSION LAYER OCCURRING OUTSIDE THE CON-
TAINMENT DIE ON AN END BONDED FUEL ELEMENT

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FIGURE 10 - TYPICAL END BOND SHOWING NON-UNIFORM
DIFFUSION DUE TO THE PRESENCE OF
ALUMINUM OR NICKEL OXIDE

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FIGURE 11 - TYPICAL EXTERNAL BOND ON A HOT DIE SIZED FUEL
ELEMENT

of oxide pockets on the surface of either the nickel or aluminum. The bond is believed to be acceptable.

In Figure 11 a typical external lateral bond is shown depicting the uniformity of the diffusion structure normally achieved.

The metallography of the closure zone has shown good bonding in almost every instance. The closure does not appear to be a problem at this time.

The completion of the next end bonding test should lead to a reliable end bonding process specification.

Irradiation of the first reactor grade hot-die size bonded fuel elements will then be initiated.

NPR FUEL DEVELOPMENTFuel Production Status

Fuel loading of the N-reactor is scheduled for April 15, 1964. Completion of fabrication of a full reactor load of N fuel elements (approximately 450 tons) will be required by February, 1964. This schedule is a modification of the one reported previously.

The initial reactor load will consist of about 90% tube-in-tube elements of 0.947 enrichment. About 10% of the load will be of natural (0.71) enrichment to be used for poison to control reactivity. The .94% enriched fuel will be nominally 24-inch length, with 10-15% acceptable as cut down multiples (18-inch and 12-inch). The natural fuel will be nominally 20-inches in length. In addition, some quantities of the fuel will have water mixer devices affixed of a design not yet determined.

To date approximately 800 outer and 650 inner extrusions have been made. Approximately 125 tons of fuel elements are in the process stream. Only about 25 tons are in finished storage. The lack of satisfactory supports is the primary reason for the relatively small quantity of completely finished elements at this time. Present plans call for resumption of full scale finishing operations about February 1, 1963, when supports of a modified design become available. It may be necessary to run the finishing operations (support welding, etching, autoclaving, and final assembly) on a two-shift basis to work out the backlog of material now awaiting supports.

At the present time the following station reject rates are being experienced:

<u>Rejection Cause</u>	<u>Average Rejection Rate</u>
Bonding (Peel Test)	1%*
Over-all Wall Thickness	1%
Cladding Thickness-Outer Tube, Inner Clad	2%**
Cladding Thickness-Other Clads	1%
Warp-Inner Tube	6%
End Closure	10%
Chemical Milling	1%
Welding of Closure	2%
White Oxide After Autoclave	No material autoclaved

* This figure does not include the unexplained bonding problem with the inner clad of the inner tubes encountered early in December.

** Using newly established limit of .012" circumferential variation. With the old range of .008" maximum variation, the rejection rate would be 15-25%.

Exact over-all yields cannot be determined at this time, but it appears that the material now in process will be completed with a total yield of 70-80%.

KER Loop Testing Program

The schedule on the included table shows the tentative test plans for the KER loops. It will be noted that the tests to be run are essentially as reported previously except that the schedules have been moved back due to equipment problems which have resulted in extended outages.

Loops 1 and 2 have been recently modified to provide increased water flow and heat dissipation capacity. These loops have zircaloy process tubes with approximately 2.10 inches inside diameter which precludes the testing of full scale NPR elements. It is possible to test NPR inner tubes (NIE) by fitting them into thin walled zircaloy shroud tubes. Specially designed elements, such as KSE- (single tube, 1.6% enriched) have been fabricated to simulate NPR fuel conditions.

Elements now under irradiation, or scheduled for irradiation, include the V-notch end closure test, the 1.6% enriched KSE-5 test and the crud test. The crud test is a redesigned thermocoupled element to study crud formation during irradiation.

Loops 3 and 4 are scheduled exclusively for testing prototypic assemblies of N-fuel. These two loops were recently modified by the installation of full size N tubes and new pumping and heat exchange facilities. These loops have been plagued with pump troubles for a number of months and little meaningful data have been gained. One charge of 14 NAE-I fuel elements (prototypic NPR) did reach an exposure of 1250 MWD/T under prototypic conditions. There was no evidence of any damage or deformation visible after discharge.

