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THORIA TARGET ELEMENT FAILURES

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DG Kilpiel 12/22/92
PM Eick 12-23-92

K. C. Bowles
J. P. Schmidt
Reactor Engineering Unit
Process and Reactor Development Subsection
Research and Engineering Section

[redacted]

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THORIA TARGET ELEMENT FAILURES

INTRODUCTION

The thoria program at Hanford was initiated in response to an order placed by the Atomic Energy Commission for the production of uranium-233 from the irradiation of thorium or thorium oxide (thoria). The initial orders called for approximately 130 kg U-233 and subsequent orders have since increased this figure. This report provides a brief history of the thoria program, an analysis of the thoria target element failures which ocurred during the irradiation phase, and the significant changes in the fabrication methods and inspection criteria and techniques which were made in response to the large number of failures that occurred in the early stages of the program.

SUMMARY AND CONCLUSIONS

To date, the thoria program has sustained 25 thoria target element failures. Eighteen of these failures are believed to have been caused as a result of water entry through fabrication defects in the closure weld. Two of the failures were attributable to water entry through the cladding as a result of charging machine damage, and the remaining five failures were caused by water entry in some undetermined manner.

The majority of thoria element failures occurred in the early stages of the program. Upon determining, from the examination of several thoria failures, the cause of failure, Production Fuels Section initiated programs to upgrade the quality of the closure weld on thoria elements. Process improvements resulted in the thoria element failure rate being reduced by about a factor of ten from 0.035 percent (approximately twice the failure rate for uranium fuel elements) to 0.0041 percent.

The current performance of thoria target elements is very satisfactory. No future problems are foreseen that in any way should prevent the fulfillment of thoria program goals. In the event of further requests for high purity U-233, the equipment and knowledge exist with which these orders may be filled with a minimum of effort.

BACKGROUND

Irradiation of thorium, in metallic or oxide form, serves as a source of secondary nuclear fuel, uranium-233. The nuclear reaction responsible for the conversion is: $Th^{232}(n,\gamma) \rightarrow Th^{233}(\beta) \rightarrow Pa^{233}(\beta) \rightarrow U^{233}$, in which a thorium atom absorbs a neutron and emits a gamma ray in being converted to thorium-233 and decaying to paladium-233 and uranium-233 by Beta emission. By using high purity thorium or thorium oxide (thoria) and limiting the reactor exposure, the U-233 produced will contain very low levels of radioactive contaminants, primarily U-232. The incentive of the thoria program is to fill, as economically as possible, the Atomic Energy Commission's requests for high purity U-233. Utilization of the oxide instead of the thorium metal results in a 35 percent savings in process cost while producing a product of equivalent quality.

Several processes are known for the production of thoria from feed ores. The method used for preparing the thoria irradiated at Hanford is known as the Sol-Gel process. The thorium in the ore is nitrated, concentrated and purified, after which the thorium nitrate is converted to thorium oxide (ThO_2). The oxide is then dispersed in a hydrosol and reduced to a gel by evaporation. Further densification is accomplished by firing the gel at high temperatures. The final particle size, and thus the density of the thoria as a packaged composite of individual particles, is controlled in this step.

Thoria target element specifications[1] require that the oxide particles be of such size that they may be compacted to 7.5 g/cc, or about 75 percent of theoretical density, while still meeting the dissolution requirements. This requirement is based upon a balance between the need for a dense material to maintain good reactor control and minimize local variations in reactivity, and a high surface-to-volume ratio for reasonable dissolution rates.

Thoria is classed as a target material; it absorbs neutrons, but the resulting reaction does not produce neutrons and as such, thoria columns are not self-sustaining but rely on surrounding columns of enriched uranium to maintain adequate reactivity levels in the reactors. Thoria columns operate at power levels of approximately 10 percent of the adjacent supporting uranium tube powers. For fringe thoria, this is about 80 kw/column and core thoria columns operate at approximately 200 kw/column. At these heat generation rates, it is not necessary to provide for internal cooling of the target element.

The irradiation time for the thoria elements is dependent upon the U-232 impurity levels specified by the product customer of less than six parts per million (ppm). These specifications set exposures at four to six weeks for core loadings and six to nine months for fringe loadings. The U-233 produced from the scheduled loadings of the program, when blended, will contain less than 5 ppm U-232.

Due to the low powers at which thoria columns operate, cladding material presents no real problem. Corrosion is very mild with maximum cladding temperatures of 70 C and coolant outlet temperatures of about 40 C. As no internal support is provided by the core in thoria elements, strength in the clad is essential as is good formability. From the separations standpoint, a very low nickel content (nickel retards dissolution rate of thoria) is desirable. Aluminum alloy C-64 was chosen in view of these requirements. This alloy has aluminum specified at 99.3 percent minimum. Iron is the major alloying addition (0.35 percent - 0.50 percent), with silicon at 0.17 percent maximum. All impurities are specified with much lower maximums than an equivalent commercial grade alloy to minimize the parasitic neutron captures occurring in reactor.

Early calendar year 1964 marked the initial test irradiation of thoria target elements in the Hanford reactors. These elements were fabricated by development groups within what is now Battelle-Northwest Laboratories. Initial thoria testing was done in D Reactor using both thoria and metallic thorium cores. After favorable results were obtained from these tests, four tons of thoria elements were fabricated for testing in F Reactor. In September, 1964, Production Fuels Section assumed responsibility for thoria target element production.

SUMMARY OF THORIA IRRADIATION PROGRAM

Four tons of thoria target elements were charged as a fringe loading in F Reactor in early September, 1964, under the authorization of Production Test IP-648-AC. A core loading at F Reactor containing about two tons of elements followed shortly thereafter. These thoria irradiations were very successful, performing much as predicted, and no failures were encountered. The tests reached the intended exposure, which in some cases was greater than the exposures encountered in later production thoria loadings.

Post-irradiation examination of the thoria elements showed no significant damage or corrosion to the target elements. In view of the favorable performance of these tests and based upon the existence of orders for substantial amounts of U-233, irradiation of thoria as a source of U-233 on a production basis began at KW Reactor in late September, 1964.

FABRICATION OF THORIA ELEMENTS

The fabrication of thoria target elements involves no complex operation. After material acceptance, the thoria is placed in aluminum cans and compacted by vibratory methods; an aluminum cap is then inserted and welded in place to effect the necessary closure. Stringent quality control is necessary to ensure the integrity of the closure weld. Present specifications require two visual inspections, a helium leak test and radiographing of the weld. Shortly after the initiation of the thoria program, high failure rates resulted in the upgrading of fabrication methods and control measures to their present level.

THORIA ELEMENT FAILURES

Since the initial thoria loading in early CY 1964 through April 30, 1966,

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approximately 180,000 thoria target elements have been irradiated in the DUN-operated reactors. Twenty-five thoria failures occurred within this time period, comprising 0.014 percent of the total number charged. This figure represents a failure rate which is about two times greater than the failure rates experienced with uranium fuel elements fabricated by the AlSi process.

The thoria failure picture gains considerable significance when calculated for two different time periods. On about April 1, 1965, the Production Fuels Section incorporated several process and control procedures into the thoria canning process in an effort to reduce the high failure rate experienced since the beginning of the program. These procedures, to be discussed later, and their effectiveness are vividly illustrated by the reduction in failure rates experienced between the start of the program and April 1, 1965, and from April 1, 1965 to March 20, 1966, of from 0.035 percent to 0.0041 percent. These rates correspond to 20 failures in 57,800 irradiated elements for the first period and five failures in 122,200 irradiated elements for the second period.

DETECTION AND CONSEQUENCES OF THORIA FAILURES

Suspect thoria failures are detected as gradual increases in the Panellit readings for a process tube containing thoria or increases in the gamma monitor activity of headers leading from regions containing thoria charged process tubes. The following criteria have been proposed by the Process Technology Subsection for reactor shutdown due to a suspect thoria failure: 1) excessive amounts of Krypton-87 present in the reactor coolant water, 2) a Panellit pressure increase of 10 pounds accompanied by a gamma monitor trip, 3) a Panellit pressure increase of 10 pounds accompanied by gamma monitor activity which is insufficient to cause a trip, and 4) a Panellit pressure increase of 20 pounds with no gamma monitor activity. A final decision is made based upon the indication received in conjunction with the present state of the reactor involved as these criteria are still informal recommendations.

