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REPORT TO THE WORKING COMMITTEE
OF THE FUEL ELEMENT DEVELOPMENT COMMITTEE
FROM THE GENERAL ELECTRIC COMPANY - HANFORD

11

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OF THE FUEL ELEMENT DEVELOPMENT COMMITTEE
FROM THE GENERAL ELECTRIC COMPANY - HANFORD

April 7, 1964

by:

M. Lewis, N-RD
J. E. Minor, HL
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CURRENT REACTOR PRODUCTION FUELS

Uranium Core Production Data

Quality of Uranium Cores

Visual Inspection of Virgin Cores

Visual reject rates for the period December 1, 1963 through March 27, 1964 were 0.09 per cent for normal uranium and 0.12 per cent for enriched uranium. This is no change from the last reporting period.

UT-2 Rejects

The UT-2 reject rates for the period December 1, 1963 through March 27, 1964 were 0.5 per cent for normal uranium and 1.1 per cent for enriched uranium cores. The improved standards for K5N and K5E cores were placed in service during this period and significantly reduced the rejects in these models. Fifty per cent of the normal rejects were K5N cores tested prior to the use of the new standards. The reject rate for enriched uranium remained high principally because of the within-core grain size variation reported in the January 14-16, 1964 Working Committee FEDC Report.

The grain-size variation observed in HAPO enriched cores was apparently caused by non-uniform quenching resulting from the condition of the heat treat rack in the old Nu-Sal furnace where the enriched cores were being heat treated. HAPO specifications required a 43-second air delay time (salt bath to quench) on normal cores and a 20-second delay time on enriched cores. The specifications were revised to provide a 20-second delay time on both normal and enriched cores, thus enabling National Lead to heat treat both normal and enriched cores in the newer more uniform Amsler-Morton furnace. This change should result in substantially improved enriched cores.

Grain Size

The average grain size of cores received during the period November 1, 1963 through March 1, 1964 was A-6 for both normal and enriched uranium.

Uranium Specifications

On November 1, 1963, Savannah River increased their iron and silicon specifications to 125-225 ppm on Mark VB inner and outer ingots. HAPO has been urged to review the specifications for I&E ingots to determine if the limits could be raised from the current maximums of 140 ppm silicon and 185 ppm iron to the new SRP levels, thus eliminating the need for separate solid-scrap remelt streams at the feed site. Currently the specifications for I&E cores are being revised and, tentatively, will specify 115-200 ppm iron and 75-140 ppm silicon. For several reasons HAPO does not believe the specification on additives in I&E cores should be further raised to the SRP levels: (1) HAPO uses additives primarily to eliminate bumping, for which the current levels

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are satisfactory, while SRP increased the additive levels to eliminate cavitation-type swelling, (2) HAP0 has no experience irradiating cores with iron and silicon in the 125-225 ppm range, and (3) NLO has not shown the problem of maintaining two scrap streams to be a problem or to involve increased costs. It should be noted that increased additive levels up to 300 ppm iron and 350 ppm silicon would not be a problem in chemical separations at HAP0.

Top Crop Remelting

HAP0 has approved the use of normal ingot top crops which have been remelted and processed to rod as recycle in I&E ingots. This type of feed is not to exceed ten per cent of the total charge weight in any ingot. Work performed at NLO showed the use of remelted top crops does not adversely effect the quality of the ingots.

UT-2 Testing

HAP0 is evaluating a sample plan for UT-2 testing at HAP0 to replace the current 100 per cent testing. Such a plan is feasible so long as the heat treat process is in control and the feed site is also UT-2 testing on a sample basis. The primary concern is that no partially or completely untransformed cores are charged into the reactors. In January HAP0 started retesting every reject core to determine the cause of rejection, and to date no untransformed cores have been found.

Cut-Crown Magnesium

Development work is being conducted at the feed site to use a cheaper form of magnesium in the uranium reductions. This is referred to as cut-crown magnesium and differs from that now being used in that it has not been remelted before being processed to chips. Apparently, the only significant difference in the uranium reduced with cut-crown magnesium is an added aluminum contamination of approximately 50 ppm. HAP0 has no objection to the use of cut-crown magnesium provided Al is the only contaminant introduced and it remains at a uniform low level.

Induction-Oil and Salt-Oil Heat Treating

A production test to evaluate the performance of induction heat treated O3N cores was discharged in March and the data will be available for analysis in April. One induction heat treated piece failed in this test (side hot spot type), but the significance of this failure will not be established until the dimensional data are analyzed. Bulk charging of approximately 65 tons of induction heat treated O3N cores will commence in April. Performance of these charges will be monitored through the quality certification program.

A production test to evaluate salt-oil heat treated enriched cores will be discharged in April.

At the present time, over-all fuel performance is not limiting the operation of the reactors, and consequently economic justification for converting to

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salt-oil heat treatment of HAP0 I&E cores is difficult to show. However, tests in progress on long fuel elements and high performance drivers for target loadings may show that oil quenching is required for these programs. If these programs justify the conversion to oil quenching, then induction heating may also be considered if it is shown to provide a product superior or equivalent to salt heated metal.

