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TECHNOLOGY--FEED MATERIALS

UNITED STATES ATOMIC ENERGY COMMISSION

RESEARCH AND DEVELOPMENT IN THE FIELD
OF THORIUM CHEMISTRY AND METALLURGY

Monthly Progress Report [for] May 16 thru
June 15, 1955.

By
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Monthly Progress Report

RESEARCH AND DEVELOPMENT IN THE FIELD OF
THORIUM CHEMISTRY AND METALLURGY

For: Manager
U. S. Atomic Energy Commission
Savannah River Operations Office
P. O. Box A
Augusta, Georgia

Att: Mr. Paul J. Hagelston

Period: May 16 thru June 15, 1955
Contract No. AT(30-1)-1335

From: Horizons Incorporated
2891-2905 E. 79th Street
Cleveland 4, Ohio

By: Dr. James L. Wyatt
Assistant Technical Manager

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DATE MAR 14 1957
For The Atomic Energy Commission
H. R. Canale
Chief, Declassification Branch TC

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Summary (Cont.)

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extrusions of cold compacted, cold compacted followed by sintering, and uncompact powders. One extrusion tested for tensile and yield strengths showed strengths of the order of 28,000 lbs. per sq. in. and 36% elongation in 1/2" gauge length. These properties compare favorably with the best melted materials ever reported in literature.

Immediate future efforts will concentrate in the cell feed department, after which further electrolytic operations will be conducted as soon as cell feed is available.

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S U M M A R Y

This report covers the period May 16 through June 15, 1955 on contract AT(30-1)-1335 at Horizons Incorporated.

An internally heated chlorinator capable of containing approximately 200 pounds of cell feed was operated for the first time on a preliminary heat-up cycle without salt, and, later, on a second campaign in which molten salt actually was melted. Certain design difficulties were encountered necessitating modifications to the equipment, but basically it now has been shown that it is feasible to heat internally using a bed of graphite chunks as a resistor element in contact with molten salt.

The first extended campaign ever to be completed in an electrolytic cell at Horizons Incorporated was run during this report period, 14 consecutive cathodes being pulled during a period of 78 hours. The feasibility of continuous operation was shown, processes for removing and re-inserting cathodes were proven, and make-up cell additions containing high concentrations of thorium were used to maintain suitable ion concentrations in the electrolyte. During this campaign some mechanical difficulties were encountered which will be corrected prior to any succeeding operation of this assembly. Metal of the highest quality ever produced at Horizons was made during this campaign, and current efficiencies ranging up to 79% were recorded.

A number of experimental chlorination runs have been made in order to evaluate effects of thermal treatments on thorium oxycarbonate as regards their rate of chlorinatability, and to evaluate various chlorinating agents more completely with respect to their efficiency of utilization and conversion time factors.

Interesting results were obtained in physical metallurgy evaluations of certain of the thorium materials produced at Horizons, these tests including

RESEARCH AND DEVELOPMENT IN THE FIELD OF THORIUM
CHEMISTRY AND METALLURGY

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INTRODUCTION

Under the auspices of the Atomic Energy Commission contract AT(30-1)-1335 work has continued at Horizons Incorporated during the month ending June 15, 1955 in the general field of thorium chemistry and metallurgy.

This has been an extremely eventful contract period, major developments having occurred in every aspect of the program. Each of the phases will be discussed in detail.

Feed Materials Department

Mark XXIII Chlorinator Operation

Two runs were made early in the report period in the Mark XXIII externally heated chlorinator. The first of these was planned to produce a high thorium concentration cell feed for use in the Mark VI electrolytic campaign. Starting material for this run was a low concentration cell feed, together with additional thorium nitrate tetrahydrate, the mix planned so as to produce a product with approximately 40% thorium. The run was concluded successfully, adding approximately 260 lbs. of cell feed to the stock pile, thus completing the requirements for the Mark VI campaign.

The following run in the Mark XXIII was the first in which the starting material consisted entirely of thorium oxycarbonate produced at Horizons. A suitable quantity, mixed with sodium and potassium chlorides, was chlorinated with phosgene to yield a product containing approximately 25% thorium. This run was not needed for the Mark VI campaign and will be stockpiled in anticipation of a Mark XXVI campaign later on in the year. No difficulties were encountered in the conversion of thorium oxycarbonate, some 273 lbs. of product being recovered.

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Structural modifications to Building B, scheduled for early June, necessitated the removal of the Mark XXIII cell and all auxiliaries. This unit will not be operated again in Building B, and will be set up in Building C only as a last resort if the internally heated chlorinator does not operate satisfactorily. Mark XXIII components will be maintained in stand-by condition as a precautionary measure.

Thorium Oxycarbonate Filtration

For the past year a vertical batch-type Oliver filter has served as the filtering unit in Horizons' process for the preparation of thorium oxycarbonate. The design of this unit was limited, inasmuch as no method was incorporated in the filter for solid removal of the filter cake. In the past the unit has been utilized also as a solution tank, in which the thorium oxycarbonate filter cake has been dissolved in hydrochloric acid, the solution being pumped from the tank through ports in the bottom.

The new cell feed preparation process requires thorium oxycarbonate in solid form, and hence the requirement for a different filter design. Some months ago an Eimco continuous rotary filter 3' in diameter with a 2' face was placed on order. This now has been received but as yet is not installed. It has been necessary to remove the vertical Oliver filter prior to placing concrete foundations for the Eimco, inasmuch as the latter will be located in the same place occupied by the Oliver unit.

The vertical Oliver was operated for a period of time sufficient to allow a nominal amount of stockpiling of thorium oxycarbonate, in order to interfere as little as possible with the early experimentation on the internally heated chlorinator. The Oliver now has been dismantled and removed from the building, and wooden forms for the concrete piers and a catch basin now have been completed. Present plans call for the construction to be finished and the filter put in

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place by the end of the next month report period. This down-time on the preparation of thorium oxycarbonate filter cake will not interfere with the over-all program, inasmuch as the chlorinator unit has been dismantled and the replacement chlorinator still is undergoing pre-trial operations.

Pelletizing

The internally heated chlorinator probably will be charged with thorium oxycarbonate in the form of pellets. Final charging techniques have not been worked out, but preliminary experiments have indicated that intermittent charging will be a satisfactory technique.

A ten-ton pelletizing press capable of turning out up to 80 pellets per minute, each 1-1/2" in diameter and in varying thicknesses, has been moved into a specially prepared room adjacent to the feed preparation department in anticipation of this pelletizing requirement. More than 400 lbs. of pellets were produced during the early part of June, the raw material being spent cell feed resulting from electrolytic operations. This material, consisting largely of sodium and potassium chloride, with substantially little thorium values, was chosen for the initial melting tests on the chlorinator, inasmuch as gross contamination would be expected during the first few runs as a result of contact with freshly laid brick, graphite parts, etc. No difficulties were encountered in pelletizing this material, and a suitable quantity has been stockpiled for the internally heated chlorinator experiments.

For actual chlorination operations thorium oxycarbonate will be the major constituent to be pelletized. Preliminary mixes have been in proportions of two-thirds thorium oxycarbonate, one-sixth sodium chloride and one-sixth potassium chloride, these materials being ground and blended together prior to pelletizing.

Preliminary pelletizing experiments have not been fruitful. The thorium oxycarbonate as dried is quite coarse, resembling in some respects a cinder. It does not have the quality of pelletizing which was exhibited by the pre-fused spent salt bath, and as yet no good quality pellets have been obtained from the operation.

It is common practice to make an additive in cases of these types in order to coat the particles with a material exhibiting better compaction properties than the material to be pelletized, common agents being molasses, dextrin, starch and certain organic compounds that decompose thermally without leaving any carbonaceous materials behind. Experiments will be run during the coming weeks to evaluate a number of such additives, not only in the pelletizing operation, but in small scale chlorination experiments, to ascertain their effects, if any, on the chlorination reaction.

Continuous Belt Drier

The continuous belt drier, designed at Horizons for dehydrating thorium oxycarbonate, has been operated sporadically throughout the present report period on oxycarbonate filter cake extracted from the vertical Oliver unit. Certain minor modifications were required in the oxycarbonate feed system, these including a by-pass back to the feed tank and an agitator in the tank itself. Belt guides on the pulley sheaves also were found necessary in order to maintain alignment of the stainless steel belts.

A single Moyno pump presently is being utilized to feed both belts, the quantity of material fed to each belt being controlled by a valve in the feed line. This system has not proven satisfactory in practice, and present plans contemplate the use of individual Moyno pumps to feed the belt separately. A double hopper feed tank will be installed simultaneously with the continuous

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Eimco filter. This arrangement should provide substantially continuous operation with a minimum amount of supervision by operating personnel.

Recent small scale drying experiments have shown that prolonged heating at fairly elevated temperatures, such as 500°C., does not affect the chlorinating properties of thorium oxycarbonate adversely. The operation of the continuous belt drier therefore will have rather wide latitude, it being possible to produce a product containing in excess of 75% thorium. The actual operation of this unit in production will be determined entirely by the results of chlorination in the internally heated chlorinator.

Internally Heated Chlorinator

As a replacement for the Mark XXIII chlorinator, a brick-lined internally heated unit has been designed and placed in initial operation. A campaign in this unit should be substantially continuous, raw materials being charged periodically, chlorinated for a pre-determined period of time and the molten product drained through a taphole at the bottom. While the operation will not be completely continuous, it will nevertheless be maintained at temperature continuously, with intermittent charging, chlorinating and tapping. This unit is designed for a capacity of approximately 200 lbs. of total cell feed, with an anticipated cycle time of perhaps 36 hours. If this type of schedule could be achieved, the unit would have a capacity of approximately 1000 lbs. of cell feed per week on a continuous basis. A campaign of this type will be attempted once the unit is considered fully operable.