After shutdown due to one of these indications, the thoria is discharged from the suspect tube at a cost of 10 to 12 hours unscheduled reactor outage time. Discharging and recharging of the tube plus reactor turnaround requires approximately seven hours; three hours are attributable to non-equilibrium losses and one to two hours are charged to incidental losses. Of the 25 thoria failures recorded as of April 30, 1966, 14 have resulted in an unscheduled reactor outage. The remaining nine* were classified as failure suspects during the course of their irradiation, but did not give large enough indications to merit reactor shutdown. As such, they were left in reactor until the first scheduled outage following their detection, during which time they were discharged and confirmed as failures.

* In two instances, multiple failures have occurred in a single process tube. See Table I.

THORIA ELEMENT FAILURE CAUSES

Thoria target elements have been observed to fail by one or a combination of three distinct methods: 1) expansion due to internal gas pressure evidenced by bulging of the can at its weakest point and consequent coolant flow restriction, 2) weld porosity allowing entry of water into the element and the escape of fission products, often accompanied by fusing and swelling of the thoria, producing flow restriction and further fission product release, and 3) mechanical damage caused by improper handling and/or charging, producing surface damage severe enough to allow penetration of the cladding by corrosion and thus fission product release and water entry prior to attainment of goal exposure (see Figures 1, 2, 3, 3a, and 4). Table I lists the 25 thoria failures with their respective lot numbers, reactor and tube number, percent of goal exposure reached upon failure and failure classification.

Four of the elements are listed as "cause of failure unknown." One of these failures (#22 in Table I) was found to have had the cap completely severed from the remainder of the element. Some unsubstantiated indications that charging machine damage may have been partially responsible were seen on the element. No other causes for this failure were apparent upon examination. The failure listed as #8 in Table I was charged in tube 3098 at KE Reactor which was equipped for non-poisonous flux traverses. At some unknown time, either during charging or irradiation, a spline apparently became lodged in the tube. Swelling of the failed element and the presence of the spline necessitated discharge of the column by tube removal. Upon examining the failed element and the corresponding tube section, water entry appeared to have occurred through a hole in the cladding caused by a type of fretting corrosion between the target element and the lodged spline. The remaining two elements which failed for unknown reasons are assumed to have failed by one of the established mechanisms, as nothing suggesting an alternate failure mode was apparent from their failure indications or subsequent examination.

THORIA TARGET ELEMENT EXAMINATION BY RADIOMETALLURGY

Initially, 11 thoria target elements were shipped to Radiometallurgy for an analysis of their in-reactor performance and/or to determine the cause of failure. Two of the failures, from a central zone loading in KW process tube 4653, resulted in an unscheduled reactor outage. Four additional elements from this central zone loading were also included in this shipment. They were discharged from tubes which indicated failure suspects. Upon discharge, the individual failures were identified and confirmed. The remaining five elements examined possessed severe mechanical damage to the cladding. They were examined to determine whether the damage occurred prior to or during charging or upon discharging.

Upon examination of the six failed elements by Radiometallurgy, it was concluded that:

TABLE I

LISTING OF THORIA TARGET ELEMENT FAILURES

Failure No.	Date	Caused Outage	Tube Number	Lot Number	% Goal Exp.	Classification
1	1/19/65		4653-KW	TK-1808	80	EM
2	1/19/65	X	4653-KW	TK-1010	80	SM
3	1/19/65		Unknown-KW	Unknown	Unknown	SM
4	2/18/65	X	5881-KE	TC-2250	24	EM
5	2/22/65		1865-KW	TK-4340	63	SM
6	2/22/65		3083-KW	Unknown	63	SM
7	2/22/65		1374-KW	Unknown	63	Unknown
8	3/1/65		3098-KE	TC-2650	29	Unknown
9	3/25/65	X	0172-C	TC-1650	52	SM
10	3/29/65		1853-KW	TK-4440	115	SM
11	3/29/65	X	4265-KW	TK-4940	115	EM
12	3/31/65	X	5885-KE	TC-2250	45	ED
13	5/3/65	X	4971-KW	Unknown	75	EM
14	5/3/65		3553-KW	TK-6940	75	EM
15	5/3/65		3559-KW	TK-Unknown	75	Unknown
16	5/12/65	} X	1196-C	TC-1550	87	EM
17	5/12/65		2396-C	TC-1650	87	EM
18	5/14/65	X	3496-C	TP-0210	88	EM
19	6/23/65	X	0358-C	TC-1250	106	EM
20	6/26/65	X	4168-KW	TK-9240	92	EM
21	7/27/65		3853-C	TC-1250	114	EM
22	9/13/65	X	2898-KE	TD-3308	17	Unknown
23	10/21/65	X	5852-KE	TD-3508	75	EM
24	12/30/65	X	4274-KE	TK-9730A	46	ED
25	4/13/66	X	3298-KE	TCC-578	13	Not yet examined

EM - End manufacturing defect (weld closure)
SM - Side manufacturing defect
ED - Charging machine damage

"The cause of failure in all six examined elements was water entry through pin holes and/or porosity in the closure weld [Figures 7 and 8]. Three elements contained pin holes, two porosity, and one element had both."[2]

During the examination, the caps of the failed elements were ground, polished and examined in increments, starting at the exterior, in an effort to trace the path of water entry through the weld. Each of the six failures displayed at least one continuous, traceable path.

Examination of the remaining five elements revealed that the severe mechanical damage to the cladding was effected during or after discharge. The damage was severe enough to allow water entry and indicates the necessity of carefully handling the thoria elements. The thoria failure on December 30, 1965, at KE Reactor was in fact directly attributable to damage inflicted by the charging machine. This failure resulted in an unscheduled reactor outage.

Examination of subsequent thoria elements shipped to Radiometallurgy revealed no failures due to causes other than those previously mentioned.

POSSIBLE THORIA FAILURE MECHANISMS

Whereas the cause of the three types of failures is thought to result from water entry into the fuel element, the actual failure mechanisms are not thoroughly understood. The thoria target element failures always display either a swelling of the welded end of the element and/or an over-all type of swelling. Table II lists actual diameter measurements from ruptured thoria elements exhibiting the two types of swelling.

The present understanding and analysis of the swelling problems postulates a distinct cause for each of the two types. Upon examining failures displaying end swelling, the thoria is still present as a loose powder and the swelling always occurs at the weld end. The irradiated thoria, as a powder, displays no apparent increase in volume and thus the swelling must be due to internal gas pressure.

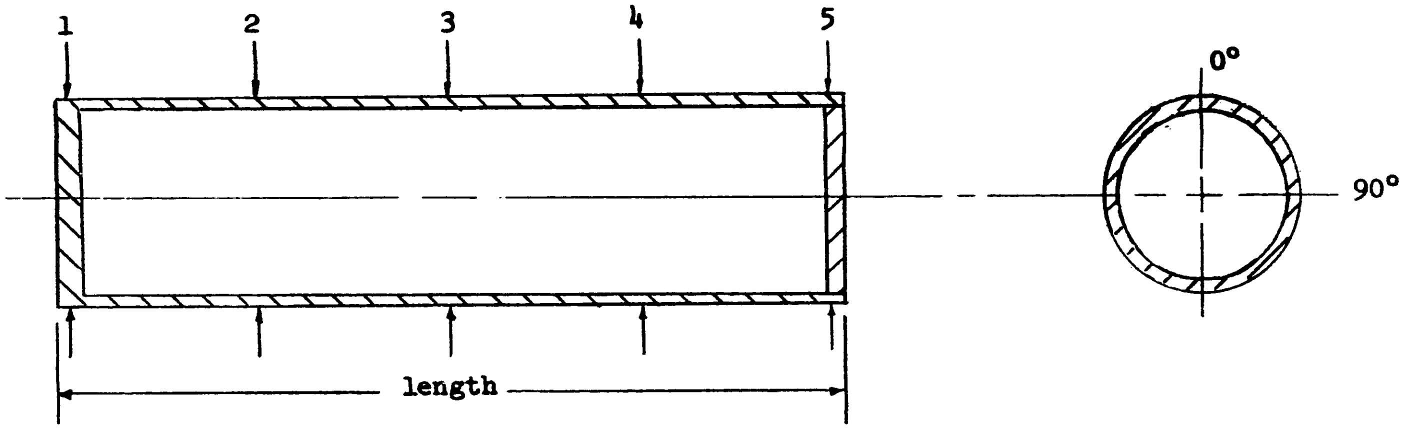
Thoria target elements experience a gradual increase in power, and thus temperature, in proportion to the U-233 buildup. Water that has entered will gradually be expelled to satisfy the conditions of pressure and increasing temperature. If the path of water entry (escape) is blocked, the pressure would then increase gradually to the point where the can could not support the internal pressure without bulging at its weakest point. As most pin holes are found to be funnel shaped with the mouth at the interior cap surface, an oxide particle could easily lodge in the funnel and block water escape quite effectively. Expansion of the aluminum cap could also seal the pin hole.