Heat Treat Variables

In April 1962 an order was placed for cores from twelve ingots and twelve dingot bars heat treated under various conditions in a statistically designed experiment. The cores were to be irradiated and the critical heat treat parameters defined and optimized. During heat treatment the cores warped excessively and could not be machined to produce acceptable pieces. Therefore, this material will not be irradiated. However, it was believed an analysis of heat treat induced warp would be valuable as this may be related to reactor warp and has a practical significance in the NLO plant. Also, x-ray studies are being conducted at HAP0 on selected treatments. The variables investigated were (1) core blank size, (2) form-hollow or solid, (3) quench medium-brine or oil, (4) delay time, and (5) quench temperature. The individual treatments are:

<u>Treatment No.</u>	<u>Core Blank Size</u>	<u>Form</u>	<u>Medium</u>	<u>Delay Time</u>	<u>Quench Temp.</u>
1	1.454	S	B	30 sec.	120
2	1.454	H	B	30	120
3	1.410	S	B	30	120
4	1.410	H	B	30	120
5	1.437	H	O	14	200
6	1.437	H	O	14	90
7	1.437	H	O	6	200
8	1.437	H	O	6	90
9	1.437	H	O	10	145
10	1.454	H	O	10	200
11	1.454	H	O	10	90
12	1.410	H	O	10	200
13	1.410	H	O	10	90
14	1.437	S	B	43	120
15	1.437	S	B	17	120
16	1.437	H	B	43	120
17	1.437	H	B	17	120
18	1.437	H	O	10	145
19	1.454	H	O	14	145
20	1.454	H	O	6	145
21	1.410	H	O	14	145
22	1.410	H	O	6	145
23	1.437	S	B	30	170
24	1.437	S	B	30	70
25	1.437	H	B	30	170
26	1.437	H	B	30	70
27	1.437	H	O	10	145

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In addition to the above variables, two types of dingot (1) outgassed after beta heat treatment, (2) non-outgassed, and two types of ingot (1) 250 ppm Fe plus Si, (2) 150 ppm Fe plus Si, are compared.

Brine Quench

For brine quenched material the data are quite consistent from dingot to ingot and indicate that the lower warps are found for:

1. larger core blank size
2. hollow form
3. lower quench temperature

The differences between types and between ingot and dingot are not significant... Analysis also indicates that in the hollow form the lower warps are found at the smaller core blank size and the lower quench temperature.

Oil Quench

A similar analysis has been performed on the warp data from the oil quenched cores. Solid oil quenching was not included in this test, so there is one less variable in this analysis than in the water quenched analysis.

The results indicate lower warps are to be expected at:

1. smaller core blank size
2. longer delay time
3. lower quench temperature

Analysis also indicates that the effect of the core blank size is not strong for the long delay time, but at the short delay time the effect of core blank size and quench temperature is very strong.

Depleted Uranium

HAP0 is currently involved in a program to produce 25-30 kg of 18 per cent ²⁴⁰-content plutonium for subsequent irradiation to curium-²⁴⁴. This program requires the irradiation of at least 22 tons of depleted uranium, ten tons in the old reactors and 12 tons in the K Reactors. The goal exposures will be to 1200 MWD/T in the old and 1400 MWD/T in the K Reactors. These high exposures and long residence times require exceptionally good and uniform fuel performance. The specifications for depleted cores are essentially the same as for standard metal, but some impurity limits were lowered and ranges were imposed on carbon (350-650 ppm), iron (100-180 ppm) and silicon (60-125 ppm) in order to obtain uniform quality. Also, a surface hydrogen specification was imposed in an attempt to prevent excessive braze-layer porosity in the canned elements, and the core blanks were salt-oil heat treated.

The feed material was derby uranium in inventory at Paducah, Kentucky. These derbies were exceptionally free of slag and met all of the chemical requirements. In the interest of uniformity, the ingots were to be all

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derby charges with no recycle scrap. However, experience with the ingots cast for the "O" size cores showed difficulty meeting the minimum carbon requirement with all derby charges, and NLO was allowed to recycle reject ingots and cores from the "O" size campaign into "K" size ingots, provided a uniform amount of scrap was added to each ingot. The grain size of the depleted cores appears to be a uniform A-4. The "O" size cores have been shipped and will be charged in May. Shipment of the "K" size cores is expected by the end of April.

Fuel Performance

Nine normal and fourteen enriched production fuel elements failed during the period December 1, 1963 through March 31, 1964. In addition, two failures occurred in C6N overbore material, two failures occurred in dingot metal, and one failure occurred in induction heat treated material in a production test.

Of the fourteen enriched production failures, thirteen occurred in non-bumpered fuel. The use of bumpered fuel was discontinued in the January E-N loading in H Reactor, and in the fringe loadings in the old reactors at the end of CY 1963. Based upon recent experience, plans now call for the production of approximately 20 tons per month of bumpered fuel starting in April for the fringes of the old reactors, and the October E-N loading will be of bumpered fuel.

Sufficient data to project the dimensional stability and bond quality trends was not available during this period.

AlSi Process Development

Increased Fuel Length Program

Studies at Hanford have shown an economic incentive exists for increasing the fuel element lengths to ten and/or twelve inches. The major savings are in labor and materials costs in fuel element assembly. A series of tests is planned to evaluate the use of longer fuel elements in the old and K Reactors. An outline of the testing schedule is:

1. Ten-inch "O" size enriched water quenched cores with six-inch control pieces to be reactor charged in April. Approximately 300 elements.
2. Ten-inch "O" size enriched oil quenched cores with six-inch control pieces to be reactor charged in May. Approximately 240 elements.
3. Twelve-inch "O" size normal oil and water quenched cores with eight-inch control pieces to be reactor charged in June. Approximately 600 elements.
4. Pilot scale evaluation of a minimum of 2500 ten-inch "O" size enriched cores to be reactor charged in July.