The first heat-up of the chlorinator took place during the week of May 23, a carbon brick bed being utilized as the resistor heater in the interior of the chlorinator. Severe arcing was encountered, apparently as a result of only a few contact points between carbon brick and the four electrodes which power the unit. At one electrode site the arcing was so severe that local

over-heating actually melted approximately 2" off the face of several bricks surrounding the electrode port. Despite these difficulties it was possible to achieve a satisfactory temperature with existing power sources, and the basic principle of using a graphite resistor bed was established on a dry basis.

On June 13 the second experimental run in the chlorinator was initiated, this time with a bed of graphite chunks approximately 1-1/2" cubed. By going to the smaller blocks it was hoped that a multiplicity of contacts would be made between the bed and the electrodes, thus eliminating insofar as possible the severe arcing which had been experienced in the previous run. This proved to be the case, but at the same time the resistance of the bed was markedly decreased, to approximately 1/4 that which had been realized in the earlier run. It therefore was necessary to place in parallel two additional power units, providing a much higher amperage, lower voltage power source than was formerly used on the operation. For this run some 500 amps at 11 to 12 volts on each pair of electrodes were realized. Thermal equilibrium at 850°C. was maintained with approximately 400 amps. at 10 volts on each pair of electrodes. The outer skin temperature of the chlorinator under these conditions was 300°F.

During the week of June 13 the chlorinator was brought slowly to temperature and pelletized spent cell feed charged periodically in small batches. Approximately 170 lbs. of spent cell feed were charged and melted with little change in the operating conditions of the power sources. The operation was surprisingly uneventful, and gives promise to the prospect of continuous operations with a minimum of manpower requirements.

On June 16, while one of the operators was making an adjustment on a lower electrode, the graphite electrode snapped in two, arcing in the electrode

port severely. This caused localized overheating, to the extent that molten salt poured out around the electrode, some 60 lbs. flowing from the chlorinator before the liquid level dropped below the electrode port opening. An incident of this type had not been anticipated, and insufficient cooling around the electrode apparently had been provided in the design of the unit.

Present plans call for the disassembly of the chlorinator down to the shell, rebuilding of electrode ports to allow entry of the electrodes on an angle instead of horizontally, the use of a somewhat larger diameter electrode, inclusion of cooling spades directly around the electrode and certain other minor modifications which were deemed desirable as a result of the above described incident. It is hoped that the unit will be back in operation before mid-July.

The run in the chlorinator in which salt was melted provided some very heartening information. It was substantially impossible to calculate even an approximate resistance for the bed of graphite chunks when immersed in molten salt, and there were certain fears on the part of several of Horizons' personnel that an extremely high amperage, low voltage power source would be required in order to power the unit satisfactorily. This now appears not to be the case, since even under molten conditions the circuit resistance is in the order of 0.025 ohm for a 16" path. The use of larger diameter chlorinators would of course improve this situation, allowing the use of somewhat higher voltages than those presently required.

The charging operation functioned quite smoothly, quick-opening valves serving as lock components for admitting pellets as desired without introducing large quantities of air into the chlorinator itself. It was also possible to observe through the charging port the actual melting of the pellets.

A surprisingly uniform thermal situation existed inside the chlorinator, temperatures at four different points and levels in the bed differing from each

other by less than 20°. Thermally the unit appears to be very easily controlled, and should lend itself well to continuous operation with a minimum of attention.

Electrolytic Operations

The only electrolytic operation carried out during the present report period was a short campaign in Horizons' Mark VI cell. This campaign actually began on June 6 and was of 78 hours duration, during which time 14 cathodes were removed from the bath. This campaign marked the first application of removal chambers, as a result of which deposits could be pulled out of the bath, cooled and removed without allowing contamination of the cell atmosphere by air. Unfortunately the removal chambers did not provide as complete protection as was desired, the base plate for the removal chamber assembly having warped either as the result of welding stresses or thermal stresses to the extent that O-rings between the base plate and its mating plate did not provide a proper seal. In principle, however, the operation went extremely smoothly, with no difficulties being encountered.

Table I presents a very brief summary of the campaign.

Table I

Statistics on First Continuous Mark VI Electrolytic Campaign

Total weight of thorium charged to cell	442 lbs.
Total weight of deposits recovered (metal + salt)	532 lbs.
* Total weight of thorium metal recovered	261.5 lbs.
Least, average and largest deposits (metal + salt)	22 lbs. 38.1 lbs. 57 lbs.
Least, average and largest thorium deposits	5.9 lbs. 15.6 lbs. 31.2 lbs.
Least, average and highest current efficiencies	21% 44.5% 79%

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Table I (Cont.)

Least, average and highest per cent metal in deposits	27%
	41.2%
	55%
Total time for electrolytic campaign	78 hours
Total number of cathodes produced	14
Average electrolytic cycle	5.5 hours
Cumulative current efficiency for entire campaign	54.2%

* Metal weight includes 219 lbs. from cathodes based upon aliquot wash evaluations and 42.5 lbs. of metal recovered from bottom of cell. Additional quantities of metal had been converted to thorium carbide in the cell crucible, accounting for the slightly low current efficiencies.

Table II presents the electrical aspects of each individual deposit produced during the campaign.

Table II

Approximate Analysis of Individual Electrolyses in Electrolytic Campaign

<u>Deposit No.</u>	<u>Total Amp.Hrs.</u>	<u>Total wt. of deposit (lbs.)</u>	<u>% metal in deposit</u>	<u>Metal Wt. (lbs.)</u>	<u>Current Effic.</u>
1	6000	42.0	30	12.6	44%
2	6000	22.0	27	5.9	21%
3	6000	34.0	32	10.9	38%
4	8000	40.5	34	13.8	36%
5	8000	57.0	44	25.1	67%
6	6000	26.0	33	8.6	30%
7	8700	25.0	50	12.5	30%
8	10,000	39.5	48	19.0	40%
9	6300	40.0	32	12.8	43%
10	8200	56.5	55	31.2	79%
11	5850	31.3	38	11.9	43%
12	10,000	47.0	51	23.9	50%
13	7000	48.5	44	21.3	64%
14	5300	23.5	41	9.6	38%

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Several points should be made clear before starting a detailed discussion of this campaign.

1. Really good quality metal has never been produced in the Mark VI cell, and high current efficiencies have never been achieved in the unit, the latter probably being associated with the actual cell design. Current concentration is at the cathode tip instead of more or less uniformly distributed over the anode and cathode surfaces, producing rather long, slender metal deposits. Inability to dissipate heat through the cathode is thought to be the cause of losing all or part of many metal deposits from the cathode, at least partially explaining why quantities of metal always are recovered from the cell bottom, and leading to the expectation of low current efficiencies even before the runs are made.

2. This campaign was the first in which a removal chamber assembly has been used on such a large scale. The prime purpose for the campaign was to prove out the operation of the removal chambers and to train green crew members, the actual results of the campaign being secondary in importance.

3. The only reason for terminating the campaign after 14 deposits was the fact that the supply of cell feed had been completely depleted. The campaign could have been continued had additional quantities of cell feed been available.

4. This campaign was the first in which continuity was attempted, i.e., all previous runs have been entirely batch-wise, the cell being emptied and recharged prior to the electrodeposition of a second, third or succeeding deposition. This run therefore was the first major experiment in which an attempt was made to replace cell feed through the addition of high thorium concentration make-up salt, and to determine just exactly what concentration of cell feed was necessary in order to perpetuate the run ad infinitum. This

run also marked the first time cell feed charging has been attempted on such a large scale, and from an appreciable height. The make-up salt was charged from a level more than 6' above the salt bath, and bath splashing was considered to be a problem.

5. Past experience in this and other electrolytic cells had generally indicated that cathode cooling was one of the major factors to be considered in a successful electrolytic operation. This series of runs was the first experiment in which water cooling was utilized over the length of the cathode, in conjunction with cathode tip cooling by means of air. Air cooling alone had been utilized in the Mark XXV cell successfully, but at considerably lower amperages and without benefit of the water cooling modification. Unfortunately this campaign was made during a period of time in which the humidity and temperature were both extremely high, and serious difficulties arose in cooling the cathode tip during the runs. Part of this was due to condensation of moisture in the aftercooler from the air compressor, and in the inability to achieve adequate air flow into the cathode tip. During the runs which ultimately showed high current efficiencies (5, 10 and 13) excellent air cooling was maintained throughout. In those runs when the current efficiency was extremely low, air cooling was ineffective, and in some cases almost non-existent, due to difficulties in the cooling system. Cooling of the cathode tip continues to be one of the most critical factors in the operation, thus substantiating previous thoughts based on Mark XXV operations.

6. The characteristic shape of metal deposits from the Mark VI has included a long, slender "carrot" some 50% longer than the cathode itself. The cathode used in these runs was 3-1/2" in diameter and 12" long, and in almost every instance the length of the deposit ranged between 17" and 20". Even with excellent cooling on the cathode it is not possible to extract an appreciable amount of heat through a deposit thickness in excess of 2" to 2-1/2".

For this reason one generally observes sagging and sliding of the lower portion of the deposit, and ultimate losses of large chunks of the deposit primarily because of excessive temperatures in the deposit some distance away from the cathode surface itself. Future cell designs, such as the Mark XXVI, will incorporate a much larger diameter cathode and will provide for current distribution minimized at the bottom of the cathode in order to eliminate some of these problems.