Process Development

Materials Development and Procurement

Uranium Billets

The inspection of N-billets received at HAP0 has resulted in essentially no rejection for metal quality defects. Some dimensional rejects (approximately 1%) have been detected for excessive wall thickness variation at the center of the billets. These rejects were related to the much higher rejection rate

TENTATIVE LOOP SCHEDULE
12/20/62

KER Loop Number	CY 1963												
	December	January	February	March	April	May	June	July	August	September	October	November	December
Loop 1	Decontamina- tion Test	Crud Test (HLO) Four 17" NIE-1 Heater Fuels	12 KSE-5 Fuels (Unalloyed)			Crud Test HLO Four 17" NIE-1 Heater Fuels		12 KSE-5 Fuels (Unalloyed)					
Loop 2	Tapered End Closure Test. NIE 5 Pcs. Brazed 5 Pcs. Not Brazed			6 U-2 w/o Zr and 6 NIN-1 or Target Element									
Loop 3	Pump Repairs	14 NAE-1's											
Loop 4	14 NAE-1's (Prototypic Assemblies)						14 NAE-1's						

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(approximately 10%) for this cause at NLO. The wall thickness variation is caused by warp of the primary extrusion. The OD surface of the billet is machined to a right circular cylinder but the machining tool follows the contour of the as-extruded inner hole. Wall thickness variation results if the original billet blank was warped prior to machining.

The uranium billet production route consists of casting 11-inch diameter by 28 inch long hollow ingots; triple-beta heat treating; alpha extruding to rough size; cutting to length and finish machining to final coextrusion billet size with a square ended configuration. The recent process changes, production problems, and new development activities are as follows:

1) Billet Heat Treatment

As explained in the last report, single beta heat treatment of the billet blank was instituted based on limited data which indicated an improvement in clad thickness control. Control of clad thickness variation of the inner clad of the outer tube is still a major coextrusion problem. These data had been obtained from coextrusion of billets single-beta heat treated in finished form at HAP0. Concurrent with the process modification for single-beta heat treatment of the billet blanks, a test was designed to compare three other billet treatment routes.

The analyses of the results from this test showed that better clad thickness control could be obtained if the billet blanks were not single-beta heat treated. As a result, beta heat treatment of the billet blanks was discontinued October 8, 1962. The other two billet treatment routes tested were (1) an increase of 0.3 inches in upset of the ingot ID during primary extrusion followed by single-beta heat treatment of the billet blanks, and (2) rough machining of the billet blank before single-beta heat treatment followed by finished machining. Neither of these process routes showed any significant improvement over single-beta heat treatment of the billet blanks.

The billets heat treated at HAP0 yielded the best results to date for clad thickness control. The one known difference between these billets and those heat treated at NLO was that the billets at HAP0 had been finish machined prior to heat treatment. This resulted

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in an as-heat treated surface in contact with the zircaloy. This effect is now being tested. Nineteen (19) billets finished machined on the ID before single-beta heat treatment at NLO are now being coextruded along with non-heat treated control billets.

2) Uranium Alloy Development

The clad thickness control problem is believed to be related to the grain structure of the billet at the time of coextrusion and also to the structure existing after casting of the ingot. Iron and silicon were added to the uranium melts starting in May, 1961, primarily to refine the structures and control clad thickness variation. A short-range program has been started to determine if minor increases in silicon will result in further gains. Test billets containing 140-190; 190-250; 400-450 ppm silicon have been ordered and will be coextruded. Half of the total number of billets in these tests will be single-beta heat treated after ID finishing to determine if additional gains can be made from this process.

Silicon was selected as the additive material because:

- a) Extensive irradiation experience of alloys has been obtained at HAP0 only with iron and silicon additives. If appreciable gains are obtained in clad thickness control from use of silicon additives, conversion of production to the new alloy would be planned before additional irradiation experience could be obtained.
- b) Silicon is as effective for grain refinement as iron based on grain size studies made at HAP0 with I&E uranium cores.
- c) Silicon is less apt to migrate at the high alpha phase temperatures the fuel will encounter during coextrusion and irradiation according to information developed at Malinckrodt.

The longer range alloy development work includes an investigation of iron, silicon, and aluminum in binary and ternary systems. The effect of these alloys on grain refinement and fabricability characteristics will be studied. The more promising alloys resulting from these studies will be analyzed for characteristics influencing irradiation stability.