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TABLE I
PRODUCTION RUPTURES
I&E FUEL ELEMENTS

Fuel Element Type	Reactor	Power	Exposure		Rupture Classification	Failure Date
			MWD/T	% Goal		
Enriched Non-Bumpered	H	1147	730	91	Side Hot Spot	12/4
Enriched Non-Bumpered	F	1281	436	68	Side Unclassified	12/21
Normal Watermix	H	1154	863	108	Side Groove Corrosion	12/24
Normal Non-Bumpered	DR	1223	307	52	Side Hot Spot	12/26
Normal Non-Bumpered	B	1204	459	77	Unknown-Not Examined	12/27
Overbore Bumpered	C	1428	227	28	End Defect	12/30
Normal Non-Bumpered	DR	1278	405	77	Side Hot Spot	1/6
Normal Non-Bumpered	DR	1242	394	65	Side Hot Spot	1/10
Enriched Non-Bumpered	F	1226	969	127	Unknown-Not Examined	1/14
Overbore Bumpered	C	1475	315	39	End Defect	1/18
Normal (Dingot) Non-Bumpered	H	1302	362	69	Side Hot Spot	1/21
Enriched Non-Bumpered	C	1394	538	81	Side Hot Spot	1/24
Normal Non-Bumpered	DR	1322	616	116	Unknown-Not Examined	1/31
Enriched Non-Bumpered	F	1283	535	83	Side Hot Spot	2/7
Enriched Non-Bumpered	C	1340	680	86	Side Hot Spot	2/9

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TABLE I (Cont'd)

<u>Fuel Element Type</u>	<u>Reactor</u>	<u>Power</u>	<u>Exposure</u>		<u>Rupture Classification</u>	<u>Failure Date</u>
			<u>MWD/T</u>	<u>% Goal</u>		
Normal Non-Bumpered	D	1234	694	114	Side Hot Spot	2/20
Enriched Non-Bumpered	H	1270	351	43	Side Hot Spot	2/22
Enriched Non-Bumpered	H	1320	343	42	Side Hot Spot	2/23
Enriched Non-Bumpered	F	1277	741	99	Side Hot Spot	2/24
Enriched Non-Bumpered	H	1290	425	53	Side Hot Spot	3/3
Enriched Non-Bumpered	F	1388	847	120	Unknown-Not Examined	3/4
Enriched Non-Bumpered	C	1547	499	76	Side Hot Spot	3/5
Normal (PT-610) Induction Heat Treat	DR	1232	646	122	Side Hot Spot	3/9
Normal (Dingot) Non-Bumpered	B	1377	445	84	Side Hot Spot	3/11
Enriched Bumpered	DR	1278	1044	141	Side Unclassified	3/11
Enriched Non-Bumpered	H	1296	547	68	Side Hot Spot	3/14

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5. Pilot scale evaluation of 1000 ten-inch "K" size enriched cores to be reactor charged in August.

The ten-inch enriched water quenched elements have been fabricated, measured, and are ready for reactor charging.

An alternate method for producing long elements is to assemble two short cores in one jacket. This approach would alleviate problems at the feed site in drilling the longer cores. No serious process assembly problems were encountered fabricating 50 double-core elements for reactor drop tests. These tests are designed to determine if the core-to-core bond can withstand the forces on the bond during discharge into the reactor basin. If the unirradiated double-core elements do not break apart, consideration will then be given to irradiation testing.

Depleted Uranium Program

The dingot uranium program showed that the chemical and physical properties of the uranium core have a direct effect on the finished fuel element bond structure. A pilot quantity of the small reactor size (100 O3D) depleted cores have been canned in the pilot plant under the following conditions:

1. Standard conditions
2. Standard conditions with a 1-2 second agitation in the AlSi layer of the duplex furnace
3. Lead temperature 8 C higher than standard
4. Lead temperature 8 C higher than standard and a 1-2 second agitation in the AlSi layer of the duplex furnace.

Metallographic examination of the bond layers indicates Condition 2 is the best, and this is the condition that will be used for canning the O3D cores. A similar test is planned for determining the optimum conditions for canning the "K" size (KID) depleted cores.

Upstream Fuel Program

The upstream fuel program consists of the use of certain low-risk sub-standard fuel elements in low-rupture potential upstream positions in the reactors. The program has been in effect since August 30, 1963, and approximately 33,000 fuel elements which would formerly have been rejected have been charged into upstream positions.

Overbore Fuel Program

Two fuel element failures in each of the last two C6N overbore fuel lots classified these lots as rupture prone. Six per cent of the irradiated elements, including those that ruptured, exhibited a "necking down" or tensile stress on the aluminum cladding at the junction of the cores and

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the end caps. Radiometallurgical examination of the irradiated elements showed little or no bond existed between the uranium and the braze metal on the end caps. A large unirradiated sample of the last C6N rupture-prone lot was ultrasonically end cap bond tested, and approximately six per cent of the elements had some detectable unbonded areas. Metallographic examination of these elements showed the ingot cores had larger than usual stringers, which in most cases were filled with lead. Also, the compound layers were too thick and had shattered. Figure 1 shows the unbonded area on an irradiated element and the stringers and bonds in an unirradiated element.

Pilot plant development tests have been performed to determine the effect, if any, of lead filled stringers on bond formation. Evaluation of the results is not complete, but changes in the canning process parameters are expected to lessen the problem.

Projection Fuel Program

Process and mechanical improvements continue to be made in the ultrasonic welding of aluminum supports to aluminum clad fuel elements. During the period April 1963 through February 1964, the rejection of projection fuels for faulty rail attachment has decreased from seventeen to two per cent. Currently, 99.8 per cent of the welds produced are satisfactory.

The significant process changes leading to the improved yields are the use of oil rather than soap as a lubricant for the welding operation and the elimination of the deoxidizing of rails prior to welding. The rails were formerly degreased and deoxidized in a commercial preparation prior to welding. Elimination of the deoxidizing step has increased the average tensile peel strength of the welds from a nominal 80 pounds to 100 pounds. The mechanical improvements in the welder basically consist of providing greater rigidity in the welder and in improving the post welding rail sizing equipment. The current collapsible rail design requires sizing after welding to maintain the height specification required by the reactors. Improved sizing equipment has reduced the standard deviation of the rail height from 0.9 mil to 0.5 mil, with attendant decrease in rejection rate for rail height.

Further improvements are anticipated from development work now in progress. A large scale test recently conducted using increased weld power and automatic rail feeding and lubricating equipment indicates the rejection rate may be reduced to one per cent and the capacity of the welders may be increased from 180 elements per hour to 240 elements per hour. In this test the lubricant was applied to the weldment side of the rail only. This keeps oil off the sonotrode tip and results in more uniform weld strengths at a shorter weld time (0.5 seconds rather than 0.8-1.0 seconds). Also, the use of fatty acids derived from soap-water solutions as lubricants shows promise for further improving the welding operation.