One might summarize by stating that it was known in advance of the campaign that the results would not be particularly good, major aims being to evaluate certain modifications to the process equipment, new ideas in product handling, and extensive training of inexperienced personnel. Perhaps somewhat fortuitously the campaign provided not only these goals but some rather gratifying results with respect to metal purity as well.

Description of Runs in Mark VI Electrolytic Campaign

A general description of each individual electrolysis in the campaign is presented below.

Prior to the start of the electrolytic campaign the Mark VI electrolytic cell underwent a series of hot evacuations to eliminate residual moisture. The cell was evacuated for two 24-hour periods at 400°F., and in a third evacuation it was brought to 1500°F. over a period of twelve hours. After this final evacuation the cell was leak tested, with the nickel slide valve closed. A leak rate of four to five microns per minute was found.

Heat-up for the electrolytic campaign started at 0800 on June 6, 1955. The cell reached operating temperature after 7-1/2 hours, and the first charge was dropped to the cell 1/2 hour later. Over a period of five hours the entire charge of 377 lbs. was charged and melted.

This initial charge was composed as follows:

136 lbs. cell feed 361-31 at 25% Th
90 lbs. cell feed 361-4 at 25% Th
145 lbs. sodium chloride
6 lbs. potassium chloride

The charge contained 57 lbs. of thorium, in the form of 92 lbs. thorium chloride. Total sodium chloride content was 206 lbs., and total potassium chloride was 83 lbs. The thorium concentration of the initial charge was 15%, and the ratio of sodium chloride to potassium chloride content was 2.5 to 1. Density of the molten charge was approximately 0.067 lb./cu. inch. The initial bath depth was 31", and the crucible wall temperature used as electrolysis control temperature was 780°C. Argon flow to the cell was maintained at 4.5 L/min., and argon flow to the removal chamber (electrolysis position) was 3 CFH.

First Electrolysis

The cathode was lowered into the cell and pre-heated to 325°C. After a 15-minute pre-heat period the DC current was started at polarizing level of 300 A and 5.5 volts. Erratic cathode temperature readings indicated a faulty thermocouple, and the cathode thermocouple was replaced. When the DC was raised to 2000 amps, it was noted that the cathode heat shields were shorting out through the slide valve assembly. This condition was corrected by placing the cathode support clamp on the cathode, and electrolysis was begun at 0010 hours on June 7, 1955. Initial DC during electrolysis was 2000 A at 11.5 V. Two separate chlorine leaks were noted between the stationary and rotating plates of the removal chamber assembly at the start of electrolysis, and the leaks increased in intensity throughout the run. The electrolysis proceeded for 5800 amp. hours, at 2000 amps. Average crucible wall temperature during electrolysis was 725°C. Cathode cooling was adequate throughout the run.

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Polarizing time at the conclusion of electrolysis was determined by the length of time required for removing the main DC cables from the cathode clamp. In general, this operation required from two to three minutes. The cathode and deposit were hoisted into the removal chamber, with argon flow to this chamber continuing at 3 CFH. The slide valve was closed, and the turntable rotated to the charging position. Following withdrawal of a bath sample, the cell was re-charged with 50 lbs. of cell feed 378-1 (containing 44% Th) and 6 lbs. NaCl. The deposit resulting from the first electrolysis weighed 42 lbs. It was 17" long and approximately 8" in diameter and appeared to be fairly well intact. Several small chunks appeared to have dropped off the deposit.

Electrolysis No. 2

Two and one half hours after withdrawal of the first deposit, the second cathode was lowered into the cell. Bath depth was 30", and polarization was carried out for 19 minutes at 400 A and 5.0 V. The electrolysis was carried out at 2000 A, with the voltage decreasing from 12.0 to 8.1. The cathode temperature thermocouple became inoperative during the run, and cathode cooling was poor throughout the run. Electrolysis control temperature was 725°C., and the run was terminated after 5700 amp. hours. A deposit weighing 21 lbs. was withdrawn from the cell. The deposit dimensions of 16" length by 6" diameter indicate that the bottom section of the deposit must have dropped into the crucible.

Electrolysis No. 3

Turn-around time between runs No. 2 and No. 3 was 1 hour, 25 minutes. Charge for this run was 50 lbs. cell feed 378-1 and 6 lbs. NaCl. The bath depth was 30". Polarization was at 400 A and 6.2 V for 10 minutes, and the

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electrolysis was carried out at 2000 amps, with the voltage at 14.5 V initially. Cathode cooling was fairly poor throughout the run. The run was terminated at 6200 amp. hours, and the deposit resulting from this electrolysis weighed 26 lbs. Deposit dimensions were 18" length by approximately 7" diameter. It appeared that considerable deposit loss had occurred along the sides of the deposit.

Electrolysis No. 4

Make-up charge for this run was 50 lbs. of cell feed 378-1 and 6 lbs. NaCl. The cathode was pre-heated to 370°C. and polarized at 400 A and 4.7 V for 18 minutes until a cathode temperature of 680°C. was reached. Electrolysis was carried out at 2000 A, with the voltage decreasing from 11 volts to 7.6 volts. Cathode cooling was insufficient during most of the run. To eliminate chlorine leakage between the stationary and revolving phases of the removal chamber assembly, a check valve in the chlorine outlet line was removed entirely and the sealing liquid was drained from the chlorine outlet. The liquid seal between the stationary and revolving plates was refilled, and no further chlorine leakage between the plates was noted for the remainder of the run. Electrolysis was terminated after 7750 amp. hours. The cathode deposit weighed 40 lbs. and measured 19" length by 7 - 8" diameter. The outside structure of the deposit was fairly open, and some yellowish-white spots were noted on the surface. In general, the deposit was fairly symmetrical, except for some losses along the sides. The bath contained approximately 7% Th after this electrolysis.

Electrolysis No. 5

Make-up charge for this run was 50 lbs. cell feed (42.9% Th) and 6 lbs. NaCl. Bath depth was 31". Polarization was at 450 A and 4 V for 10 minutes.

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Cathode cooling was very effective during this electrolysis. Electrolysis conditions were 2500 A at 11.0 to 8.4 V. The electrolysis was terminated after 8100 amp. hours. Total deposit weight was 57 lbs. The deposit dimensions were 21" length by 8-1/2" diameter. There was a slight necking at the top portion of the deposit. The deposit exterior was partially covered with a white material, and the layer of the deposit immediately adjacent to the cathode consisted of solidified salt plus metal for a distance of 1-1/2". The outer inch layer of deposit had a fairly open structure. A sample of bath following electrolysis showed almost no thorium content. The slide valve was not closed after pulling this deposit. Thirty-five minutes after withdrawal of the cathode from the cell the cathode temperature was 315°C. After one hour of cooling, the temperature dropped to 210°C., and after 1 hour, 35 minutes the cathode temperature reached 80°C. This rate of cooling was fairly representative for all of the runs.

Electrolysis No. 6

The make-up charge for this run was 50 lbs. of cell feed and 6 lbs. of NaCl. Bath depth was 31". The cathode was pre-heated to 300°C. and polarized for five minutes at 450 A and 4.6 V. Electrolysis was carried out at 2500 A, with the voltage ranging from 12.2 V to 9.2 V. At 6150 amp. hours, the voltage rose from 9.2 to 9.5, and the current dropped to 2400 amps. Electrolysis was terminated at this point. Cathode cooling was poor through most of this run. A deposit weighing 26 lbs. was obtained. It was 14" long, and the diameter ranged from 9" at the top to 6-1/2" at the bottom, indicating a large deposit loss. The deposit surface contained numerous cavities and was covered with yellowish-white salt. When the deposit was raised from the cell, a part of the cathode support clamp broke off. The nickel slide valve was closed

while welding repairs were made on the clamp. Hold-up time was 45 minutes. Thorium concentration of the bath following electrolysis was 4%.

Electrolysis No. 7

Make-up charge for this electrolysis consisted of 60 lbs. of cell feed and 7 lbs. NaCl. The cathode was pre-heated to 300°C. and polarized for 10 minutes at 400 A and 5.2 V. The first 15 minutes of electrolysis were carried out at 1200 A. Recurring trouble with the fiberglass-sleeve covered 14 ga. thermocouple wire used for cathode temperature determination necessitated the substitution of 20 ga. thermocouple wire, and no further trouble was encountered in subsequent runs. Electrolysis was continued at 2500 A, with the voltage decreasing from 11.7 V to 9.1 V. At 5800 amp. hours the voltage rose from 9.1 to 9.4, with probable accompanying deposit loss. At 8300 amp. hours, the current dropped from 2500 A to 2400 A. Final voltage reading was 9.7 volts. Cathode cooling was poor throughout the run.

Electrolysis was terminated after 8100 amp. hours. The deposit was 16" long and tapered from 7-1/2" diameter at the top to 6" at the bottom. A large portion of the bottom and some of the sides appear to have been dropped.

Electrolysis No. 8

Turn-around time between runs 7 and 8 was 45 minutes. The make-up charge consisted of 60 lbs. of cell feed, and 20 lbs. NaCl. The cathode was polarized for 10 minutes at 400 A and 5.7 volts. Electrolysis was carried out at 2000 A, with an initial voltage of 9.5 volts. During the run a voltage rise from 7.6 to 7.9 indicated loss of deposit at one point, but the run was continued for another 3000 amp. hours to a total of 9850 amp. hours. Cathode cooling was fair throughout the run. Prior to pulling the cathode, argon flow to the removal chamber was increased from 3 CFH to 50 CFH. A deposit weighing

39 lbs. 10 oz. was recovered from this electrolysis. It was 18" long and 7" in diameter. Apparently a part of the bottom and sides was lost. The surface of the deposit was contaminated with a very small amount of white and yellowish material.