Calculations based on samples of gas taken from non-failure irradiated thoria

TABLE II

THORIA TARGET ELEMENT MEASUREMENTS

Measurement Position	Orientation	Specifications	Over-all Swelling	End Swelling
1	0°	1.596" to 1.603"	1.606"	1.609"
2	0°	1.596" to 1.603"	1.645"	1.632"
3	0°	1.596" to 1.603"	1.636"	1.638"
4	0°	1.596" to 1.603"	1.634"	1.632"
5	0°	1.596" to 1.603"	1.635"	1.713"
1	90°	1.596" to 1.603"	1.609"	1.606"
2	90°	1.596" to 1.603"	1.650"	1.634"
3	90°	1.596" to 1.603"	1.632"	1.633"
4	90°	1.596" to 1.603"	1.635"	1.629"
5	90°	1.596" to 1.603"	1.644"	1.696"
Length		8.785" to 8.985"	8.859"	9.006"



elements indicate that the pressure increase from absorbed and fission product gases does not approach the pressure necessary to produce end swelling. Thus, water entry through a defect is believed to be essential to produce the required gas volume upon which the preceding explanation is based.

The fact that bulging always occurs at the welded end of the thoria element is explained as follows: in attaining a good closure weld, it is necessary to reach temperatures which are sufficient to cause changes in the micro-structure of the aluminum within the heat affected zone. These changes, referred to as annealing, result in deterioration of the tensile properties of the aluminum within this zone. The cladding within this region will thus yield at lower internal gas pressures than the remainder of the cladding that is unaffected by the heat from the closure weld.

Examination of thoria failures exhibiting over-all swelling reveals that the oxide is no longer in powder form. Instead, it is in a fused state and occupies a larger volume. The volume increase of the oxide occurs in somewhat of a uniform manner and produces what is referred to as over-all swelling. Burst tests on thoria cans welded under present process specifications indicate the magnitude of the pressures required to produce swelling and rupture of the cladding. No swelling was evident in this test at internal pressures of 500 psig, and rupture of the cladding occurred at internal pressures on the order of 1300 psig.[3]

This type of swelling is often attributed to thoria exhibiting behavior very much as would be expected of fire clays.[4] The thoria, containing only 0.15 percent H_2O maximum as received at Hanford, absorbs some of the water entering the fuel element via pin holes or weld porosity. The water, probably absorbed as water of hydration, results in an increase in volume which causes stresses to be exerted on the aluminum cladding. When the stresses exceed the yield point of the cladding, over-all swelling will occur. The fusing of the thoria, then, would be explained by a time at temperature effect sufficient to produce the fused mass.

Experimental work[5] has also related the fusing of irradiated thoria to the firing temperature used in the Sol-Gel process. Firing temperatures of 600, 800, and 900 C resulted in the thoria's fusing during irradiation, whereas thoria fired at 1100 and 1500 C remained as a loose powder when subjected to similar conditions. The thoria utilized at Hanford is fired at approximately 1050 C. A direct relationship between this experiment and thoria performance at Hanford cannot be made due to the low exposures used in the experiment (2.4×10^{-3} Mwd/t) which may well be a factor in the fusing of the oxide powder. The experiment does indicate, however, that the stability of the oxide powder increases with firing temperatures.

The thoria elements rupturing as a result of over-all swelling display nothing else to suggest alternate mechanisms for the fusing and swelling of the thoria. The cladding exhibits a ductile type of failure at the rupture

[redacted]

(Figure 9), and the microstructure exhibits the expected distortion in grain structure within the deformed region of the rupture. Elements displaying over-all swelling may or may not exhibit additional swelling at the welded end as a result of internal gas pressure.

The fused thoria, found in elements exhibiting over-all swelling, is characterized by the occurrence of color changes to the thoria. These colors occur in circular bands running parallel to the longitudinal axis of the element, with natural yellow colored thoria, dark blue, and pale blue bands occurring from the outer diameter region to the center of the element (Figures 8 and 9). These color bands have been associated with definite grain structures[6] and are also dependent on adsorbed gases present at elevated temperatures. Below 860 C, thoria exhibits its natural yellow color and no grain growth has been observed at any lesser temperature. Between 860 and 1000 C normal grain growth proceeds and the thoria generally assumes a light blue color. Above 1000 C grain growth occurs at an accelerated rate yielding abnormally large grains having a dark blue or purple color. Failures at Hanford display the deepest blue color in a band occurring about half way between the OD and the center of the element instead of at the very center as would be concluded from the above findings. This discrepancy may be a result of inherent differences in the thoria in question, varying amounts of adsorbed gases, and variations in irradiation conditions, or more probably, some combination of these factors.

The tendencies for thorium oxide to fuse appear to be best described as a function of the firing temperature of the gel, and to be dependent upon the presence of water in the fuel element. As the information on this subject is limited, as is the information gathered from thoria element failures at Hanford, over-all swelling cannot be attributed to any one definite mechanism at this time.

FABRICATION PROCESS IMPROVEMENTS

The majority of thoria failures is a result of water entering the target element through some type of closure weld defect. The failure rates sustained from January, 1965, through March, 1965, were intolerable from an economic viewpoint of reactor operation. Upon recognizing the probable cause of the high failure rate, Production Fuels Section set about initiating both short and long range programs to reduce the fabrication defect oriented failure rate of the thoria target elements to tolerable levels.

Prior to March 1, 1965, thoria target element inspection consisted of bubble testing* and visual weld examination. The high failure rates obtained with these inspection criteria illustrated the need for more reliable methods of weld integrity analysis. On March 1, 1965, the following steps were taken: 1) all welds were radiographed, 2) all thoria elements were autoclaved, and 3) tightened visual inspection was initiated including the brushing of all welds prior to examination. The addition of the quality control measures resulted in increased reject rates.

Element reject rates increased further upon incorporating helium leak testing into the methods for weld integrity analysis. Leak testing was initiated on March 21, 1965, on which date autoclaving of fabricated thoria elements was discontinued due to the capabilities and confidence obtainable from helium leak testing and radiographing.

The significant difference in reject rates for thoria elements manufactured prior to and after March 31, 1965, is illustrated in Table III by runs of comparable sizes from these respective periods.

The effect of tighter quality inspection methods and techniques is immediately apparent from both Table III and the decrease in failure rates. The thoria target element failure rate was reduced from more than five times the rate experienced by uranium fuel elements ($\sim$0.006 percent of uranium elements fail) to approximately one-half of the uranium fuel element failure rate.

An effort was made in the summer of 1965 to utilize a 90 mil can wall thickness in place of the standard 45 mil can. It was intended that by providing a step cut in the can wall for the use of two caps and two welds, a closure of higher integrity might be produced. A lack of time for complete development, and improved techniques and efficiencies of single weld closures, resulted in a return to the 45 mil can and a single closure weld during subsequent canning runs. Further developments and process improvements have resulted in increased efficiencies for thoria element fabrication.

In conjunction with the 90 mil can, a chill block was used in an attempt to reduce the number of weld blowouts experienced. Its application proved somewhat successful in this area. Upon returning to the use of 45 mil cans, use of the chill block was retained both to reduce the number of weld blowouts and as a means of reducing the heat affected zone occurring as a result of closure welding.

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* Bubble testing consists of submerging a finished thoria element in water and drawing a partial vacuum over the surface of the water. The reduced pressure around the element should result in gas escape from the element if any pin holes or weld porosity exist. The test was found to be inadequate for the detection of fine leaks which proved to be significant to reactor performance.

[redacted]

TABLE III

REJECT DATA

	Fabricated Before 3/31/65	Fabricated After 3/31/65
Total number of elements	42,242	41,933
Elements accepted	38,307	31,068
Elements rejected by leak testing	103 (bubble)	1,082 (helium)
Number of weld rejects	2,254 (visual)	10,865 (visual and X-ray)

The elements accepted plus the elements rejected are not additive in the above table due to rewelding of weld rejects and the rejection of thoria elements for other than the reasons listed above.