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A. Irradiated C6N
Element 5X Cap End
Lot CP005D

B. Unirradiated C6N End Bond 250X
FIGURE 1

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N-RD PRODUCTION FUELS

Shop Yields

During the first quarter of CY 1964, 110.2 tons of fuels were produced. Uranium utilization in the period was 72.6 per cent.

Major shop problems for the past quarter were clad-to-core unbonding and inner clad variation of the outer tube. The unbond problem was severe for N-outer tubes during January (4.4 per cent) but returned to normal levels in the next two months. The increased rate for inner cladding defects was the result of the dimple problem described elsewhere in this report.

The cumulative rates of major reject categories for the first three months of 1964 are shown below:

<u>Station</u>	<u>Reject Rate - Per Cent</u>	
	<u>Outer Fuel</u>	<u>Inner Fuel</u>
Bonding (Peel Test)	.3	3.4
Clad Thickness Variation	11.9	0.0
End Closure	5.0	3.2
Dimple Defects	7.3	0.0
Thin Cladding	4.3	1.5

Billet Fabrication Problem

A large increase in defective rates for billets at the feed site resulted from casting problems which were discussed by National Lead in the last report. The billets produced during the period were characterized by a higher incidence of surface pitting and lower carbon levels.

A study was made at Hanford to determine the fabrication characteristics of the within-specification billets produced during the out-of-control period. Density, hardness, and ultrasonic testing failed to show any significant differences between these billets and a number of billets from earlier casting periods.

Finished fuel elements from this material were examined; mechanical properties, chemical-etch rates, and metallographically-observed stringer content all failed to show difference from control material. Over a hundred samples, which represented material throughout the history of N-fuels, were evaluated for non-metallic stringer content and, though variation was found in the population, there was no significant difference evident between the test material and the control.

Large Diameter Ingot Study

The first four hollow cast ingots for tipped mandrel extrusions have been processed through the finished billet stage. These ingots had a nominal 13 inch OD and 6.5 inch ID and were primary extruded to the nominal 7.2 inch OD by 2.6 inch ID N-outer billet size. The purpose of this ingot design is to increase the amount of working of the uranium near the ID and reduce the inner cladding thickness variation due to residual ingot effects. Metallography, completed

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at National Lead, has shown the new geometry to have superior heat treat characteristics resulting in (1) greater grain refinement and (2) following primary extrusion, the crystallographic texture of the billet ID closely approximates that normally found at the OD.

The ingots were primary extruded at Ashtabula in mid-February and were characterized by higher running pressures, greater full length bow, and longer end defects. The running pressures were approximately 40 per cent higher than the standard 11 inch ingots while the reduction ratios were only about 10 per cent greater. This pressure increase is in part reflected in greater work on the ingot ID with subsequent improved grain structure.

The full-length bow increased from an average of .050 inch to .175 inch. Although the extrusions were made at a time of generally higher than normal extrusion warp, no satisfactory cause has been assigned to the condition. The full-length bow problem resulted in a 60 per cent reject rate for finished billets for wall thickness variation.

The increased length of end defects did not result in any short billet losses. The increased defect was expected as all of the ingots were square ended. Work is continuing with the casting of 16 more ingots during which time ingot pre-shapes will be studied to minimize end defects and attempts will be made to determine the causes of increased warp.

Sleeved Billet Concept

A potential approach to reduced cladding thickness variation on the ID of outer fuels is the fabrication of sleeved billets wherein greater working can be concentrated on the metal surrounding the inner zircaloy component.

Two sleeved billets have been assembled for single-tube fuel element designs. The billet assemblies were made up of standard NOE billets with uranium sleeves of 2.780 inch OD X 2.380 inch ID. The sleeves were made with a reduction ratio of 14:1 and the coextrusions with reduction ratios of 16.2:1. The outer cladding thickness variations were normal, but the inner cladding thickness variations resulting from these coextrusions were considerably less than the 0.008 inch normally encountered with standard N-outer tube coextrusions. The results for the 18 pieces produced were as follows:

	<u>Maximum Cladding Variation</u>	
	<u>Average</u>	<u>Range</u>
OD Cladding	.003"	.002" - .005"
ID Cladding	.004"	.002" - .009"

Samples were taken from each extrusion and examined for uranium-to-uranium bonding. In all samples, bonding was complete and recrystallization extended across the interface.

Further work is in progress to adapt this concept to standard NOE coextrusions.

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Outer-Tube, Inner-Clad Dimpling Defects

A severe increase in clad thickness variation rejects was experienced during the period. The increase was a result of the phenomenon known as dimpling, which is seen as a streak of thin zircaloy clad caused by a drag of metal during extrusion followed by periodic deposits of the dragged metal forming abrupt thick spots. The defects appear to be time related and on the worst days a reject rate of 50 per cent for this cause was experienced. By late January the reject rate averaged eight to ten per cent.

A study of billet component histories and coextrusion variables showed that more dimples occurred in the center section of a coextrusion, that dimpling reject rates varied with respect to coextrusion order and time, and that there was no relationship to component histories. No significant differences were found in composition of copper or zircaloy in dimpled vs non-dimpled areas nor were there differences in microhardness in the copper, zircaloy, or uranium in these areas.

A correlation was found between dimpling and different batches of extrusion lubricants. The lubricants were colloidal graphite and differences were noted between batches from the same vendor as well as within batches. It has been postulated that these lubricants can degenerate from causes such as freezing or contamination. New batches of lubricants are being used in production and reject rates for dimples are now less than one per cent.

REACTOR FUEL DEVELOPMENT

N-Reactor Fuel Development

Process Development

Alternate Uranium Composition

Studies are in progress to determine the effect of altered fuel components upon fuel element fabrication, corrosion behavior, and irradiation-swelling resistance.