An N-outer ingot containing 640 ppm Al and 400 ppm Fe was processed through ingot heat treatment, primary extrusion, billet heat treatment, and coextrusion to N-outer tube. Sections of the coextrusion have been studied to evaluate the effect of (1) alpha treatments from 1-24 hours at temperatures from 450-640°C following beta treatment; (2) effect of time in the beta phase (730°C) on the clad-core bond, U_6Fe and UAl_2 size and dispersion, grain size, and hardness. No significant change in room temperature hardness resulted from either complete solution of Fe and Al or extended alpha treatment to cause coarse precipitation of their compounds. Wide variations in the size and distribution of U_6Fe and UAl_2 could be effected by variations in treatment. At high alpha temperatures the U_6Fe phase is very coarse and grain boundary oriented, while the UAl_2 growth is not as pronounced, nor is the precipitate grain boundary oriented. Additional electron microscopy evaluation is being conducted to more accurately determine the precipitate sizes.

3) Ingot Uniformity

The initial fabrication shape in the production of coextruded fuels is the cast uranium ingot. Any non-uniformity introduced at this step is manifested throughout the remainder of the fuel fabrication process. Hollow casting of the ingot results in some eccentricity. Because of this eccentricity and the roughness of the as-cast surface, the primary extrusion must be made oversize to allow about 1/8 inch of metal for machining cleanup and dimensional truing.

Plans have been made to cast solid ingots and drill the hole. This conversion will not occur until equipment can be put in place, but test quantities will be available in February or March. As a further step in demonstrating the benefits of ingot uniformity, the OD will be machined on a test group of ingots and the lead end will be squared and chamfered. The machined OD should cause the ingot to align horizontally in the primary extrusion press. The chamfer should cause the lead end of the ingot to lift on the die cone and center in the container.

Drilling and machining of the ingots will cause chip generation. However, no over-all increase in chip production should result if the added uniformity is used to reduce the cross-section of the primary extrusion and reduce the chip generation from billet finishing.

DECLASSIFIEDFuel Element Supports

Fuel element supports, both inner and outer, have continued to be the key problem area in the N-fuel fabrication. The fundamental problem has been inability to make supports of the designs desired from commercially rolled zircaloy-2 strip. It appears that the problem was one of not recognizing the importance of crystallographic texture and its relationship to bendability in zircaloy strip. Previously used strip was rolled with various intervening anneals and stress reliefs. These latter were designed to improve ductility, but the effect was always marginal.

The laboratory program to investigate fabrication methods for N-inner supports has consisted of development work to improve the bend ductility of zircaloy-2 sheet as well as the development of support forming procedures. Fabrication schedules designed to control the crystallographic texture of the sheet have produced laboratory quantities of sheet with greater ductility than is normally found in commercially produced sheet. Procedures based on rolling sheet from extruded bar or tubing have been the most effective in producing a ductile product. A support forming method has been developed which utilizes the electrical resistance of the part for heating during forming. The sheet fabrication and support forming procedures are being used to manufacture a pilot quantity of inner supports for use on N-reactor fuel elements.

1) Inner Tube Supports

The inner support design is essentially the same as discussed in previous reports, i.e., the "buggy spring." The key problem was to find methods of fabrication and it appears that one or more of the processes discussed will provide the strip material. Thus, the main technical problems appear to be solved. At the present time the HLO facilities are producing supports. It is planned that 20,000 will be produced on-site and then industrial supplies will be available.

2) Outer Tube Supports

The outer tube support has consisted of the "suitcase handle" design. However, the properties of impact and crushing strength have been lower than desired. A new support, the "arch" design (Figure 11) is now planned for production use, and tests are nearly completed. The crushing strength of the new support is about twice that of the suitcase handle (approximately 450 lbs., versus 1000 lbs.) when both have their thin steel shoes

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BLDG. NO. 306	INDEX NUMBER 490020			
DWG. NO.	SHEET NO. SHEETS			

DWG. NO.	DRAWING TITLE		
	REFERENCE DRAWINGS		
CLASSIFICATION	CLASS. BY	DATE	
U. S. ATOMIC ENERGY COMMISSION			
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GENERAL ELECTRIC			
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crimped on. The impact necessary for .010 inch permanent deflection is about 0.4 foot-lbs. for the suitcase handle and about 1.0 foot-lbs. for the arch support. The "arch" design is being firmed up and it appears conversion can be accomplished by about February 1, 1963, with initial supplies made on-site.