FUTURE PERFORMANCE

Thoria target element fabrication efficiencies have steadily improved. Product performance using the failure rate as a comparison is presently twice as good when compared to uranium fuel elements. Past performance by Production Fuels in handling problems which were a serious threat to the success of the program gives confidence that future problems will be met and disposed of with the same efficiency and results. Continuation of the thoria program would be expected to be accompanied by improved performance of the thoria elements as a result of continued process development.

Note: See appendices for further information on thoria target element performance.

K. C. Bowles
Reactor Engineering Unit
Process and Reactor Development Subsection
Research and Engineering Section

J. P. Schmidt
Reactor Engineering Unit
Process and Reactor Development Subsection
Research and Engineering Section

[redacted]

APPENDIX A

SPECTROGRAPHIC ANALYSIS

The two ruptured thoria elements from KW tube 4653 were subjected to a spectrographic analysis of the thoria along with non-failure control samples from the same fuel lots. A complete analysis for all impurities as listed in the Thorium Oxide Target Element Engineering Specifications (RL-REA-2177) was prohibited by the high cost of this type of analysis. All impurities which produced lines corresponding to concentrations that may have been significant, however, were recorded and identified. Table IV lists the elements identified and their concentration, and Table V lists the specified impurity levels.

Examination of the table reveals an increase in aluminum concentration by a factor of 10 to 100. The source of this aluminum was undoubtedly the cladding. The increase is considered insignificant as no problems resulted in processing the irradiated thoria containing those magnitudes of aluminum, and no marked reduction in can wall thickness due to internal corrosion was observed on any of the failures. The concentrations of nickel, silicon and manganese increased by a factor of from two to ten. The only one which could be detrimental at such concentrations would be nickel as it greatly affects the dissolution rate of thoria. The maximum concentration recorded in the analysis was 200 ppm nickel and was not considered as significant in affecting thoria dissolution rates.

[redacted]

TABLE IV

SPECTROGRAPHIC ANALYSIS RESULTS ON RUPTURED THORIA ELEMENTS FROM KW PROCESS TUBE 4653

Significant Impurities	Lot TK-1808 Control Sample (Composite)	Lot TK-1808 Rupture (Composite)	Lot TK-1010 Control Sample (Composite)	Lot TK-1010 Rupture (Composite)	Lot TK-1010 Rupture (Center Sample)	Lot TK-1010 Rupture (Outside Sample)
Al	10	>1000	100	>1000	<1000	>1000
B	0.1	0.1	<0.1	<0.1	0.1	0.2
Cd	1	5	<5	--	--	5
Cr	<5	<5	5	10	5	10
Cu	50	200	50	50	50	50
Fe	50	100	100	200	200	200
Mg	100	100	100	100	100	100
Mn	<1	<1	<1	5	2	5
Ni	20	200	100	100	50	500
Si	50	50	20	100	50	50
Zn	<50	50	<50	<50	<50	50
Sn			<1	--	--	1
Bi	<5	--	2	1	--	--
Ag			--	10	--	--
Pb	--	50	10	50	--	--
P	--	200	<20	100	--	--

Concentration in parts per million, thorium oxide basis.

TABLE V

ALLOWABLE IMPURITIES IN THORIUM OXIDE -- THORIUM OXIDE TARGET ELEMENT ENGINEERING SPECIFICATIONS (RL-REA-2177)

Specific Impurities

Uranium	10 ppm max.
Boron	TNT* content + 3 ppm max.
Chloride	50 ppm max.
Silicon	TNT content + 50 ppm max.
Lithium, Cadmium, Samarium, Gadolinium, Dysprosium, and Europium	TNT content + 2 ppm max.
Total Impurities	TNT content + 500 ppm max.

Total Impurities

To determine total impurities, analyses shall be performed for the following elements: Al, Cd, Eu, Na, Sm, B, Cl, Fe, Ni, U, Be, Co, K, P, Zn, Bi, Cr, Li, Pb, C, Cu, Mg, Si, Ca, Dy, Mn, and Sn.

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* Thorium Nitrate Tetrahydrate; i.e., impurity content taken during Sol-Gel process, when thorium exists as TNT, plus specified amount. Average total impurity content obtained from TNT determination = ~600-700 ppm.

[redacted]

APPENDIX B

GAS ANALYSIS

A volume determination and quantitative analysis of the gas content have been performed on all unruptured thoria target elements examined at Radiometallurgy. The gas samples are obtained by tapping the unruptured elements and evacuating them into a bulb under a partial vacuum of 100 mm Hg. The volume was determined for standard conditions of temperature and pressure (STP) and the gas analysis was performed on a mass spectrometer. The results from a representative sample of these analyses is shown in Table VI.

The contained gas volume of a thoria element depends largely on the history of the particular thoria within an element, its canning history, as well as the temperatures and fluxes experienced within that particular element during irradiation. These factors affect the initial gas content of the thoria, the amount of gases adsorbed during fabrication, and the amounts of fission gas produced within that element during irradiation.

Using gas volume data from the element which contained the largest volume of gases, calculations showed that this volume of gas alone was insufficient to result in deformation of the cladding. Thus, water entry into the cladding is necessary to provide sufficient gas pressures to exceed the yield point of the cladding and cause swelling.

[redacted]

TABLE VI

VOLUME AND ANALYSIS DATA ON GASES FROM UNRUPTURED THORIA TARGET ELEMENTS

Capsule or Lot Number	Gas Volume (STP) - ml	Gas Analysis - Mole %					
		H_2	N_2	O_2	Kr	Xe	CO, CO_2, CH_4, He, Ar
TK-1808*	59.3	98.1	1.7	<0.1	<0.01	<0.01	<0.09
TK-1808*	55.2	1.54	95.7	<0.1	<0.001	<0.01	<0.8
TK-1010*	29.5	7.18	87.0	0.01	0.13	0.27	<6.5
36-H-15	26.7	7.16	34.6	<.01	10.07	37.14	<11.5
36-H-3	26.1	0.09	96.1	0.04	0.06	0.14	<4.5
36-H-20	27.0	12.9	24.4	<0.01	14.78	44.35	<4.5
7650-20	36.4	0.05	97.6	<0.01	<.01	<.01	<2.5
2-2-T6	7.4	3.85	86.8	0.06	<.01	<.01	<9.5
3-2-T6	16.6	0.07	94.1	0.01	<.01	<.01	<5.8
2-3-T6	7.0	35.3	56.4	0.01	0.01	0.01	<8.3
2-4-T6	5.90	15.3	75.2	0.01	0.01	0.01	<9.5
1-1-T6	9.3	75.6	13.15	0.01	0.01	0.01	<11.2
51-H-7	22.6	0.19	26.70	<.01	<.01	<.01	<3.1
29-17	34.7	<.01	96.1	<.01	0.03	0.08	<4.8
3818	35.0	<.01	97.1	<.01	<.04	0.10	<2.8
6-7	42.4	<.01	94.1	<.01	<.05	<.20	<5.7

* Lot Numbers; all other identification numbers are fuel element numbers (gas capsule numbers).