The swelling resistance of the fuel elements fabricated from the standard fuel composition is to be compared to an alloy containing 400 ppm Fe and 800 ppm Al at the high fuel core temperatures attained with a single-tube geometry (1.790 inch OD X 0.975 inch ID) enriched in U-235. The major impurities in these two fuel core materials are: 510 ppm C, 137 Fe, 6 Al, 127 Si for the standard composition and 537 ppm C, 386 Fe, 837 Al, 47 Si for the altered fuel. Both compositions are being irradiated in the as-beta heat treated condition.

Structural characterization of the ingots, primary extrusions, coextruded fuel and completed fuel elements was made. The increased iron and aluminum content of the altered fuel composition caused an increase in recrystallization temperature, a reduction in as-extruded grain size, an increase in hot hardness at 300-400 C, and an increase in the number of second-phase compound inclusions. No significant

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differences were observed in either the beta heat treated grain size or the clad-to-core bond appearance.

Work has started on the preparation of 14 different uranium alloys for NaK-capsule irradiations. The relative swelling resistance is to be compared as a function of composition, heat treatment, and irradiation conditions.

The composition of eight of these alloys, which have been received from the Mallinckrodt Chemical Works, are given in Table II. These alloys, produced as dingots from the bomb reduction process were fabricated by gamma extrusion of the dingots into two-inch diameter rods. To provide small coextrusion billets, the rods were cut into six inch lengths. Prior to final machining, the billet blanks of the U + Si, U + Al + Si, U + Al, and U + P alloys were beta heat treated and water quenched. Cracking occurred along the radially-oriented grain structures in the first three alloys; the U + P alloy did not crack. Quenching into salt or air cooling to lower the cooling rate still resulted in cracking of the billets. The cracked billets and original stock of the U + Si + Al and the U + Al alloys will be remelted and cast in an effort to produce sound coextrusion billets.

The beta heat treated U + P and the remaining alloys with Zr or Nb additions and three sound U + Si billets, all in the as-gamma extruded structure, were coextruded with Zr-2 cladding to 0.6 inch diameter rods. Preheat temperatures and extrusion constants for each rod are given in Table III. Cladding thickness variation was good on all the rods except the U + P, U + Si, and the U + Nb + Al alloys. From previous experience with the irradiation instability of non-uniform Zr-2 cladding, these two alloys will be decladded before they are irradiated.

The coextrusion of the U + Nb + Al alloys was difficult. Using coextrusion preheat temperatures in the high alpha range, the finished rod had a longitudinal cladding thickness variation ranging from 0.020 to 0.040 inch. The variation is believed to be related to the greater "stiffness" of the U + Nb + Al alloy compared to the Zr-2 in the alpha-phase temperature of the uranium. Attempts to alpha coextrude the alloy using a "stiffer" alloy of Zr + 3Nb + 1Sn for the cladding were unsuccessful. The problem was solved by coextruding with Zr-2 cladding in the low gamma-uranium temperature range (750 C). Extrusion variables and preheat temperature for these tests are listed in Table IV.

Seven other alloys of uranium with varying amounts of Fe, Si, Al, and carbon are now being produced using either vacuum induction or arc melting facilities at Hanford.

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TABLE II

COMPOSITION OF URANIUM ALLOYS FOR CAPSULE IRRADIATION PROGRAM

<u>Alloy</u>	<u>Si</u> (ppm)	<u>Al</u> (ppm)	<u>Zr</u> (w/o)	<u>Nb</u> (w/o)	<u>C</u> (ppm)	<u>P</u> (ppm)
1	---	700	1.75	---	25	---
2	---	---	1.55	---	17	---
3	---	380	----	1.7	35	---
4	---	800	----	---	55	---
5	800	---	----	---	37	---
6	300	260	----	---	38	---
7	---	---	----	---	40	450
8	---	---	2.3	---	18	---

TABLE III

COEXTRUSION CONSTANTS AND PREHEAT TEMPERATURES FOR URANIUM ALLOY RODS

<u>Alloy</u>	<u>Rod No.</u>	<u>"K" Start</u> (TSI)	<u>"K" Run</u> (TSI)	<u>Preheat</u> Temp. -°C	<u>Remarks</u>
U + Zr + Al	1	24.2	21.7	630	Coextrusion good
	2	22.4	20.1	630	
U + Zr	1	23.8	20.5	630	Coextrusion good
	2	24.2	21.0	630	
U + Nb + Al	1	>30.0	Stalled	610	Non-uniform longitudinal cladding thickness ***
	2	22.4	18.7	655	
	3	23.8	22.0	610	
	1*	23.8	21.1	630	
U + Si	1	24.8	22.2	610	Non-uniform cladding thickness **
	2	24.8	21.7	610	
	3	24.3	22.0	610	
U + P	1	19.6	15.7	630	Non-uniform cladding thickness **
	2	17.8	14.5	630	
	3	18.7	15.4	630	
U + Zr	1	24.3	20.1	630	Coextrusion good
	2	24.3	20.5	630	

*Stalled extrusion billet #1 was recanned and coextruded.

**To be decladded before irradiation.

***See Table IV for the coextrusion performance of this alloy at a preheat temperature of 750 C.

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TABLE IV
COEXTRUSION CONSTANTS AND PREHEAT TEMPERATURES FOR U + Nb + Al ALLOY

<u>Alloy Rod</u>	<u>"K" Start</u>	<u>"K" Run</u>	<u>Preheat Temp. °C</u>	<u>Remarks</u>
1	30.0	Stalled	610	Non-uniform longitudinal cladding thickness.
2	22.4	18.7	655	" "
3	23.8	22.0	610	" "
1*	23.8	21.1	630	" "
4**	22.9	21.5	650	" "
5	11.7	11.7	750	Coextrusion good, uniform cladding thickness

*Stalled billet #1 recanned and extruded.
**Clad with Zr + 3Nb + 1 Sn alloy.