Electrolysis No. 9

Make-up charge for this run was 50 lbs. of cell feed and 6 lbs. NaCl. Bath depth was 31". Between charging the cell and starting electrolysis, the crucible wall thermocouple used as electrolysis temperature control became inoperative, and the measurement of the crucible wall outside temperature was used as a guide for all subsequent runs. The cathode was polarized for 18 minutes at 400 A and 5 V until a cathode temperature of 700°C. was reached. Electrolysis was started at 2500 A and 11.5 V. Cooling was very good throughout this run.

At 5300 amp. hours a voltage rise of 0.2 volts was observed, and DC was cut to 2000 amps at 8.2 volts. At 6300 amp. hours an attempt was made to raise the DC back to 2500, but a small voltage rise was noted immediately, and the run was terminated. For pulling the deposit and subsequent cooling, the argon flow to the removal chamber was increased to 30 CFH. Color of the deposit was black. It weighed 40 lbs. and was 18" long. Diameter of the deposit was 7" except for a section at the top. The deposit sides appeared to be intact. If any deposit was lost, it appears that it must have dropped from the bottom. The bath sample following electrolysis showed essentially no thorium present.

Electrolysis No. 10

Make-up charge was 70 lbs. of cell feed. The electrolysis started at 2500 A and 10.8 V, but was changed to 2200 A and then to 2000 A at 9.0 V when analytical results pointed to a low thorium content of the bath at the end of

2

cooling was fairly ineffective throughout most of the run. The deposit obtained in this electrolysis was 15" long and very irregular in shape, indicating definite deposit loss of both bottom and sides. Color of the deposit was a dark gray, possibly indicative of sodium formation as a result of bath depletion.

Electrolysis No. 12

Make-up charge for this electrolysis was 103 lbs. of cell feed and 1-1/2 lbs. NaCl. Bath depth prior to electrolysis was approximately 36". The cathode was polarized for 8 minutes at 400 A and 5.7 volts. Later examination of the cathode showed that it was immersed in the bath to a depth of approximately 1-1/2" more than desired. Electrolysis was carried out at 2500 a up to 4200 amp. hours, at which time the DC was cut to 2000 A. A further cut to 1500 A was made after 6800 amp. hours. The run was terminated at 9900 amp. hours. Final DC was 1500 A at 6.4 volts. Cathode cooling was only fair until the DC was dropped to 1500 A. The deposit obtained in this electrolysis weighed 47 lbs. Its diameter was 9 - 10", and it was approximately 18" long, not counting a 3" long x 2" diameter growth at the bottom. In general, the surface was dendritic and covered with a slight amount of yellowish contamination. A portion of the bottom appeared to have been dropped. A sample of the bath taken near the top showed approximately 4% Th, while a sample taken at the bottom showed almost 8% Th.

Electrolysis No. 13

Make-up charge for this electrolysis was 75 lbs. of cell feed and 5 lbs. of NaCl. Bath depth was approximately 35". The cathode was polarized for 26 minutes at 425 A and 5.0 volts until the cathode reached the proper operating temperature. DC current was 2500 A during the first 5200 amp. hours, then cut to 2000 A for the next and final 1000 amp. hours. Cathode cooling was good and

easily controlled throughout this run. The electrolysis was terminated when a slight voltage rise was indicated. A sample of the bath taken near the top showed no thorium present. The bath level had dropped to 31" after removal of the deposit. The deposit obtained in this run was 20" long and approximately 7-1/2" in diameter. Its weight was 48-1/2 lbs. It was uniform and symmetrical and was contaminated with only a very small amount of surface oxidation. There was very little dendritic structure in evidence. A very smooth structure at the bottom was indicative of some possible deposit flow. It appeared that a portion of the bottom may have been dropped.

Electrolysis No. 14

Make-up charge for this run was 65 lbs. of cell feed. The cathode was polarized for 19 minutes at 400 A and 5.1 volts. 635°C. was the cathode temperature at the start of electrolysis, and DC was 2000 A throughout the run. Initial electrolysis voltage was 12.2, dropping to 8.5 but rising to 9.0 volts to cause termination of the run at 5300 amp. hours. At the conclusion of electrolysis, the deposit was polarized for 8 minutes at 450 A and 4 volts, then pulled from the cell. The nickel slide valve was closed and the cell was then shut down. The deposit resulting from this run was dendritic in structure at the top, smooth at the bottom. Its length was only 14", making it apparent that almost the entire bottom had been dropped. The bottom of the cathode was almost visible. Diameter of the deposit was 7-1/4". There was no solid salt-metal layer next to the cathode, confirming that cooling was inadequate throughout the electrolysis. Weight of the deposit was 23-1/2 lbs.

Tables III through XVI present auxiliary data on this campaign. In addition, Figs. 1 through 3 show plots of cathode temperature as a function of ampere hours during the run to illustrate the effectiveness or lack of effectiveness of deposit cooling during electrolysis.

Table III

Mark VI Electrolytic CampaignElectrolysis No. 1

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps</u>	<u>Volts</u>				
0	0	790	300	5.5	0	1100		Started polarization
	10	785	300	3.0				Replaced cathode therm.
	25	775	450	2.5	175			Raised DC
	35	770	700	2.8		815	120	
	55	765	2000	4.6	600			Started electrolysis, cathode shorting across heat shield to valve.
1	5	760	2000	11.5	725	830	125	Straightened cathode stopped short.
1	15	760	2000	10.8	1050	775	115	
1	25	760	2000	10.3	1400	675	110	
1	45	750	2000	9.5	2100	600	100	
1	55	750	2000	9.2	2450	600	100	
2	15	750	2000	8.9	3050	595	100	
2	25	750	2000	8.6	3750	585	105	
2	55	745	2000	8.4	4425	565	105	Air at 41% flow
3	15	745	2000	8.2	5100	540	100	
3	55	745	2000	8.0	6400	520	95	End electrolysis
		745	500	5.0				Polarizing

* Temp. in crucible wall

AC power input during run was 30 KW high - 5 KW low

Table IV

Mark VI Electrolytic CampaignElectrolysis No. 2

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps.</u>	<u>Volts</u>				
0	0	725	400	5.5				Polarizing
	15	725	400	5.5		840		Polarizing
	19	725	2000	12.0	0	700		Started electrolysis
	30	725	2000	10.5	325	750	85	
	55	725	2000	9.7	1090	740	95	
1	5	725	2000	9.5	1550	680	95	
1	15	725	2000	9.4	1850	-	100	Cathode thermocouple defective. Use same air flow as for No. 1
1	40	725	2000	8.8	2650		100	
2	0	725	2000	8.6	3425		100	
2	20	725	2000	8.4	4200		100	
2	40	725	2000	8.3	4700		100	
3	0	725	2000	8.1	5400		100	
3	05	725	2000	8.2	5700		100	Voltage rise End electrolysis

* Temp. in crucible wall

Table V

Mark VI Electrolytic CampaignElectrolysis No. 3

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps.</u>	<u>Volts</u>				
0	0	725	400	6.2				Polarizing
0	25	725	2000	14.5				Start electrolysis
	40	725	2010	11.3	400	675	45	Cathode temp. erratic
1	10	725	2000	9.8	1400		50	
1	30	725	2000	9.3	2100	625	55	Air flow 36%
1	50	725	2000	9.0	2700		55	
2	0	725	2000	8.8	3000	600	60	
2	15	725	2000	8.5	3550	600	60	
2	30	725	2000	8.4	4050	600	60	
3	0	725	2000	8.2	5000	600	60	
3	35	725	2000	8.1	6200	600	60	End electrolysis
			400	4.6				Polarizing while withdrawing cathode.

* Temp. in crucible wall

Table VI

Mark VI Electrolytic CampaignElectrolysis No. 4

Elapsed Time		Temp. *	DC		Amp. hrs.	Cathode Temp.	Cathode exit air temp.	Remarks
Hrs.	Min.		Amprs.	Volts				
0	0	720	400	3.3		370		Started polarization
0	15	705	2000	11.0		680		Started electrolysis
0	25	705	2000	10.0	400	725		Air at 45% flow
0	40	705	2050	9.6	1100	710		Air at 60% flow
1	10	715	2100	9.3	1800	690	75	Air at 75% flow
1	40	720	2150	8.8	3150	680	80	Air at 80% flow
1	55	725	2150	8.6	3500	670	80	
2	10	730	2000	8.0	4300	625	80	
2	40	735	2000	8.0	4900	610	55	Air at 100% flow
3	0	735	2000	7.8	5600	610	55	
3	10	740	2050	7.8	6100	610	65	Added oil to liquid seal between plates
3	25	740	2050	7.8	6600	620	60	
3	40	740	2000	7.6	7000	615	60	
3	55	740	2100	7.7	7550	610	60	Air cont. at 100%
4	5	745	2100	7.7	7750	605	60	End electrolysis
			420	3.5				Polarizing

* Temp. in crucible wall.

Table VII

Mark VI Electrolytic CampaignElectrolysis No. 5

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps.</u>	<u>Volts</u>				
0	0	750	450	4.0		385		Polarizing. Lowered AC
0	15	730	2500	11.0	0	725	85	Start electrolysis
0	25	735	2570	11.0	200	700	85	
0	35	740	2570	10.6	750	660	85	Air flow 80%
0	40	740	2500	10.3	1000	630	85	Air flow 55%
0	55	745	2500	10.2	1500	610	80	
1	10	745	2500	10.1	2050	590	80	Air flow at 70%
1	25	750	2500	9.5	2700	580	85	Cut back on AC
1	40	745	2500	9.2	3275	565	85	
2	10	745	2500	9.0	4700	540	80	Air flow at 65%
2	40	745	2500	8.6	5760	525	80	
2	50	745	2500	8.6	6150	520	80	Air flow at 70%
3	0	740	2500	8.5	7390	500	75	
3	33	740	2500	8.4	7800	495	75	Stopped electrolysis
			500	3.5				Polarized while removing cathode.