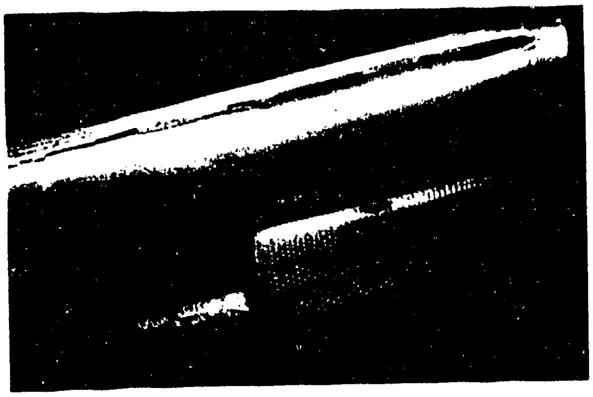

Photo #1

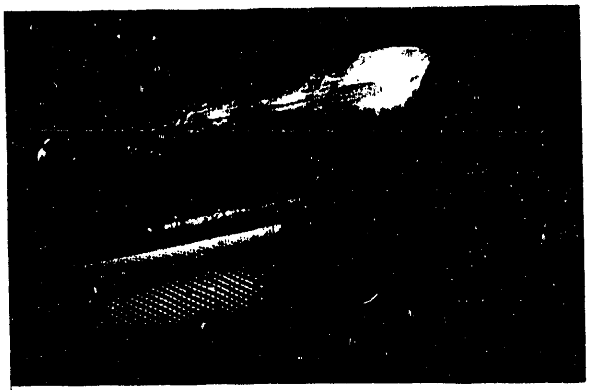

Photo #2

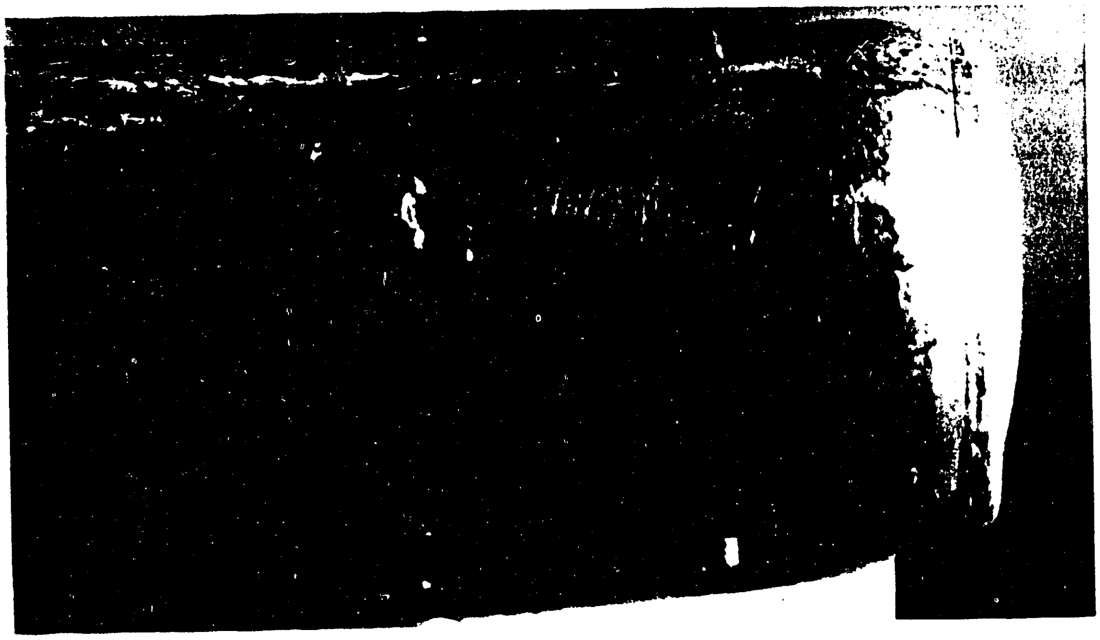

Photo #3

FIGURE 1

In Photo Number 1 the Thoria Fuel Element Rupture Displaying Overall Swelling and Longitudinal Cladding Failure. In Photos Number 2 and 3 the Thoria Fuel Element Ruptures Displaying End Swelling

DUN 1010
Uncl.

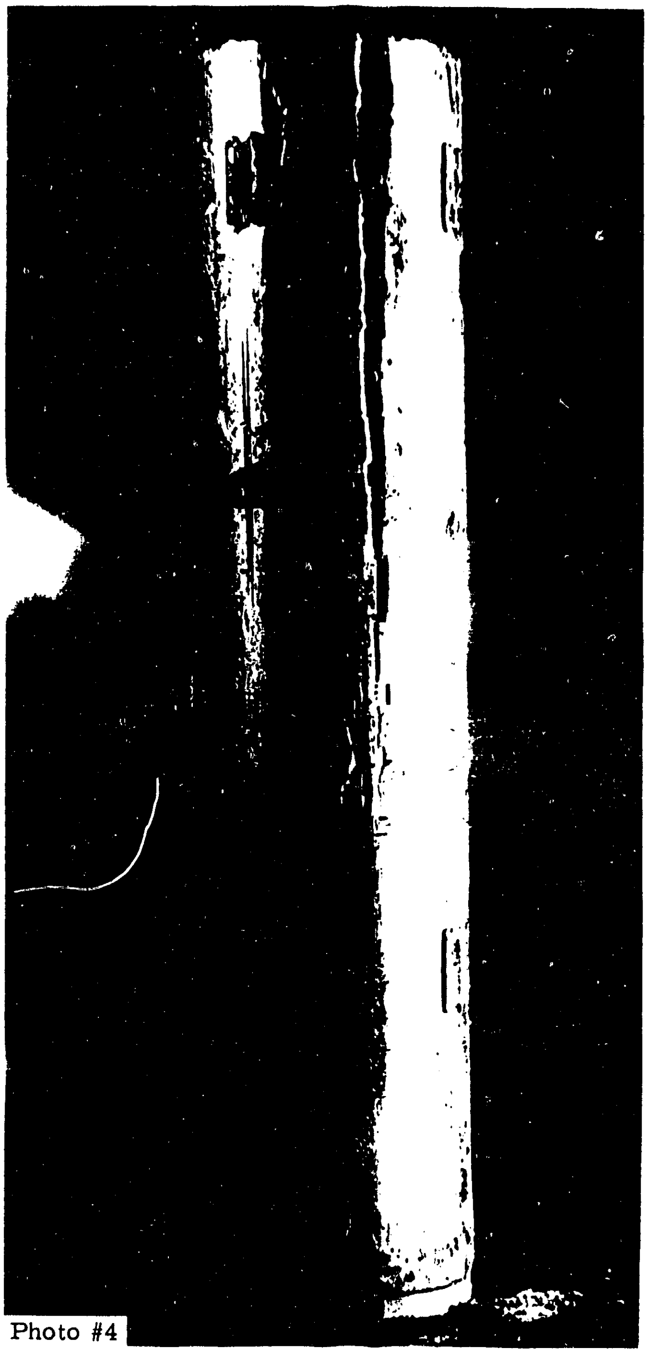

FIGURE 2

Longitudinal Cladding Failure as a Result of Overall Swelling. Note that the cap had been Removed for Examination Before this Photograph was Taken.

Photo #5A

FIGURE 3

Photos Number 5A and Number 5B: Close ups of Cladding Failure in Figure Number 2. Note that Fused Thoria is Visible Through the Failure.

DUN-1010
Uncl.