The "suitcase-handle" supports have been attached to over 1,000 finished fuel elements and will be attached to an additional two or three thousand prior to conversion. KER loop testing has not shown that the suitcase handle is unsatisfactory in-reactor; rather, it is highly susceptible to damage before it gets in place. Thus, extra care will be required in handling those fuel assemblies with the suitcase handle supports.

Coextrusion Process

Uranium: Zircaloy Bonding

A billet design change from conical preshaped uranium billets to square-end uranium billets was made during an inner tube production campaign in late November-early December. Concurrently, a condition of essentially no bonding of the inner tube inner cladding developed.

The general characteristics of this bonding problem differ from bonding problems experienced in the past in that (1) the coextruded clad:core interface is relatively free from contamination (oxides, nitrates), (2) there is no apparent loss of billet vacuum, and (3) approximately 40-50% of the unbonded extrusions have had 6-8 inches of "good" bond at the front and rear of the extrusion.

A number of test coextrusions have been performed in an attempt to isolate the most logical variables. A further series of test coextrusions is planned for early January, in which the tests are designed to isolate variables in the uranium billet geometry, uranium billet quality, billet component cleaning processes, and billet evacuation.

Cladding Thickness

Producing coextrusions which met original clad thickness variation specifications for the outer tube-inner cladding was one of the major fabrication problems in the coextrusion process. Re-evaluation of the requirements of the outer tube-inner cladding has

allowed a change in the circumferential range specification for this specific clad thickness variation from 0.008" to 0.012". With this new specification, rejection of fuel elements for this cause will be in the range of 1-2 percent.

Inner Tube Warp

Warp on the inner tube is at approximately predicted yield levels and is not considered a major problem. Reverse bending limits are being raised to permit straightening of pieces with up to .100" TIR warp as compared to .075" previously. This will improve yields by about 1% over-all.

End Closure Development

Beryllium-Zircaloy Braze Closure

The beryllium-zircaloy end closure process is producing high quality closures which have performed very satisfactorily in all irradiation tests to date. The closure reject rate has been higher than predicted and has been increasing recently. No one single thing can be held responsible for this increase.

It has been established that an intermetallic compound layer, which is felt to be indicative of a good metallurgical bond, forms much more readily between the Be-Zr alloy and the uranium when the uranium is "non additive"; that is if it has no iron or silicon additions. It has not been determined whether it is the iron or the silicon which tends to prevent compound layer formation.

A study is under way to establish a technique for measuring the failure resistance of various closures. The first attempt has been to use alpha-beta cycling of the brazed closure to induce failure. Some failures have been induced by up to 50 complete cycles but the uranium has been so grossly distorted that interpretation of the results are subject to question.

The satisfactory use of Be-Zr brazing alloy made with zirconium rather than zircaloy has been demonstrated on a small scale. This change will effect some saving in cost and may improve the closure quality as well as process yield.

Alternate Closures

1) Gas Pressure Bonded Closure

Development work is continuing on the gas pressure bonded end closure concept. This concept entails

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chemical milling recesses in the ends of the fuel tube sections, inserting closure rings, electron beam sealing the ends in vacuum, and gas pressure bonding the closure rings to the clad and core at a temperature just below the alpha-beta transformation.

It has been demonstrated that zircaloy can be bonded to uranium very satisfactorily at 650°C, 10,000 psi, in one hour if the surfaces have been cathodically etched and kept under vacuum. It has not been possible to get completely satisfactory bonding of zircaloy to zircaloy under the same conditions. About 25% of such closures autoclaved have failed due to incomplete cap:clad bonds. Introduction of a third "diffuser" metal between the cap and clad has been only partly successful in promoting a satisfactory bond.

Closure rings of metals other than zircaloy have been tried in an attempt to find a more likely bonding combination. Pure zirconium closure rings bond more readily than zircaloy, but instances of incomplete bonding have occurred which have resulted in autoclave failures. Also, gross corrosion during 400°C (725°F) steam autoclaving of one group of zirconium closures indicates a corrosion problem may be inherent with pure zirconium. Titanium appears to bond exceptionally well and the closure ring itself has adequate corrosion resistance. The intermetallic compounds which form between titanium and zircaloy are subject to very rapid corrosion. Other metals such as niobium and thorium have been tried but do not appear promising.