* Temp. in crucible wall.

Table VIII

Mark VI Electrolytic CampaignElectrolysis No. 6

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps.</u>	<u>Volts</u>				
0	0	740	450	4.5				Start polarization
0	12	745	2500	12.2	0	715	50	Air flow 35%
0	20	740	2500	11.4	250	755	70	Air flow 55%
0	30	745	2500	10.8	750	755	75	Air flow 65%
0	40	745	2500	10.6	1150	750	75	Air flow 85%
1	0	750	2500	10.1	2000	730	70	
1	15	750	2500	10.1	2525	720	70	Air flow 90%
1	25	740	3000	5.0				Cathode shorted. Raised 1/8" Short stopped
1	30	740	2500	9.5				
1	50	735	2500	9.5		695	65	
2	10	735	2500	9.3	4900	685	60	Air flow 80%
2	20	735	2500	9.2	5200	690	60	Much water in air
2	30	735	2500	9.2	5600	685	60	
2	45	735	2500	9.5	6150	685	60	Very rapid voltage rise
2	46	735	2400	9.7				Definite end point. Current drop - voltage rise
								Stopped electrolysis

* Temp. in crucible wall

Table IX

Mark VI Electrolytic CampaignElectrolysis No. 7

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps.</u>	<u>Volts</u>				
0	0	725	400	5.5				Start polarization
0	7	725	1200	9.3		455		Start electrolysis
0	23	725	1300	7.5		685		
0	24	725	2500	12.0		690		Raised DC to 2500
0	35	715	2550	11.5	700	675		Lowered air cooling from 40% to 20%
0	50	735	2500	10.9	1450	710		
1	15	725	2500	10.5	2740	700	110	Air flow at 30%
1	50	725	2500	10.2	3900	685	110	Air flow at 40%
2	10	730	2550	9.4	4700	665	110	Air flow at 50%
2	20	735	2500	9.3	5100	655	105	Air flow at 60%
2	40	735	2500	9.1	5900	625	105	
2	50	785**	2500	9.5	6300	620	105	Temp. measurement outside of crucible Voltage rise
3	10	785	2500	9.8	7200	595	100	
3	40	785	2500	9.7	8200	580	100	
3	50	785	2450		8700	565	100	End electrolysis
			400	3.3				Polarizing while removing cathode.

* Temp. inside crucible wall.

** Temp. outside crucible wall.

Table X

Mark VI Electrolytic CampaignElectrolysis No. 8

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps.</u>	<u>Volts</u>				
0	0	730	100	5.8				Start polarization
0	15	730	2000	10.6		705		Start electrolysis
0	25	730	2000	9.5	350	725	75	Air flow at 50%
0	40	730	2000	9.0	900	735	85	Air flow at 60%
1	0	730	2000	8.8	1450	725	80	
1	30	730	2000	8.4	2550	725	75	
1	50	730	2000	8.2	3250	675	75	
2	5	730	2000	8.0	3750	675	60	
2	35	730	2000	7.8	4650	660	60	
2	55	710	2000	7.7	5350	660	55	Raised AC
3	20	720	2050	7.8	6150	645	65	Air flow 100%
3	35	720	2000	7.6	6650	650	60	
3	42	730	2000	7.9	7050	635	60	Voltage rise
3	50	730	2000	8.0	7200	630	55	Voltage rising
4	5	735	2000	8.0	7550	625	50	
4	20	735	2000	8.1	8150	625	55	Voltage rise
4	35	735	2000	8.1	8700	625	50	
5	5	735	2000	8.1	9550	620	60	
5	12	735	2000	8.1	9850	615		Ended electrolysis
			600	5.5				Polarized while removing deposit

* Temp. in crucible wall.

Temp. outside crucible wall was 785°C.

Table XI

Mark VI Electrolytic CampaignElectrolysis No. 9

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps</u>	<u>Volts</u>				
0	0	785	400	5				Start polarization
0	18	785	2500	11.5		700	130	Start electrolysis
0	30	785	2500	10.5	600	635	105	
0	45	785	2550	10.3	1150	600	90	Air flow 100%
0	53	785	2580	10.2	1500	565	85	
1	15	785	2500	9.6	2300	530	110	
1	50	785	2550	9.4	3900	490	110	
2	15	785	2490	9.1	4855	480	110	
2	25	785	2450	9.3	5300			Voltage rise plus current drop
2	26	785	2000	8.2	5500	440	90	Lowered DC to 2000
2	35				6100			Ended electrolysis

Tried to raise current to 2500. Corresponding voltage was 9.4.
Obvious end point.

* Temp. outside crucible wall.

Table XII

Mark VI Electrolytic CampaignElectrolysis No. 10

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps</u>	<u>Volts</u>				
0	0	785	400	5.0				Start polarization
0	15	785	2500	10.8		705	30	Start electrolysis
0	20	785	2200	9.8	200	715	30	Cut DC to 2200
0	25	785	2000	9.0	350	715	30	Cut DC to 2000
0	35	785	2000	9.0	600	710	45	Air flow 76%
0	55	785	2000	8.6	1200	700	70	Air flow 95%
1	21	785	2000	8.3	1830	675		Cathode exit air
1	40	785	2000	8.2	2750	655		Thermocouple out
1	50	780	2000	8.0	3025	645		
2	12	775	2000	8.0	3750	625		
2	40	775	2000	7.8	4650	665		
3	0	765	2000	7.6	5325	595		
3	45	765	2000	7.5	6900	585		Air flow 97%
4	03	770	2000	7.3	7425	580		
4	20	770	2000	7.3	8000	580		
4	25	770	2000	7.4	8200	580		Voltage rise End electrolysis

* Temp. outside crucible wall.

Table XIII

Mark VI Electrolytic CampaignElectrolysis No. 11

Elapsed Time		Temp. *	DC		Amp. hrs.	Cathode Temp.	Cathode exit air temp.	Remarks
Hrs.	Min.		Amps	Volts				
0	0	760	400	5.6		585		Start polarizing
0	20	760	2500	13.4		660		Start electrolysis
0	30	760	2600	11.9	390	695	110	Air flow at 30%
0	50	760	2525	11.0	1100	700	113	
1	0	770	2525	11.0	1500	700	125	
1	10	765	2500	10.9	1900	695	125	Air flow to 40%
1	20	765	2550	10.6	2350	680	130	Air flow to 50%
1	45	775	2500	9.0	3500	660	110	
2	15	770	2500	9.4	4600	645	110	Voltage rise
2	30	770	2500	9.3	5300	635	110	
2	45	770	2500	9.5	5450	620	110	Voltage rise
2	50	770	2500	10.0				Another voltage rise
2	55	770	2000	8.6	6200	590	110	Lowered DC to 2000
3	5	770	2000	8.8	6500	550	95	Voltage rise
			400	3.9				Ended electrolysis
								Polarized

* Temp. outside crucible wall.

Table XIV

Mark VI Electrolytic CampaignElectrolysis No. 12

Elapsed Time		Temp. *	DC		Amp. hrs.	Cathode Temp.	Cathode exit air temp.	Remarks
Hrs.	Min.		Amps	Volts				
0	0	770	400	5.6		550		Started polarization
0	8	770	2500	13.9		590		Started electrolysis
0	13	770	2650	13.2	200	690		
0	23	770	2550	11.9	650	722	50	Air flow at 65%
0	33	770	2550	11.6	1000	718	57	
0	43	775	2550	11.5	1400	725	50	
0	53	775	2560	11.3	1800	745	60	Air flow at 70%
1	23	775	2500	10.6	3200	685	95	
1	48	775	2000	8.4	4200	660	60	Cut DC back to 2000
1	58	775	2000	8.4	4600	620	60	
2	13	775	2000	8.2	5000	620	60	
2	37	775	2000	8.1	5800	615	60	
3	08	775	1500	6.5	6800	605	60	Cut back DC to 1500
3	13	775	2000	7.8		605	60	Raised DC to 2000
3	27	775	1500	6.5	7500	605	60	Cut DC to 1500
4	3	775	1500	6.5	8300	575	55	
4	31	770	1500	6.4	9000	570	55	
5	17	770	1500	6.4	10,000	565	55	Ended electrolysis
			600	4.2				Polarized. Raised argon to 60 CFH

* Temp. outside crucible wall.

Table XV

Mark VI Electrolytic CampaignElectrolysis No. 13

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit air temp.</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>		<u>Amps</u>	<u>Volts</u>				
0	0	775	425	5.0				Start polarization
0	23	775	2500	12.3		650		Start electrolysis
0	30	775	2575	12.0		690		
0	40	775	2500	11.1	800	665	100	
0	50	775	2525	10.9	1100	645	105	
1	0	775	2540	10.7	1550	615	115	
1	25	775	2540	10.1	2550	550	100	
1	35	775	2500	9.8	3000	540	100	
2	0	775	2510	9.5	4000	520	105	
2	11	780	2550	9.5	4550	510	105	
2	26	780	2000	7.6	5200	505	95	Cut DC to 2000 amps
2	35	775	2000	7.5		485	90	Air set at 75%
2	40	775	2000	7.5	5400	480	90	Lowered AC input
3	0	765	2000	7.5	6200	470	85	
3	15	755	2000	7.5	6800	460	85	
3	22	760	2000	7.6	7000	450	85	Voltage rise. Ended electrolysis. Raised argon flow to chamber to 35 CFH

* Temp. outside of crucible wall.