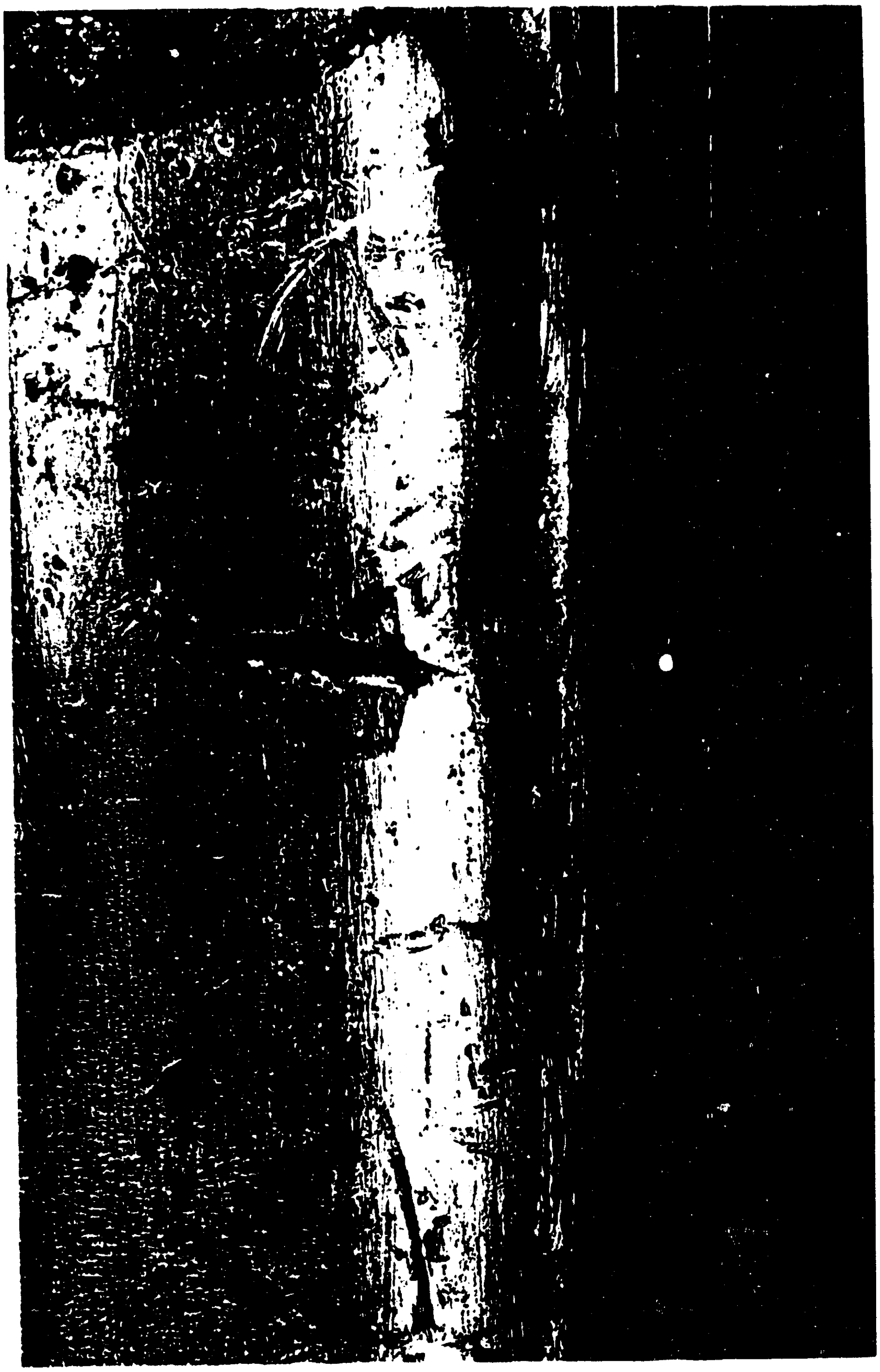

Photo #5B

FIGURE 3A

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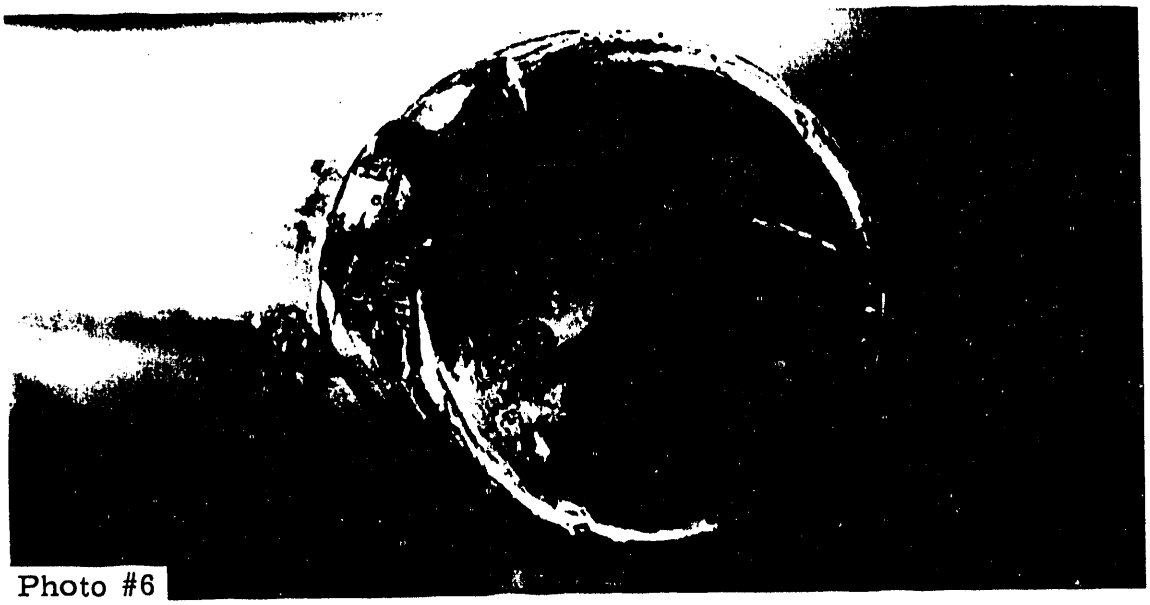

Photo #6

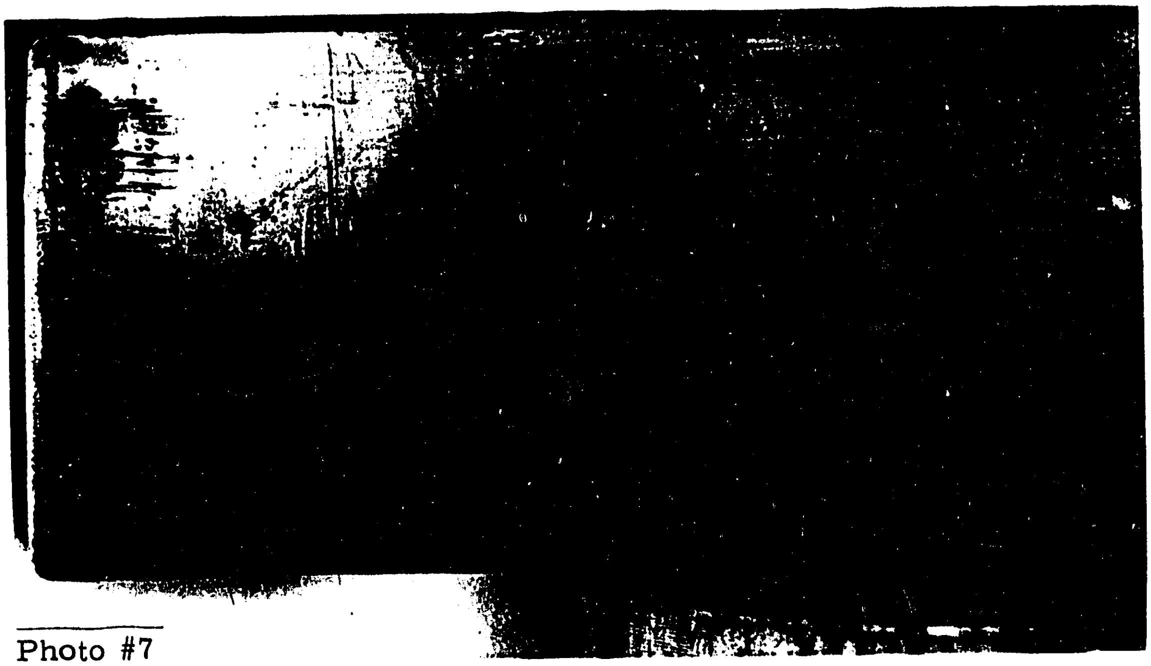

Photo #7

FIGURE 4

Thoria Fuel Elements Exhibiting Mechanical Damage. The Damage Shown here was Inflicted During or After Discharge but is Indicative of the Serious Nature of any such Damage Occurring Prior to or During The Charging Operation.