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End Closure Development

Corrosion Studies of Braze Materials

Corrosion studies are being continued on zirconium-base alloys containing varying amounts of beryllium and uranium in 360 C water at pH 7 and pH 10. Several alloy systems have been added to the study: zirconium-beryllium with iron, copper and thorium, zirconium-iron-nickel, and zirconium-iron-chromium and nickel. This work is aimed at defining the relation of braze metal corrosion rates vs composition for the current braze metal and other braze metal compositions.

Because a large mass of data has been accumulated, a Fortran program was written to analyze the results. This program, called CORAN (for corrosion analysis), calculates the surface area, weight differences, weight gain/loss per unit area, and normalizes the light data.⁽¹⁾ Normalized light data plotted as a function of time yields a curve very similar to the weight gain curve up to the point of breakaway corrosion. These light data are analogous to the optical properties of pure metals and represent the approach to the stoichiometric condition. The basic program is being enlarged to incorporate a regression analysis of the optical properties as a function of weight for each measured wave length. This analysis will show any deviation from ideal and should indicate those alloys which have either improved or poorer corrosion properties.

To date the zirconium alloys with varying beryllium content have amassed 3400 hours in 360 C pH 7 water. After 2900 hours the 1.13 per cent Be alloy began to rapidly lose weight the same as in the pH 10 test after 750 hours. Zircaloy with 0.5 per cent uranium had the same weight gain after 3500 hours as zircaloy-2 after 3400 hours. Additions of iron to a Zr-2 + 4.5 per cent Be alloy shows decrease in the weight gain with increasing iron content, up to 12 per cent Fe. The Ni-Cr-Zr-2 alloys had half the weight gain of Zr-Cr-Fe and Zr-Cr-Fe-Ni alloys. Copper additions up to 14 per cent Cu resulted in large weight gains for the Zr-2 + 4-1/2 per cent Be alloy. Thorium additions of only 120 ppm resulted in large weight gains for the basic Zr-2-4-1/2 per cent Be alloy.

These tests will continue during the next quarter and all available data will be analyzed by means of the CORAN program.

Fuel Element Support Fabrication Development

Hot Forging of Zircaloy-2 Wire into Complex Shapes

Development of stronger and less expensive outer fuel tube supports, fabricated by means of hot forging techniques, is being conducted.

(1) The measure of an optical property of the specimen surface which is analogous to absorption and/or reflectance.

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The outer fuel tube support (Figure 2) is hot forged directly from 1/8 inch diameter zircaloy-2 wire to finished dimensions. The increased depth of section (Tee shape) in the crown area markedly increases the support strength and permanent set characteristics.

Supports formed thus far have been forged on a 600 KVA resistance welder because of easily and accurately controlled heating conditions coupled with up to 18 tons forming force. The 1/8 inch diameter zircaloy-2 wire is resistance heated into the beta phase (950 C estimated from color) and formed by a 15 ton blow. Total heating time is 1.5 seconds followed immediately by the forming blow. The part experiences an extremely rapid quench in the die. Approximately two seconds elapse after the die closes till the part is cool enough to be held by hand. For surface finishing the part is given a light sand blast and bright etch.

Approximately 300 pieces have been autoclaved in 400 C steam for 72 hours with no evidence of abnormal corrosion behavior. Ten pieces are in an extended autoclave run in 360 C water, and had accumulated 504 hours at the last examination. No evidence of abnormal corrosion behavior was seen.

Irradiation Experience

KER Loop Irradiations

Additional swelling and operational data have been obtained from two KER charges of N-fuel element assemblies. The charges were irradiated to tube average exposures of approximately 1600 and 2000 MWD/T at prototypic fuel-core temperatures and 1600 psi coolant pressure. Maximum fuel swelling observed in the 1600 MWD/T exposure test was 0.85 v/o in an inner tube (@2000 MWD/T) and 2.50 v/o in the mating outer tube (@2500 MWD/T). Maximum fuel swelling in the 2000 exposure test was 1.33 v/o in an inner tube (@2200 MWD/T) and 3.64 v/o in an adjacent outer component (@3100 MWD/T). These new data, along with previously obtained N-fuel element component and single tube element data (KSE-3's and 5's) have permitted the continuing development of an expression for fuel swelling. Analysis of the data from 130 elements has resulted in three similar expressions, one for each fuel geometry. Agreement between the calculated and experimentally determined fuel swelling is shown in Figure 3 in which perfect agreement is indicated by the dashed line. The expressions are:

$$\text{for NOE, } \% \Delta V \text{ in fuel} = B \left[4.15 + 550 \left(\frac{T_r}{P_r} \right) \left(\frac{1}{1 + e^{-\frac{T + 100B - 420}{20}}} \right) \right]$$

$$\text{for NIE, } \% \Delta V \text{ in fuel} = B \left[3.05 + 550 \left(\frac{T_r}{P_r} \right) \left(\frac{1}{1 + e^{-\frac{T + 100B - 480}{20}}} \right) \right]$$

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FIGURE 2 - N.O.T. II FUEL SUPPORT
APPROX. 3X SCALE

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FIGURE 3 COMPARISON OF MEASURED AND
CALCULATED FUEL SWELLING

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and for KSE 3 and 5,

$$\% \Delta V \text{ in fuel} = B \left[3.95 + 550 \left(\frac{Tr}{Pr} \right) \left(\frac{1}{1 + e^{\frac{T + 100B - 460}{20}}} \right) \right]$$

where, B = a/o burnup, (1 a/o BU $\approx$ 8650 MWD/T)

$$Tr = \frac{\text{Vol. Mean Fuel Temp. } ^\circ K}{273}$$

Pr = Restraining pressure from outer clad and coolant, (on basis of Zr-2 strength 20,000 psi)

T = Vol. mean fuel temperature, $^\circ C$.