Table XVI

Mark VI Electrolytic CampaignElectrolysis No. 14

<u>Elapsed Time</u>		<u>Temp.*</u>	<u>Amps</u>	<u>DC</u>		<u>Amp. hrs.</u>	<u>Cathode Temp.</u>	<u>Cathode exit</u>	<u>Remarks</u>
<u>Hrs.</u>	<u>Min.</u>			<u>Volts</u>	<u>air temp.</u>				
0	0	750	400	5.1					Start polarizing
0	19	750	2000	12.2			635		Started electrolysis
0	23	750	2000	10.8			680	55	Air flow 85%
0	30	755	2000	10.5	400		685	55	
0	40	755	2000	10.3	600		695	55	
1	5	750	2000	9.7	1580		690	65	Air flow 100%
1	15	750	2000	9.6	1900		675	65	
1	26	745	2000	9.4	2300		670	65	
1	55	745	2000	9.0	3250		655	60	
2	20	745	2000	8.8	4025		650	55	
2	30	745	2000	8.7	4700		645	55	
2	50	750	2000	8.5	5000		640	55	
3	0	750	2000	9.0	5300		650	55	Voltage rise. End electrolysis.
			450	4					Polarize while removing deposit.
									Raised argon flow to chamber to 35 CFH

* Temp. outside crucible wall.

Cathode Temperature as a Function of Ampere Hours for Electrolyses Carried out at 2000 Amperes.

Figure 1

Figure 2

Cathode Temperature as a Function of Ampere Hours for Electrolyses Carried out at 2500 Amperes.

Figure 3

Cathode Temperature as a Function of Ampere Hours for Electrolyses Carried out at an Initial Current Density of 2500 Amperes, Followed by Decreasing Ampere Flows

HORIZONS INCORPORATED

Discussion of the Electrolytic Campaign

The results of this first concerted campaign in the Mark VI cell in general were quite gratifying. To be sure, certain of the results were not those desired, and additional campaigns will have to be made before the operations are completely perfected. In general, difficulties were largely mechanical in nature, and represent items which can be corrected with nominal expenditures of time and money.

A number of factors pertaining to the process and its operation are worthy of detailed consideration. These will be taken up individually except in those instances when more than one factor has contributed to the problem.

Cell Feed

Based on very small scale electrolytic operations over the past years, it had been estimated that a cell feed containing approximately 45% thorium as metal would be required for a continuous electrolytic campaign. The exact metal concentration in the makeup salt is a function of several factors, including the per cent metal dragged out in the deposit, the thorium content of the salt being dragged out and the actual composition of the electrolyte. In general it seems that electrolytic results are improved by decreasing the proportion of potassium chloride to sodium chloride in the bath, whereas in the production of cell feed by high temperature chlorination the chlorinating operation is improved the higher the potassium chloride to sodium chloride ratio is. A 1:1 molar ratio has been used in the cell feed preparation, whereas electrolytic results seem to have been improved if the sodium chloride to potassium chloride is maintained at a 2:1 weight ratio or higher. This largely stems from the type of deposit formed on the outer perimeters of the cathode, potassium chloride concentrations in the bath giving rise to loose, dendritic

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type structures that are relatively friable and easily lost into the bath.

Electrolytes containing higher percentages of sodium chloride do not appear to be as susceptible to the formation of this type of deposit, and hence the desire to control the sodium chloride to potassium chloride ratio in the bath.

For this campaign in the Mark VI cell a cell feed ranging between 40% and 45% thorium, with 1:1 molar ratio sodium to potassium chloride, was utilized as the makeup charges. Additional quantities of sodium chloride were added in order to counteract the salt ratio, though there was a definite limitation to the amount of total salt which could be added due to the size limitation on the crucible.

Table XVII presents a hypothetical calculation using a 70% current efficiency factor, the table illustrating, using data from the Mark VI campaign, the approximate bath composition after each electrode deposit and the trend in sodium chloride to potassium chloride ratio as a result of charging the cell feed which was used in the campaign. Also listed in the table are analyses of the bath taken after each successive deposit, these being included in the column parallel to the column showing theoretical thorium based on a 70% current efficiency during electrolyses.

It may be noted that, with one exception, all thorium salt analysis samples are lower than the theoretical calculations. Part of this undoubtedly is due to inability to get representative samples of the bath, though a major portion of the discrepancy probably lies in the fact that appreciable quantities of thorium chloride are volatilized out of the cell and deposit on the walls of the upper structure of the cell and auxiliary components. Additional quantities are carried out mechanically, entrained in the chlorine gas stream.

Table XVII

Hypothetical Calculations of Electrolyte Compositions

Prior to Run No.	Total Wt. of electrolyte was (lbs.)	Wt. ThCl ₄ was (lbs.)	Wt. NaCl was (lbs.)	Wt. KCl was (lbs.)	NaCl: KCl ratio	% Th in bath was	Theor. % Th left after electrolysis	Sample salt analysis	Make-up salt added after run
1	377	91	205	81	2.53:1	15	9.85		50 lbs. (44% Th) + 6 lbs. NaCl
2	371.6	89.5	200.0	82.1	2.44:1	14.95	9.93		50 lbs. (44% Th) + 6 lbs. NaCl
3	372.5	89.1	198.8	84.6	2.35:1	14.85	10.43		50 lbs. (44% Th) + 6 lbs. NaCl
4	379.8	89.9	201.4	88.5	2.27:1	14.7	8.65	7.0	50 lbs. (42.9% Th) + 6 lbs. NaCl
5	366.2	77.8	198.3	90.1	2.20:1	13.18	8.40	2 ⁺	50 lbs. (44% Th) + 6 lbs. NaCl
6	352.9	66.9	194.8	91.2	2.14:1	11.77	6.72	4.2	60 lbs. (44% Th) + 7 lbs. NaCl
7	370.5	75.5	199.0	96.0	2.07:1	12.64	5.47	10.4	{ 40 lbs. (42.9% Th) + 20 lbs. NaCl 20 lbs. (44% Th)
8	394.9	69.5	221.8	103.6	2.14:1	10.91	2.86	6	{ 27 lbs. (43% Th) 23 lbs. (39.4% Th) + 6 lbs. NaCl
9	376.7	48.1	221.9	106.7	2.08:1	7.93	3.05	0	70 lbs. (43% Th)
10	393.7	62.0	219.0	112.7	1.95:1	9.79	3.20	0.2	{ 40 lbs. (43% Th) 20 lbs. (39.4% Th) + 10 lbs. NaCl
11	392.4	57.1	221.9	115.4	1.92:1	9.03	4.42	3.4	103 lbs. (39.4% Th) + 1.5 lbs. NaCl
12	448.1	89.6	228.3	130.2	1.75:1	12.41	5.65	4 ⁺	75 lbs. (39.4% Th) + 5 lbs. NaCl
13	451.3	81.4	232.1	137.8	1.68:1	11.20	6.58		65 lbs. (39.4% Th)
14	451.2	82.1	227.2	141.9	1.60:1	11.30	7.85		

From a purely mechanical standpoint, the actual operation of charging cell feed necessitates a change in procedures prior to any further electrolytic campaign. Make-up cell feed additions were charged through a double valve port which was built into the cell removal chamber assembly. The technique involved rotating the removal chamber plate until the charging port was directly over the cell, after which the entire charge of make-up salt was allowed to run directly into the cell through a 3" pipe. This material, weighing from 50 to 100 lbs., had a free fall drop of approximately 6 ft. before hitting the level of the electrolyte bath, which caused severe splashing. A sizeable poundage of salt was found layered above the level of the crucible top on portions of the heat reflectors and insulation in the upper portion of the cell as a result of splashing. Prior to the next campaign the crucible will be extended with a cylindrical sleeve which will prevent layering of such splashed salt, and will deflect the material back into the crucible. Because of the design of the cell it is not convenient to make a charging port on the cell itself. However, prior to final fabrication of the Mark XXVI cell, a change in feeding procedures will be incorporated which will allow direct feeding of the make-up charge at or near the cell level rather than from the removal chamber level. This was a factor which had not been considered previously, and is only one of several beneficial observations that have resulted from the Mark VI campaign.

Mechanical Handling of Cathode Deposits

The system devised for handling cathode deposits worked quite well in theory but not so well in practice. This removal chamber assembly consists of a stationary cylindrical water-cooled plate nearly 6 ft. in diameter, with an 18" diameter hole centered some 15" from the center of the plate and coinciding

with the electrolytic cell opening. A companion plate of comparable diameter, supporting two removal chambers, was designed to seat perfectly against the base plate, gas-proof seals consisting of two O-rings running the entire perimeter of the plate near the outside edge. As a further precaution, an oil well was built into the outer rim of the two plates so that a short cylinder welded to the upper plate dipped down into a channel in the lower plate, effectively making a positive oil seal against air inleakage. When the upper plate was to be rotated it was raised by a fairly simple lever mechanism onto four wheels affixed to the plate perimeter, thus raising the upper assembly approximately 1/8" off the lower plate and clearing the O-ring seals. The oil seal on the perimeter of the plates was designed so that even in the raised position oil would still make a positive seal and prevent air inleakage.

In addition to the perimeter seals of O-rings and oil, additional O-ring seals were placed around the 18" diameter holes in two positions, one around the cell top opening, the other in a position 180° from the cell opening.

The sequence of operation developed during the Mark VI campaign was approximately as follows:

1. On completion of electrolysis, raise cathode into removal chamber positioned over electrolytic cell, the lower tip of the deposit clearing the removal chamber plate.