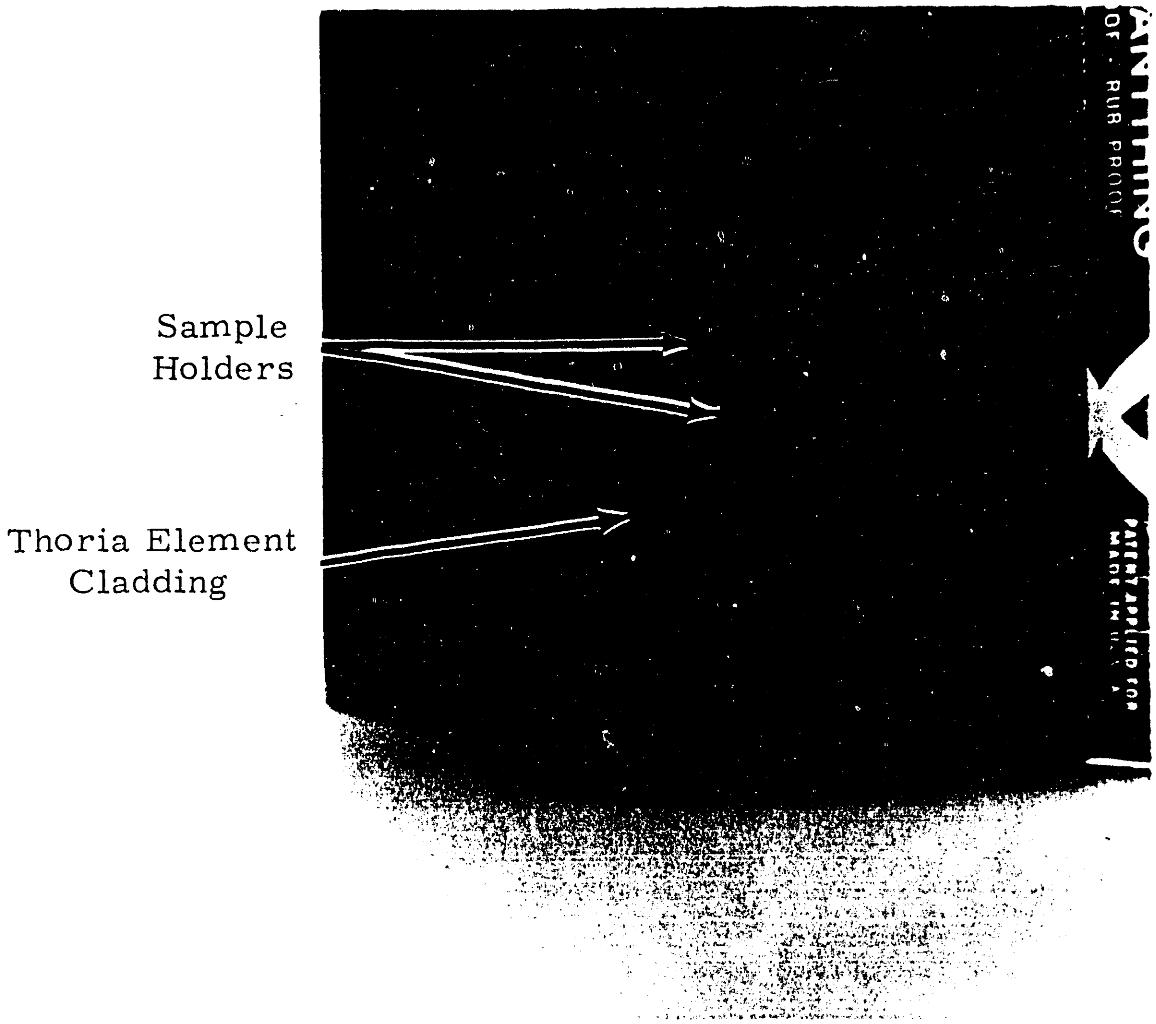

Photo #8

FIGURE 5

This Section of a Thoria Target Element Failure Displays the Colors Observed when the Thoria is Found as a Fused Mass. The Photograph was Taken Perpindicular to a Radial cut on the Element with the Color Bands Being Parallel to the Longitudinal Axis. The Section Shown is one of the OD, Yellow Band, to near the Center of the Element.

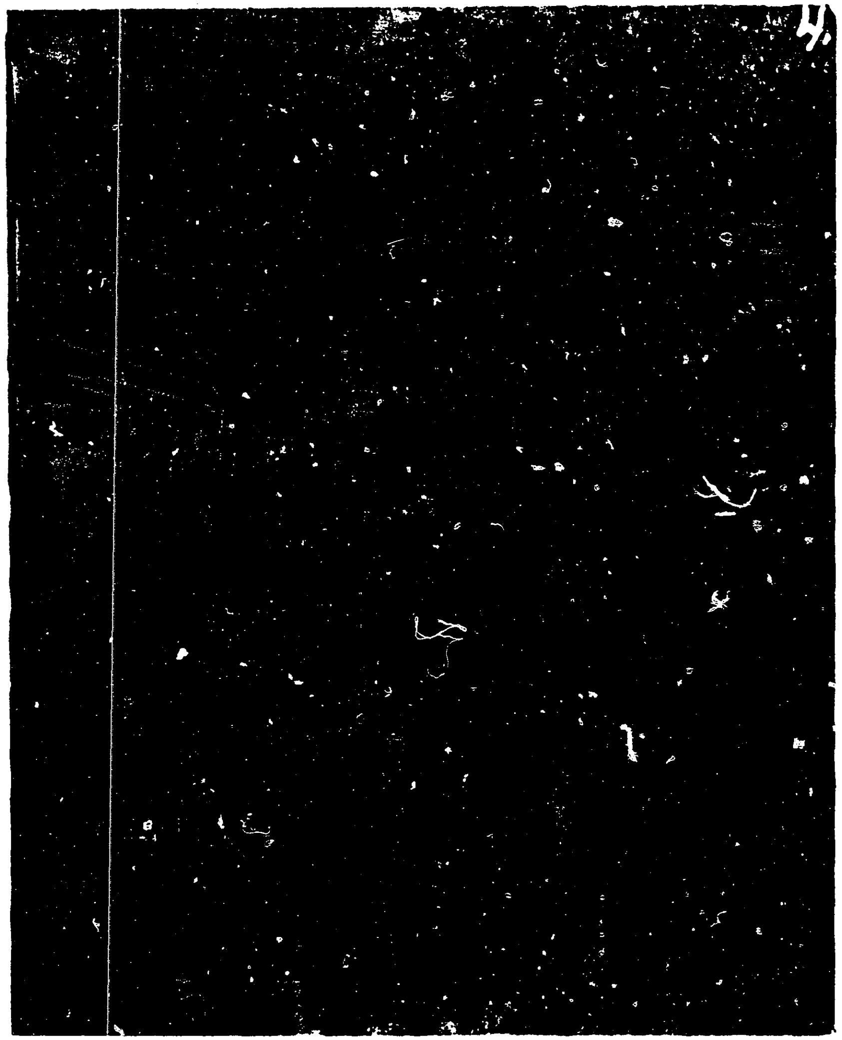

Photo #9

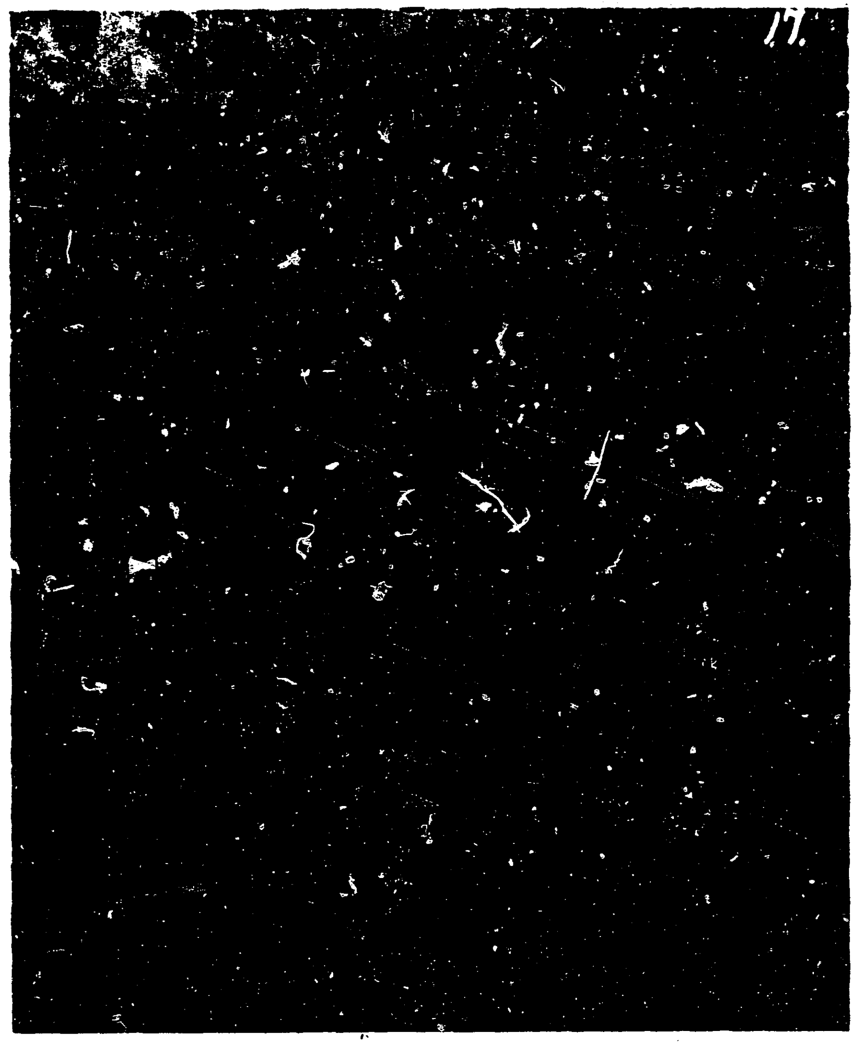

Photo #10

FIGURE 6

Photographs 9 and 10, Taken from a Large Composite of Photo #8 were from the Light Yellow and the Light Blue Color Bands Respectively. Note the Occurrence of some Dark Blue Particles Across the Entire Section.

DUN 1010
Uncl.