Metallographic examination has been made of an NOE irradiated to 3100 MWD/T near the center of a NAE charge in a KER loop. The component operated during the first 3/4 of its exposure with a calculated volume mean fuel temperature of 400 C. The last 1/4 of its total exposure was accumulated at reduced coolant temperature. A 2.33 v/o increase in the fuel was experienced during the irradiation. Superficial examination has revealed no deficiencies in performance, no warp, no bumping or rippling of the clad surface, no crud deposits or corrosion effects, and the inner bore appears undistorted. Metallographic examination of the fuel, with both light and electron microscope, does not show swelling porosity, tearing, or cracking that is adequate to account for the 2.33 v/o increase in the fuel. The uranium shows the highly distorted structure typical of low temperature irradiation. Metallographic examination of the closure has shown no deficiencies in the closure or the heat-affected zones in the fuel and clad.

Examination has also been made on the companion NIE of the above NOE. Exposure of the NIE was 2500 MWD/T and fuel swelling was 0.76 per cent. The component performed well and there were no indications of warp, surface bumping or rippling, clad striations, and deposits or corrosion effects, and the brazed end-closures and inner bore appear unchanged. Metallographic examination indicates no deficiencies in the performance of the closure or the heat-affected zones in the fuel, clad, or bonds. Some micro and macro cracks have been observed in the fuel, but these are believed to have been caused in post-irradiation handling.

Effect of Low-Phosphorus Content on In-Reactor Performance of N-Fuel Elements

In the previous section, the performance of a charge of N-fuel assemblies, irradiated to 1600 MWD/T, was described as being typical. Six of the 14 inner elements in this charge came from a single ingot that had been analyzed at 169 ppm P. Based on the swelling data of these six fuel elements, it is apparent that minimal risk is to be associated with low-level phosphorus contents in N-fuel core metal.

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Zr-2 Cladding Strain Study

Localized thinning or necking* and ultimate failure of zircaloy-2 cladding has occurred in two fuel elements irradiated in pressurized water, and on several rods irradiated under a variety of conditions in NaK-filled capsules. The characteristics of the failures in the two environments is similar. Briefly, the necking and failures run longitudinally along the cladding and can be seen as obvious indentations on the outer surface of the cladding. In transverse section, the cladding shows highly localized strain, from both the short gage length over which the necking occurs and the indications of strain within the grains.

To study the effects of cladding thickness, cladding thickness variations, and cladding temperature on the susceptibility to failure, an irradiation program has been conducted on approximately 91 one-half inch diameter Zr-2 clad uranium rods with intentional striations in the inside surface of the cladding. About 3/4 of the samples were irradiated with various combinations of one, two, or four striations that were 15 per cent or 30 per cent of the nominal cladding thickness. The remainder of the 91 irradiated samples had no striations and served as controls. After irradiation, each sample was visually examined for indications of cladding instability or necking and then measured on several diameters. The increases in diameter from the pre-irradiation values was used to calculate a maximum total cladding strain on each fuel rod.

In Figure 4, the maximum total cladding strain is plotted as a function of the mean cladding temperature during irradiation. The curve in Figure 3 was established from an analysis of data, from both prior irradiations and the tests just described, on the effect of temperature on the failure or necking strain limits of Zr-2 cladding with variations of seven per cent or less in the cladding thickness. This curve shows the maximum total cladding strain that was observed on Zr-2 clad samples with a relatively smooth uranium-cladding interface before plastic instability was observed. The data indicate that below an average cladding temperature of 325-350 C the total cladding strain limit before plastic instability or necking occurs is approximately 1.5 per cent. At cladding temperatures above 350 C, the strain limit increases rapidly (as shown on the curve). This high temperature effect of increasing the strain limit is thought to be related to the recovery of irradiation damage.

Figure 5 is a plot of the data of total cladding strain as a function of the striation depths for samples both without intentional striations and with a single striation. The limit of the total cladding strain for a given striation depth below which plastic instability was not observed is drawn as a diagonal line L, on the left. Above this line

* The term "necking" is used to describe the localized deformation that occurs during irradiation as a result of instability. The term striation is used to describe the intentional or unintentional thinning of the cladding, resulting from extrusion or machining.

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FIGURE 4 EFFECT OF IRRADIATION TEMPERATURE ON
THE MAXIMUM TOTAL CLADDING STRAIN OF
Zr-2 CLAD RODS

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FIGURE 5 EFFECT OF STRIATION DEPTH ON TOTAL CLADDING STRAIN OF Sr-2 CLAD RODS

the data are scattered, but in general they show varying degrees of instability. This plot shows the severe influence of a single striation on the stability of Zr-2 cladding. In most cases, ruptures occurred in the series of samples with two and four equally spaced intentional striations and no worthwhile information could be obtained from them.

Several samples shown in Figure 3 had initial striation depths of approximately 15 per cent and they incurred measured cladding strains from 2.5 to 3.5 per cent without necking. On closer metallographic examination the cladding of these samples was found to have several unintentional striations approximately all of the same depth concentrated in a circumferential arc of about 90°. It is believed that each of these several striations sustained part of the total cladding strain, but no one sustained enough individually for the Zr-2 to become unstable and start necking.

Other observations made in this test were:

1. As a function of the percentage of striation depth, the stability of samples with 0.035 inch thick cladding follows about the same relationship as samples with 0.025 inch nominal cladding thickness. For absolute depths of striations, the samples with the thicker cladding are more stable.
2. There was no difference seen between the stability of cladding with 130° Vee striations and 1/8-inch radius striations.
3. Although total cladding strain or uniform strain was low on all samples, the ductility of the Zr-2 remained good as evidenced by large reductions in thickness of the cladding over the short gage lengths in the striations.

In conclusion, the results of this study on small diameter Zr-2 clad uranium rods showed:

1. At temperatures below 350 C the total cladding strain limit before necking failure is only 1.5 per cent. Above 350 C the strain limit increases rapidly.
2. The above observed low strain limit is probably related to irradiation damage to the cladding and the increasing capability for strain with increase in temperature is believed due to annealing of the irradiation damage.
3. At all temperatures, the presence of single striations greater than ten per cent of the cladding thickness reduced the limit of the total cladding strain to one per cent. At greater striation depths, the strain limit is further reduced.