2. Close 18" nickel gate valve at top of cell.

3. Rotate cathode assembly 90°, charge make-up salt, and rotate plate another 90° to move fresh cathode and removal chamber to position over cell.

4. Lower fresh electrode into position just above electrolyte, opening 18" gate valve just before cathode is lowered. Pre-heat cathode, then lower into bath with polarizing voltage applied.

5. When cathode deposit in cooling position has reached 80°C., remove cathode plus removal chamber to lower level and chip off deposit.

6. Replace electrode and removal chamber to cooling position on removal plate, evacuate and flush with argon. Cycle is then ready to be repeated.

The steps outlined above are a simplification of the operation, since a number of other steps are involved. These include the disconnecting of power leads, attachment of polarizing voltage leads, changing cooling lines, both air and water, switching thermocouple leads from one cathode to the other, etc. Also involved are the use of two overhead hoists, as well as clamping mechanisms on the top of each cathode removal chamber, which hold the cathodes in position during the rotation operations. It was interesting to note that the total turn-around time between the first and second electrolysis in the Mark VI campaign took approximately three hours, whereas toward the end of the campaign the turn-around time was less than 45 minutes. In general it is concluded that this operation can easily be scheduled to coincide with the length of time required for melting the make-up cell feed, so that the actual electrolytic down-time is not interfered with as a result of mechanically handling the cathode system.

The only difficulty encountered in the use of the removal chamber assembly was in the sealing between the two plates. In order to insure that no air enters the electrolytic cell, thus allowing contamination of the bath and ultimately of the deposits, a positive seal must be maintained not only around the perimeter between the two removal plates, but also around the removal chambers themselves. This is necessary in order to be able to evacuate removal chambers after chipping the metal and flush them with argon. After the first run in this series it was found impossible to evacuate the chambers and accomplish the argon flushing, and hence major contamination into the cell resulted. This was

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evident by the appearance of the deposits which were pulled up into the removal chambers; almost without exception the surfaces were noticeably contaminated with yellowish and whitish deposits, these undoubtedly being oxides or nitrides of thorium. After completion of the campaign the plates were carefully checked and it was found that the lower plate in the removal chamber assembly had warped severely into a dome shape, the center of the plate being nearly 1/2" higher than the perimeter of the plate itself. This warpage may have resulted from welding which was done on the plate just prior to its use, since it had been machined and was perfectly flat at one time. Of incidental interest, the plate was fabricated from solid nickel 1" thick. Most of the surface is covered with a water-cooled jacket, so thermal stresses could not play an important part in the warpage. An attempt will be made to straighten the plate on a large press in the very near future. If this is not successful, it will be necessary to rebuild that portion of the removal chamber assembly.

Cathode Cooling

Work in the Mark XXV cell has shown that cooling of the cathode is an extremely important factor in determining the success of an electrolytic operation. The Mark VI campaign was the first attempt to use a cathode with water-cooled portions inside the cell. The cathode, which measures some 12 ft. in over-all length, is water-cooled on the outside to within approximately 30" of the lower tip. This serves a multiple purpose of improving the rate of heat extraction from the cathode tip, allowing the use of a rubber packing gland at the top of the removal chamber to prevent air leakage around the cathode, and contributes somewhat to decreasing the over-all cell voltage by allowing the cathode to run at a lower temperature, and hence with a slightly lower path resistance for DC electrical circuitry.

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In addition to the water cooling on the shaft of the cathode, the actual cathode tip, where the metal deposit is formed, was supplied with air-cooling in order to extract heat directly at its source. Good air-cooling on the cathode tip was achieved only during three, or possibly four, of the 14 heats conducted during the campaign. The air compressor used as the air source apparently was somewhat under-capacitated, and an after cooler installed in the line did not have sufficient cross-sectional area to be effective. These factors, coupled with an extremely high humidity at the time of the campaign, caused water freezing in the cooling coils of the after cooler, thus shutting down on the total through-put of air and hence the lack of effectiveness of cooling on the cathode tip. In those runs when effective cathode cooling was maintained, the current efficiencies were respectable and little or no metal loss occurred from the deposit. At the present time it is considered that this is the major factor in the success or failure of an electrolysis.

Prior to the next campaign a much larger air compressor and after cooler will be installed, and possibly the line sizes will be increased in order to allow a greater through-put of air. If difficulties continue to be encountered in cathode cooling it may be necessary to consider some sort of a liquid coolant, though this would unnecessarily complicate what presently is considered to be a fairly simple and potentially effective technique for extracting heat.

Cathode Deposit Shape

The crucible in the Mark VI cell is slightly tapered on the inside, running from 16" at the bottom to approximately 18" at the top in diameter, the internal depth being approximately 36". Cathode dimensions in the tip area are 3-1/2" O.D. and 12" long.

It is a proven characteristic of this cell that almost without exception

the cathode geometry yields a deposit length approximately 50% longer than the length of the tip. This results partially from high concentration of current at the lower perimeter of the cathode tip, since the anode leads to the crucible are in the bottom of the cell. This is a distinct disadvantage, inasmuch as cathode cooling from inside the cathode becomes extremely ineffective as distance from the cathode surface to the outer perimeter of the deposit increases much beyond about 3". Undoubtedly as a result of localized high temperatures, the lower portions of the deposits appear actually to have undergone plastic deformation, or deposit flow, this resulting from the very low strength of the thorium metal deposit at elevated temperatures.

There is no obvious way to eliminate this undesired deposit shape characteristic of the Mark VI cell, though it is hoped that it can be alleviated in part by the insertion of a ceramic plate in the bottom of the crucible. This will of necessity force the DC path to originate along the side walls of the crucible, though undoubtedly a major concentration still will be around the lower portion of the crucible wall. In anticipation of another campaign in the Mark VI the bottom of the crucible has been covered with alundum slabs, which may prove partially effective in off-setting this undesirable cell characteristic. This factor has been taken into consideration in the design of the Mark XXVI cell, where DC anode leads will be attached to the sides of the cell, rather than through the bottom. In the Mark XXV cell, where this also was done, the results produced shorter and fatter cathode deposits which were easier to control with respect to sloughing off of the metal deposit itself.

With reference to Table II, it is probable that those runs which reflected a current efficiency lower than 60% suffered a loss of some portion of the deposit. This was not too unexpected as a result of poor cathode cooling and the cell characteristic. In all probability the next campaign also will suffer from

this difficulty, though it should be alleviated in part with more effective air cooling and with a ceramic plate in the bottom of the cell. With reference to Table I, it may be seen that the average current efficiency for the entire run was 44.5% based on the individual run current efficiencies. However, if one includes the quantity of metal recovered from the bottom of the crucible after the campaign was over, the over-all cumulative current efficiency for the campaign was raised 10%, to a level of 54.2%. Additional quantities of metal had been converted to carbide in the bottom of the cell and as such are not recoverable. It is probable that the total current efficiency throughout averaged closer to 60% for this campaign.

Cell Atmosphere

As a result of these runs it is increasingly confirmed that no air must enter the cell or removal chamber assembly at any time during the campaign. It is hoped that straightening of the removal chamber plate will alleviate leakage conditions that were encountered in this area. The one other source of air entry is the chlorine exit, which must be provided the cell in order to discharge the chlorine produced during electrolysis. Inasmuch as the chlorine quantity is fairly low, there is an obvious possibility for back diffusion of air into the cell, and ultimate contamination.

In order to prevent this occurrence, the campaign was started out with a one-way swing valve in the chlorine outlet line, the gases in addition passing through a dust collector and a bubbler before being discharged into a gas scrubber system. Because of the air leakage in the removal chamber system it was not possible to maintain either the swing valve or the liquid in the bubbler, so in essence a 2" diameter port for air diffusion back was open throughout the runs. The best method of handling this problem is still under consideration,

though it is hoped that the very simple expedient of being able to use the swing valve will prove satisfactory in due course of time.

Power Sources and Controls

This campaign marked the first extensive use of a constant current control unit on the DC power supply. It operated perfectly throughout the campaign, and generally within the specifications provided by the manufacturer. In a typical run, during which time the voltage may have dropped from a level as high as 14 to a low of approximately 9 volts, amperage control was constant to within about 10%. Slight adjustments were made periodically during the run to hold the amperage at an exact desired figure, but in general little or no attention was paid to the DC aspect of the program as a result of having this very fine piece of constant amperage controlled equipment.

Unavailable to the project at the time of the campaign was a continuous recording voltmeter. This unit has just been received and will be placed in service prior to the next campaign. It is deemed very desirable to have a drawing of the voltage as a function of time, inasmuch as a voltage rise at any time during the campaign may be indicative either of bath depletion or of the removal of some or all of the deposit as a result of over-heating, mechanical agitation or other factors.

It would be very desirable to have a precise temperature of the salt bath during each run. Because of the crucible and cell design it is not easy to insert a thermocouple directly into the salt, and hence reliance must be placed on a wall temperature taken in the crucible itself. The crucible wall may be higher, the same as, or lower than the temperature of the bath, since the crucible is heated externally by resistance bars, within the wall itself as a result of DC current flow, and internally as a result of high temperatures

generated at the surface of the cathode deposit. Provision is being made in the Mark XXVI cell for direct reading thermocouples in the bath, but it is not at the present time considered feasible for the Mark VI.