Future plans call for use of pressures up to 20,000 psi to promote bonding of zircaloy-zircaloy and zircaloy-zirconium interfaces. Also, novel means of applying thin layers of diffuser metals will be studied.

2) Hot-Headed Closure Studies

Since the last reporting period on the projection welded diffusion-bonded hot-head closure, interface bonding material thickness, methods of preplacing the interface material, and projection design have been explored. Closures were made using several

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thicknesses of copper interface (approximately 0.2, 1.0, 2.0 and 2.5 mils thick) material between the end cap and the uranium with two, four and six ring projections machined on the end caps. Bonds between the uranium and the end cap were obtained with all thicknesses; however, the bonds with the thinnest copper appeared to be the best. Two methods of preplacing the interface material have been used, copper shims cut from foil and plating. Plating appeared to be the more simple and more precise method. Most of the plating has been done by the "immersion plating" process, however, the vacuum vapor deposition process will be attempted. Closures made with two ring projections consistently allowed the uranium to move into the interface areas between the cap and the fuel element cladding. The projection welds on the four ring and the six ring projection caps were sound and prevented movement of the uranium into end cap and element cladding interface. The six ring projection type caps were made in an attempt to eliminate the small voids between projection welds. While continuous welds without voids have been made in small areas, it has not been possible to produce these results over the entire zircaloy-to-zircaloy faying surfaces. Currently effort is being applied to the four ring projection caps utilizing copper as interface material between the uranium Zr-2 and Zr-2 to Zr-2 void areas between the projection welds.

3) Self-Brazed Closure

Continued experimentation has shown that the self-braze procedure developed for forming closures on the NPR inner tube stock produces a high percentage of sound bonds. The principal problem recognized at last writing was that of extrusion of uranium into the annuli between cap and jacket both at the bore and external clad. This problem has been largely overcome by redesigning the cap to provide a 0.075 inch skirt at both inner and outer peripheries, with the uranium core face shaped by mechanical and chemical milling to match the cap contour. When the forge pressure is applied during the closure processing, these skirts are spread out to contact the walls, thus effectively valving off the flow of uranium into the annuli. These skirts further serve

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to anchor the caps more strongly in place, though some attachment strength is presumably sacrificed by giving up the V-notch cap contour.

Preliminary high temperature autoclave tests on both the finished closures and on specimens whose weld beads had been machined off to lay bare the annular bonding layer, have indicated an encouraging degree of corrosion resistance for this braze alloy.

A group of test elements with closures formed by this process are being prepared for in-reactor testing; meanwhile, other tests to determine various mechanical and chemical quality aspects are being pursued.

4) Copper Braze Closure

Further corrosion testing has been carried out on Zr-clad uranium fuel elements closed with a Cu-Zr eutectic braze. Brazed and welded closures and brazed-only closures have been autoclaved for 72 hours in 400°C steam and for 100 hours in 300°C water. No failures have occurred. The welded closures showed slight discoloration but no severe corrosion while the braze alloy in the as-brazed closures was corroded about 0.020 inch, indicating that the braze alloy will corrode but has sufficient resistance to limit the penetration in case of primary weld failure.

Melting point studies on a series of arc-melted alloys has shown the existence of a ternary eutectic at approximately 80 w/o Zr - 10 w/o Cu-10 w/o Ni which melts at about 850°C. The alloy is fluid at 860°C and bonds tightly to both zircaloy and uranium, but may be too brittle for reactor conditions.

Zircaloy Etching and Autoclaving

Earlier reports have indicated that undue amounts of material were being rejected because of unsatisfactory autoclaved surface. Since very little material has been autoclaved since last report, it is not possible to quote any current rejection rates. However, before suspension of this operation, etching and rinsing techniques had been modified sufficiently to eliminate any serious problem which may have existed.

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Irradiation PerformanceKER Loop Irradiation of Tubular Elements

Hydrogen contents of the Zr-2 clad samples from five coextruded fuel elements irradiated in the KER loops to exposures up to 3600 MWD/T have been determined and indicate little pickup during irradiation. The fuel exposures and hydrogen contents of the clad were:

<u>Element</u>	<u>Exp.</u>	<u>Hydrogen</u>
KSE-3	1200 MWD/T	70 ppm
KSE-3	2000 MWD/T	47 ppm
KSE-3	3600 MWD/T	47 ppm
NIE	2300 MWD/T	30 ppm
NIE	3000 MWD/T	25 ppm

Pre-irradiation values of the fuel clad varied from 25-35 ppm and the low pickup during irradiation substantiates the observation that negligible clad corrosion occurred during irradiation.