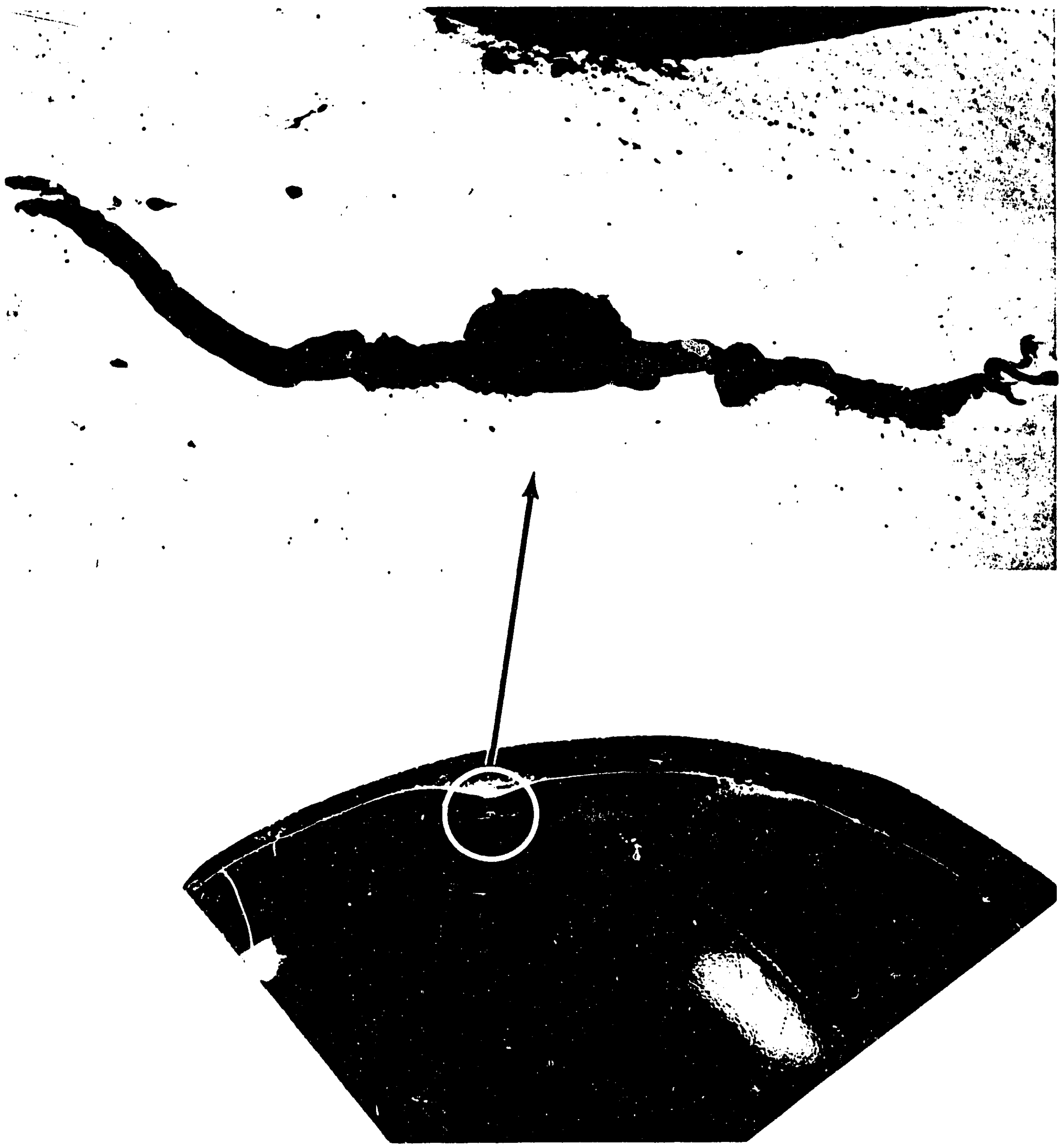

Photo #11

FIGURE 7

These Photographs were taken of a Partially Ground Through Cap of a Thoria Element Exhibiting Weld Porosity (Circled Area). This Particluar Weld Porosity Provided a Continuous Path for Water Entry to the Element Core Resulting Ultimately in a Thoria Failure.

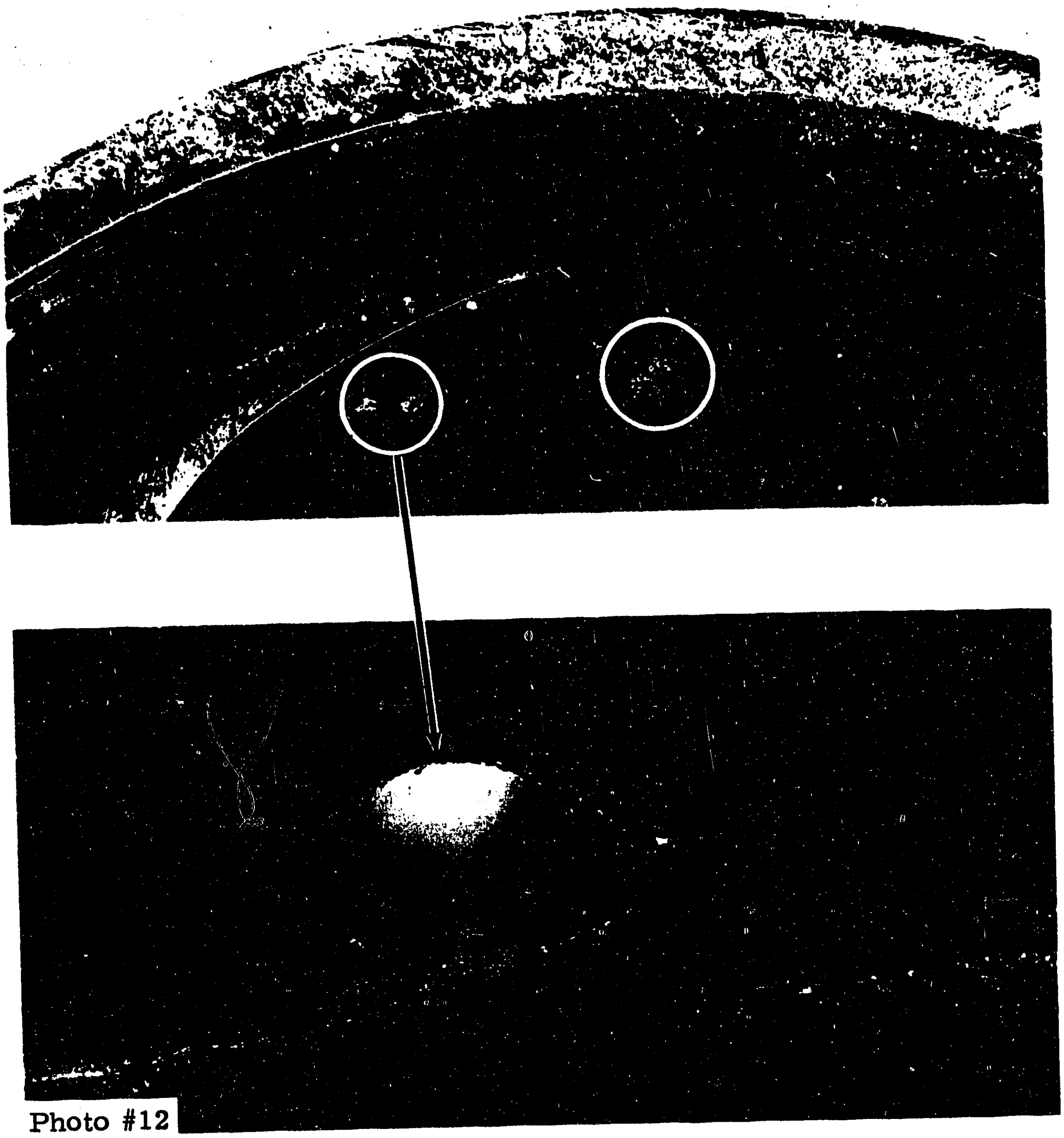

FIGURE 8

The Two Small Circular Areas on the left are Pinholes in a Thoria Element Weld. The one Corresponding to the Blow up View Provided a Continuous Path for Water Entry to the Element. The Larger Circular Area on the Right Contains an Additional Type of Weld Porosity.

DUN 1010
Uncl.

Photo #13

FIGURE 9

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

DUN 1010
Uncl.


Photo #14

FIGURE 10

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

DUN 1010
Uncl.

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