Depth of Electrolyte

In reference to Table XVII it may be noted that there was a general increase in the calculated weight of the electrolyte on a theoretical basis. In the write-up also it may be noted that at certain times the level of the electrolyte approached 36", whereas the starting depth was 30" to 31". This increase in salt stems largely from the general increase in potassium chloride, and a slight increase in sodium chloride which was added in order to counteract the potassium chloride content. It may also be noted that the sodium to potassium chloride ratio changed materially during the run, and would have necessitated terminating the campaign before an additional ten cathodes had been pulled because of the high potassium chloride content. Based on the data in Table XVII, which in turn were based on the actual campaign, it now appears mandatory that make-up cell feed contained at least 49% thorium by weight. If a cell feed could be supplied which contained 50% thorium, then there would be no increase in potassium chloride through the runs. Much larger sodium chloride additions would have to be made in conjunction with the cell feed additions, but this would result in maintaining the starting sodium chloride to potassium chloride ratio throughout the campaign. Efforts for future operations therefore will be aimed at the production of a 50% cell feed.

General Remarks on the Mark VI Campaign

This campaign in the Mark VI cell was quite informative and educational. The basic design of cell removal chamber assemblies was proven to be satisfactory,

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its operation having also been proven out to the point where no time is lost in the electrolytic cycle as a result of turn-around. The mechanics of removing deposits intact, cooling them, and recovering the metal have been proven throughout an extended operation, metal losses easily being accounted for because of other factors. The feasibility of semi-continuous operation has been shown, and nothing in the basic process has been found which would tend to indicate a lack of commercial feasibility.

This campaign was operated with five men on each shift. Because of the physical layout of the operation a crew of four men would allow operation with desired ease, and a crew of three would at times prove marginal. Proper building layouts and the slight re-design of cathode clamps, together with slightly improved locations of electrical and cooling water connections might improve upon the personnel requirements to the extent that two individuals per shift would be adequate, but this would not be too feasible at Horizons. Inadequate working space and building head room would preclude these possibilities.

The Mark XXVI cell will contain nearly six times as much salt as the Mark VI, and as such will involve much more prodigious operations in starting up and shutting down than the Mark VI. The quantity of materials involved also is very appreciably larger, and it is Horizons' opinion that every assurance should be made prior to the initial operation of the Mark XXVI that no metal deposits will be lost into the cell or other factors encountered which would necessitate its shutting down. It therefore is planned that at least one more campaign in the Mark VI will be run, since the time and materials at stake in proving out the operational techniques of the process are very much less than in the Mark XXVI. The further fact that the final design of the Mark XXVI still has not been formalized would preclude its being finally constructed for several weeks to come adds to the logic of further runs in the Mark VI.

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Experimental Program

Experimental Preparation of Cell Feed

Some small scale experiments were run in which an attempt was made to produce pure thorium chloride at a temperature of 725°C. using alternatively carbon tetrachloride and hydrogen chloride gas as the reducing-chlorinating agents. The materials have been submitted for analysis but no results as yet are available.

An experiment was conducted in which thorium oxycarbonate was chlorinated in a sodium chloride-potassium chloride eutectic solution, sparging through a mixture of hydrogen chloride and chlorine gases. Timewise the results showed that there was no advantage in using the gas mixture as compared with the straight use of hydrogen chloride gas alone. Several experimental runs were made in order to evaluate the effectiveness of several chlorinating gases on the rate of chlorinating thorium oxycarbonate in a molten system of different compositions. These experiments included chlorination in straight sodium chloride, in various ratios of sodium chloride and potassium chloride, and in potassium chloride. Using hydrogen chloride gas as the chlorinating agent, it was found that nearly three times the amount of time for total chlorination was required to convert a unit quantity of oxycarbonate to chloride in straight sodium chloride as was required in the eutectic mixture. In another experiment, in which a weight ratio of two sodium chlorides to one potassium chloride were used as the menstruum, again the reaction was much slower than in the eutectic mixture.

The relative chlorination rates in the various salt systems, and comparisons of various chlorinating agents in specific salt systems, are presented in Figures 4 and 5.

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Figure 4

Comparative Rates of Chlorination of Thorium Oxycarbonate in Different Fused Salt Systems with Hydrogen Chloride as the Chlorinating Agent.

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Figure 5

Comparative Rates of Chlorination of Thorium Oxycarbonate in Different Fused Salt Systems with Phosgene as the Chlorinating Agent.

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Figure 6

Comparative Rates of Chlorinating Thorium Oxycarbonate in Various Fused Baths as a Function of Drying Times.

Material "A" - Belt dried oxycarbonate

Material "B" - Material "A" re-dried at 300°C. for two hours.

Material "C" - Material "A" re-dried at 500°C. for two hours.

Hydrogen chloride gas as chlorinating agent.

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Figure 7

Comparative Rates of Chlorinating Thorium Oxycarbonate in Various Fused Baths as a Function of Drying Times.

Material "A" - Belt dried oxycarbonate

Material "B" - Material "A" re-dried at 300°C. for two hours.

Material "C" - Material "A" re-dried at 500°C. for two hours.

Phosgene as chlorinating agent.

An evaluation of thorium oxycarbonate, dried at various temperatures for different period of time, also was made with respect to the rate of chlorination in a fused salt system. Hydrogen chloride gas was utilized as the chlorinating agent. The results of these experiments are shown in Figure 6.

Comparable experiments on dried thorium oxycarbonate treated for two hours at elevated temperatures also were run, phosgene being utilized as the chlorinating agent. The results of these experiments are shown in Figure 7. Several other groups of experiments are planned for the coming week, including a group in which carbon plus chlorine will be utilized as the combination reducing and chlorinating agent. Qualitatively preliminary experiments would tend to favor this system over hydrogen chloride gas.

Physical Metallurgy Investigations

Powder Metal Extrusions

Two extrusion runs were made during the last month. For the first of these extrusions a cold-pressed compact weighing 162 grams was compacted under 10,000 pounds pressure. The unit measured 1" in diameter and 1.76" long. It was extruded without prior sintering, the die components being coated with colloidal graphite.

After evacuating the extrusion chamber and flushing with dried argon the assembly was heated to 1280°F. Extrusion rates as a function of load were as follows:

<u>Extrusion Load (lbs.)</u>	<u>Ram Travel in inches/minute</u>
25,000	0.017
28,000	0.037
30,000	0.051

Extrusion No. 2 in this period was from a cold press and sintered thorium compact. This compact was heated in vacuum for one hour at 1000°C., after which it was extruded under conditions identical to the one described above.

The surface appearance of the extruded rods was generally satisfactory. The front end had a somewhat rough finish, inasmuch as complete compaction had not occurred. This condition extended only about 1" back from the outlet end of the extruded rod, after which point the surface became smooth and uniform. A photograph of the first of these extrusions is shown in Fig. 8.

The densities of the extruded sections were very near theoretical for thorium. These values were determined by the usual method, i. e., weighing of the samples in water and then in air. These two extrusions had densities of 11.59 and 11.60 grams/cm³ respectively.

Hardness values were obtained using a Vicker's diamond pyramid under a load of 500 grams. The first of these extrusions averaged 62 vpn while extrusion No. 2 had an average value of 69 vpn.

Tensile specimens were machined from a previous extrusion and were tested during this report period. The photograph in Fig. 9 shows an as-machined specimen and two specimens after testing. The elongation was determined over a 1/2" gauge length, and averaged 36%. The tensile strengths obtained so far have averaged 28,000 psi; these values are normal for high purity thorium.

A typical microstructure of an extruded section is shown in Fig. 10. The dark gray phase is thorium oxide, while the white precipitate phase has not as yet been identified. The microstructure of an arc-melted sample of electrolytic thorium powder is shown in Fig. 11 for comparative purposes.

Micro-hardness determinations of the white phase indicated a hardness in excess of 400 vpn.

Figure 8

Thorium metal rod formed from a sintered powder metal compact by extrusion at 1285°F.

Magnified 2X

Figure 9

Tensile specimens from the third extrusion of thorium.

No. 1 Tensile - 26,200 psi Elong. 34%

No. 2 Tensile - 31,000 psi Elong. 38%

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Magnified 250X

Figure 10 - Photomicrograph of typical thorium rod cross-section after powder compact extrusion at 1285°F.

Figure 11 - Photomicrograph of melted thorium powder in as-cast form. Specimen melted in water-cooled copper hearth furnace.

Of incidental interest, the photomicrograph in Figure 11 is that of a melted specimen from electrolysis No. 14 in the recent Mark VI campaign. This material in the as-cast form showed a Brinell hardness of 55 and, after annealing 15 minutes at 800°C., showed an average Brinell hardness of 48. This is the softest metal ever produced at Horizons Incorporated and is indicative of what can be achieved in the electrolytic process when properly implemented.

Conclusions

The month ending June 15 has been quite productive, it having been shown without question that it is possible to operate Horizons' electrolytic process on a semi-continuous basis without major difficulties. Problems encountered were almost entirely mechanical in nature and can be corrected by proper design of key pieces of equipment and minor modifications in processes.

Despite the fact that potentially severe air leakage into the cell and components could have occurred throughout each run, some of the metal produced in the cell was the best that has ever been produced at Horizons. The outlook for excellent quality metal therefore looks extremely good when the process is expanded to a somewhat larger scale than has heretofore been attempted.

Current efficiencies as high as 79% were registered in the campaign, and rapid turn-around times were achieved on most of the runs during the campaign.

Future plans contemplate a several week interim during which time no major campaigns will be attempted. Efforts will be concentrated in the cell feed preparation area, further major electrolyses to be postponed until such time as an adequate quantity of cell feed again has been stockpiled. Efforts therefore will be concentrated during the coming weeks on the operation of the internally heated chlorination, with the anticipation of successful preparation

of 50% thorium cell feed in adequate quantity and quality to supply the Mark VI and ultimately the Mark XXVI cells.

This concludes the progress report for the month ending June 15, 1955.

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