Warp data has been obtained on five "N" inner fuel tubes (NIE's) irradiated to 3000 MWD/T, which is about 2-1/2 times initial "N" goal for these components. Residual single throw warp on the 24" long elements varied from 0.005 to 0.015 inch. Additional metallographic examination of the Be-Zr eutectic braze weld closures of these elements has revealed no deficiencies or any indications of inadequacies. A total of 32 NAE's ("N" assembled, enriched - NPR fuel element prototypes) have been superficially examined and weighed for fuel swelling evaluation after attaining exposures up to a maximum of about 1700 MWD/T in modified KER loops 3 and 4. The components were observed to be free of severe warp, fuel bumping, clad wrinkling, clad thinning or longitudinal striations, and no unusual corrosion behavior had occurred to the clad. Some of both the inner (buggy spring type) and outer (suitcase handle type) supports were deformed, cracked, partially missing or completely gone. However, it is probable that most, if not all, of these observed shortcomings of the supports resulted from discharge operations and subsequent handling in the basin. The Be-Zr eutectic braze-weld closures were generally discolored from grey to white, with the degree of discoloration increasing in the downstream or higher coolant temperature direction.

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A maximum fuel swelling of approximately 1% was observed and swelling in the outer tubular component was approximately 25% greater than in the inner tube component. The measured swelling compares favorably with the swelling predicted by a swelling model developed from earlier data obtained from KER irradiations of KSE-3 elements. The model was developed by K. R. Merckx and is based upon the temperature, burnup, and pressure dependent swelling behavior of an incremental volume of fuel. The model is expressed -

$$V = AB + \frac{GBT}{PC + \theta_1 B \theta_2 T \theta_3 [1 + e^{-\theta_4 (T + \theta_5 B - \theta_6)}] + \theta_7 TR}$$

V = v/o increase of a volume element

A = constant = 3.0

B = a/o burnup

G = constant = 550

T = °K/273

PC = coolant pressure, atmosphere

TR = ratio, outer clad radius to thickness

θ_1 to θ_7 , parameters

Parameters 1 through 6 in the above model have been statistically evaluated using a computer program to obtain the best agreement between the model and the experimentally determined KSE-3 fuel element swelling data. Values of the parameters are -

$\theta_1 = 19.5067$	$\theta_5 = 0.11204$
$\theta_2 = 1.1470$	$\theta_6 = 2.50385$
$\theta_3 = 1.0739$	θ_7 was set = 0
$\theta_4 = 195.00$	

The calculated and observed swelling data for 14 NAE's of the most prototypic irradiation test (1250 MWD/T average exposure) are tabulated below for comparison.

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<u>Element</u>	<u>Component</u>	<u>Calc. %ΔV By Model</u>	<u>Measured %ΔV</u>
Upstream	Inner	0.12	0.03 (±0.15)
	"	0.27	0.40
	"	0.42	0.54
	"	0.76	0.60
	"	1.13	0.54
to	"	1.29	0.55
	"	1.19	0.68
	"	1.21	0.77
	"	1.22	0.86
	"	1.02	0.63
	"	0.45	0.55
	"	0.27	0.43
	"	0.21	0.29
		0.15	0.09
Downstream			
Upstream	Outer	0.15	0.38 (±0.10)
	"	0.36	0.54
	"	0.51	0.62
	"	0.63	0.69
	"	0.66	0.72
	"	0.66	0.90
	"	0.60	0.92
to	"	0.60	0.77
	"	0.60	1.08
	"	0.54	0.93
	"	0.45	0.58
	"	0.33	0.50
	"	0.27	0.45
	"	0.18	0.47
Downstream			

Shaped Single Tube Element

The fabrication of test elements for an evaluation of the fluted single-tube concept for N-reactor fuel was completed. The elements were formed by coextrusion of 0.95 percent enriched uranium with zircaloy-2 cladding. The nominal OD of the elements was 2.327 inches; the ID was 1.286. The outer surface shape was composed of circular arcs forming flutes 0.1 inch in height. The end closures were brazements using zircaloy-2 caps about .5 inch in thickness. The cap-uranium joint was mitered into a V-groove in the uranium to provide a transition in the amount of swelling and consequent cladding strain adjacent to the cap. Two elements

10-1/2 inches in length were shipped to the ETR for irradiation in the M-3 high pressure loop facility.