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Current Reactor Fuel Development

Alternate Process Development

Hot die sizing is being investigated as an alternate cladding process for the production of fuel elements for the Hanford reactors. This cladding method produces diffusion bonded fuel elements by the application of heat and pressure to aluminum clad, nickel plated uranium cores. Development work has proceeded to the issuance of provisional process specifications and the production and irradiation of three groups of test fuels. The fourth production test is expected to be charged into C Reactor during April 1964.

Post-irradiation measurements of the first two production tests revealed unforeseen growth behavior in the hot die sized fuels as compared to the matched AlSi brazed control pieces. As reported previously, the outside and inside diameters of the hot die sized fuels increased an average of 0.008 and 0.006 inch, respectively, more than the companion AlSi brazed fuels. Radiometallurgical examination of the fuels indicated no deterioration of the bonds, and there was no indication of grain boundary tearing. Density determinations of the irradiated cores have not as yet been completed.

In February 1964, ten columns of hot die sized fuels were charged into C Reactor for irradiation to a goal exposure of 500 MWD/T. Five of the columns contained hot die sized fuels and were equipped with thermocouples. The other five columns contained alternate hot die sized and AlSi brazed control fuels. The rail support height on all the fuels was reduced 0.005 inch to compensate for dimensional changes which are expected. Discharge of this test is expected by the end of April 1964.

In an effort to determine the cause of the in-reactor dimensional changes, investigations were initiated in (1) metallurgical examination of uranium cores, both irradiated and unirradiated, (2) preferred orientation determinations by x-ray diffraction, (3) studies of dimensional changes as measured through the various steps in the hot die sizing process, (4) determinations of dimensional changes induced by thermal cycling of hot die sized fuels, and (5) continued irradiation testing of hot die sized fuels.

Metallurgical examination of uranium cores clad by the hot die sized process and the AlSi braze process revealed a slight difference in the grain structure of the hot die sized uranium cores. There was some evidence of recrystallization produced by hot working, and an increase in twinning.

X-ray diffraction studies of uranium cores taken from close positions on the original rolled rod and clad by hot die sizing and by AlSi brazing revealed different growth indices close to the outer surfaces of the fuels. The differences may be caused by heat treatment effects on the core blanks. The hot die sized cores are 0.026 inch larger in diameter than the corresponding AlSi cores, so different amounts of the high

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growth surface material were removed in the finished core machining. Figures 6 and 7 show plots of growth indices calculated from x-ray diffraction data for both cladding processes.

A study of the dimensional changes which took place as the fuels progressed through the hot die sizing process was completed. Fuel cores were plated with a double thickness of nickel and then used for the measurement of dimensional changes. The changes in core dimensions as measured after various steps in the process are shown in Figure 8 along with changes caused by process modifications.

Hot die sized fuel elements were thermally cycled to determine the effect of temperature changes on fuel dimensions. No changes were noted in 50 cycles to 350 C with quench in 10 C water; however, marked changes were observed after 50 cycles to 450 C with quench in 10 C water. Average length increase was 0.0103 inch and the internal diameters increased 0.0041 to 0.0055 inch at the ends of the fuel. As may be noted in Figure 9, the fuel took on an hour-glass configuration. Outside diameter changes were slight but tended toward a similar hour-glass effect.

A production test of twelve columns of hot die sized and AlSi brazed control fuels is being prepared for charging into C Reactor in April 1964. The test will contain resistance heat and induction heat end bonded hot die sized fuels along with AlSi control fuels. Goal exposure for the test will be 800 MWD/T.

The end bonding operation was modified by the use of induction heat as a replacement for the previous resistance heat units. Initial tests and metallographic examination indicate that the use of induction heat would permit a reduction in end bonding time and pressure. The bonding press rams are preheated and applied to the ends of the fuel element for a predetermined length of time. Thus, the necessary heat is applied to the ends of the fuel element while the midpoint of the fuel element remains at a somewhat lower temperature. Surface temperatures at the midpoint of induction heat end bonded fuel have been measured at 220 - 260 C. Midpoint core temperatures of resistance heat end bonded fuels have been measured at 400 - 450 C. A comparison of the end bonding processes follows:

	End Bonding Processes	
	Resistance Heat	Induction Heat
Preheat	5-3/4 min. in 680 C air resistance furnace.	3 min. in induction heated* press at 1 ton/in. ² pressure
Bonding	4-1/2 min. at 4 ton/in. ² with head temperatures at 655 C	1 min. 10 sec. at 4 ton/in. ²

*Starting head temperatures of induction press at 775 C with unit at constant power of about 5 kw. Temperature dropped to 600-625 C and regained temperature of 650-660 C at the end of the end bonding cycle.

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FIGURE 6
LONGITUDINAL GROWTH INDICES

FIGURE 7
RADIAL GROWTH INDICES

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FIGURE 9
HDS FUEL DIMENSION CHANGES DUE TO THERMAL CYCLING

Silicon-Aluminum Alloy Cladding

Studies have been initiated of the corrosion characteristics of high silicon-aluminum alloys under simulated reactor conditions. Preliminary work indicates that the high silicon alloys (approximately 10 per cent Si) bond more readily to the nickel plated uranium cores. Four high silicon alloys (6 - 10 per cent Si) have been ordered for further studies of high silicon-aluminum die sizing. Initial sizing work produced excellent bonds between the cladding and core.

Tube Cladding

The feasibility of cladding hot die sized fuel elements with aluminum tubing has been demonstrated. It was found that improved end bonding was obtained by the use of a thicker and tighter fitting end cap. Fabrication of a production test of hot die sized fuel clad in tubing is scheduled to begin in April 1964.

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