Dual Enriched Single Tube Element

An experimental dual-enriched single-tube N-reactor element failed during its fifth cycle of reactor operation in the ETR-M3 loop. The failure resulted from a cladding hot spot which developed in an area where warp of the element had restricted coolant flow. The element had received a maximum exposure of about 900 MWD/T.

Irradiation of Zircaloy-Clad Fuel Elements with Brazed Closures

Post-irradiation examination is complete on the variable braze-thickness irradiation test, GEH-4-68, 69, and 70. The tests consisted of three Zr-clad uranium tubes with end closures brazed with the Zr - 5 w/o Be brazing alloy. Two of the tubes had braze layers 0.030 inch thick between the cap and the core and the other tube had braze layers 0.015 inch thick. The test completed six cycles of irradiation at the MTR in low pressure cold water for a total exposure of approximately 1360 MWD/T. Observations from the radiometallurgical examinations may be summarized as follows:

- 1) The external condition of all elements was good.
- 2) None of the end-closures, regardless of braze width, showed cracking in the braze. All closures were in good condition.
- 3) Extensive cracking was found in the uranium in the braze heat affected zones (within 1 inch from the braze layer).
- 4) In at least one end, cracking appeared to originate in a cluster of voids in the uranium. These voids may have been introduced by the brazing process prior to irradiation or may have been caused by grain boundary tearing during irradiation.

Fuel Charging Studies

N-Outer Fuel Element Support

N-outer fuel element support - process tube compatibility testing was scaled up to charge a full length process tube. Previous testing in the short (5 foot) process tube sections did not reveal the scratch precipitating mechanism which

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severely scratched the process tube in the 314 full scale charging mock-up. A full length process tube charging test was designed to eliminate as many possible sources of scratching as practical. This test was performed under laboratory conditions, using building process water which is filtered through a 50 micron filter. System cleanliness was considered to be of paramount importance, and is more difficult to maintain as process tube lengths increase. Special cleaning and drying equipment as well as charging facilities were built or adapted for use with the long process tubes.

A 17-foot long section of process tubing was used to shake-down the facility and equipment for longer tube tests. Seven (7) elements, 5 under static water conditions, and 2 under a 30 gpm water flow rate, were charged through this tube with no evidence of damage.

A 52-foot, 8 inch long process tube was cleaned in the same manner as the 17-foot tube (1 hour cold water and detergent soak, flush rinse at 30 gpm, high velocity water jet impinged on tube wall full length), and set up to charge at a flow rate of 30 gpm. The first fuel element scratched the tube on one set of supports. A single scratch started approximately 33 feet 8 inches from the charge end of the tube. At about 35 feet the scratch divided into two scratches. At this point the second support apparently crossed the original scratch. The two scratches continued in a narrow criss-cross pattern to the end of the tube. The scratches are about .0002 to .0006 inches deep, and .002 to .004 inches wide. The other set of supports passed directly over a projection on the tube wall (visible in the borescope) which severely gouged the supports. The projection remained in place on the tube wall and no scratching was initiated by the damaged supports.

The tube was rotated approximately 90° to expose unused surface to the supports, recleaned with the jet, and testing resumed. Seventeen (17) fuel elements were charged through the tube under both flowing and static water conditions (elements 2 through 18). Borescopic examination did not reveal any tube damage.

The process tube was recleaned with the jet and rotated. Element 21 induced a very light scratch in the last 5 feet of the tube. Element 22 induced a scratch in a butt weld joint 18 feet from the charge end of the tube. Cleanliness was gradually relaxed as the tests were progressing without incident of scratching. This factor may account for the scratching produced in the last two tests. Testing was suspended at this point.

Detailed examination of supports and process tube surfaces indicates that the scratch precipitating mechanism is particulate in nature. A minute hard particle (sand, oxide flake, weld slag, metal chip, etc.) imbedded in the relatively soft iron support surface would produce the behavior witnessed in these tests.

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