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AEC RESEARCH AND DEVELOPMENT REPORT

PREPARATION OF A HIGH ENERGY GAMMA SOURCE BY ELECTRODEPOSITION OF ^{228}Th FROM FUSED SALTS

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Springfield, Virginia 22151
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DP-1166

Chemistry
(TID-4500)

PREPARATION OF A HIGH ENERGY GAMMA SOURCE BY ELECTRODEPOSITION OF ^{228}Th FROM FUSED SALTS

by

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June 1969

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CONTRACT AT(07-2)-1 WITH THE
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ABSTRACT

A remote operation procedure is described for the preparation of sources containing several curies of ^{228}Th for photoneutron counting. The neutrons from (α,n) reactions in the source are reduced by electrodepositing the ^{228}Th from a molten cesium chloride-lanthanum chloride mixture into a liquid bismuth cathode. Sources prepared by this method have less than 6% of the neutron background of $Th(NO_3)_4$ sources. The deposition efficiency for the process is 60-75%.

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INTRODUCTION

Deuterium irradiated with gamma rays of energy greater than 2.23 Mev emits neutrons that can be detected with conventional counting equipment. This (γ,n) reaction provides a convenient interference-free method for determining D_2O in natural or deuterium-enriched water, and is potentially valuable as a means of control for D_2O production plants and for monitoring D_2O in tracer studies. However, the technique has not been exploited because of the unavailability of suitable long-lived gamma sources with low neutron backgrounds.

Of the isotopes emitting gammas of sufficient energy to fission deuterium, only ^{208}Tl in secular equilibrium with its long-lived ^{228}Th parent has a half-life long enough for continuous monitoring. However, such sources must be free of nuclei of $Z < \sim 45$ to eliminate high neutron background produced by (α,n) reactions.

This report describes the preparation of a ^{228}Th gamma source containing only matrix elements of $Z > 50$. Two possible methods of source preparation were considered: the direct conversion of freshly precipitated thorium oxide to the iodide via a vapor phase reaction with an organic iodide and electrodeposition of thorium into a liquid metal cathode from a molten salt solution. The molten salt deposition was selected because it appeared to require less development time.

SUMMARY

A procedure was developed for the preparation of ^{228}Th sources (>1 Ci) with low neutron backgrounds. Neutron production from (α,n) reactions was reduced by electrodepositing the thorium as the metal into a bismuth metal matrix. The deposition efficiency for the process was 60-75%.

A 4.3-curie source prepared by this technique exhibited a neutron background of 3.3×10^3 n/(sec)(Ci) as compared to 5.8×10^4 n/(sec)(Ci) for a crystalline salt source. Preliminary results indicate that the quality of the source is adequate for applications such as control of deuterium production processes and tracing of deuterium in soil moisture studies.

DISCUSSION

EXPERIMENTAL

Johnson[1] deposited thorium metal from molten lithium chloride-potassium chloride eutectic into a liquid zinc cathode. However, a small scale ^{228}Th source prepared by this procedure exhibited high neutron backgrounds attributable to (α,n) reactions on the isotopes of zinc.[2]

There was no previous report of the electrodeposition of thorium from melts of heavy metal salts. Of the many molten salt mixtures available, $LaCl_3$-CsCl eutectic (mp 543°C) was selected for study on the basis of the following criteria:

(1) Low melting point. (Thorium halides are volatile above 650°C.)

(2) Low mutual solubilities of salt and cathode metal.

(3) Adequate solubility of thorium in the cathode metal.

Preliminary studies using cathodes made of lead were abandoned because much of the ^{228}Th was found to concentrate in a black deposit between the cathode and the glass container. Negligible deposits were found using bismuth cathodes, and bismuth was used in all subsequent work.

Reagents

All chemicals in this study were reagent grade except for the CsCl-$LaCl_3$ eutectic which was specially prepared by Anderson Physics Laboratories, Inc., Champaign, Illinois. Two lots of eutectic were prepared: one with 0.5% and one with 10% thorium chloride.

The lithium chloride-potassium chloride-silver chloride (8.75%) for the silver reference electrode was prepared by grinding 14.3 g AgCl with 74.6 g LiCl and 74.6 g KCl. The mixture was placed in an electrolysis cell mounted in the furnace, and hydrogen chloride gas was passed through the mixture as it was slowly heated to the melting point. The molten salt was purged with HCl gas for 30 minutes. After cooling, the salt was transferred to a dry box, pulverized, and stored in a desiccator.

Pure silver wire (70 mil) for the reference electrode was purchased from Handy and Harmon, New York.

The helium and hydrogen chloride gases were passed through a column of Type 5A Linde molecular sieve and two columns of anhydrous magnesium perchlorate prior to introduction into the electrolysis cell. Argon was substituted for helium for preparing curie-level sources.

Radiothorium, ^{228}Th, was purchased from the Radiochemical Centre, Amersham, Buckinghamshire, England. The material was supplied as the crystalline nitrate in 0.75-in.-dia. x 1.5-in.-long platinum crucibles; the quoted specific activity was 100 curies/gram. The platinum crucibles were encased in welded stainless steel capsules to prevent the release of ^{220}Rn to the environment. For the tracer studies, about 5 millicuries of ^{228}Th was incorporated into about 10 g of $CsCl-LaCl_3 + 0.5\% ThCl_4$ as follows: About 100 μl of a hydrochloric acid solution containing the tracer were pipetted onto 10 g of the crushed salt in an electrolysis cell. The salt was dried under vacuum and then slowly heated to melting as HCl gas was passed through it. The molten salt was then purged with HCl gas until clear (~30 min), cooled, crushed, and stored over anhydrous magnesium perchlorate. A weighed aliquot of the salt was dissolved in HCl and electrodeposited,[8] and the thorium was determined by alpha pulse height analysis and gross alpha counting.

The 1/4- x 12-in. spectrographic-grade carbon electrodes were manufactured by the National Carbon Company.

The 99.99% pure bismuth metal is available from Alfa Inorganics, Beverly, Mass. For the tracer studies, the metal was melted and cast in 10-ml "Pyrex"* beakers to give wafers weighing 35 to 40 g.

Equipment

The equipment for the tracer studies is described in this section. The equipment developed for remote operation is described in Appendix A.

Two furnaces (Heavy Duty Heating Equipment Co. Type 82) were used: one to preheat the electrode assembly and one to dehydrate and melt the salt. Each furnace was equipped with a thermocouple and "Pyr-O-Vane"** controller. The furnace, cell, "Vycor"* liner, and cooling flange assemblies are shown in Figure 1. The "Teflon"*** cap, electrodes, and stirring rod are shown in Figure 2.

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* Trademark of Corning Glass Works

** Trademark of Minneapolis Honeywell

*** Trademark of Du Pont

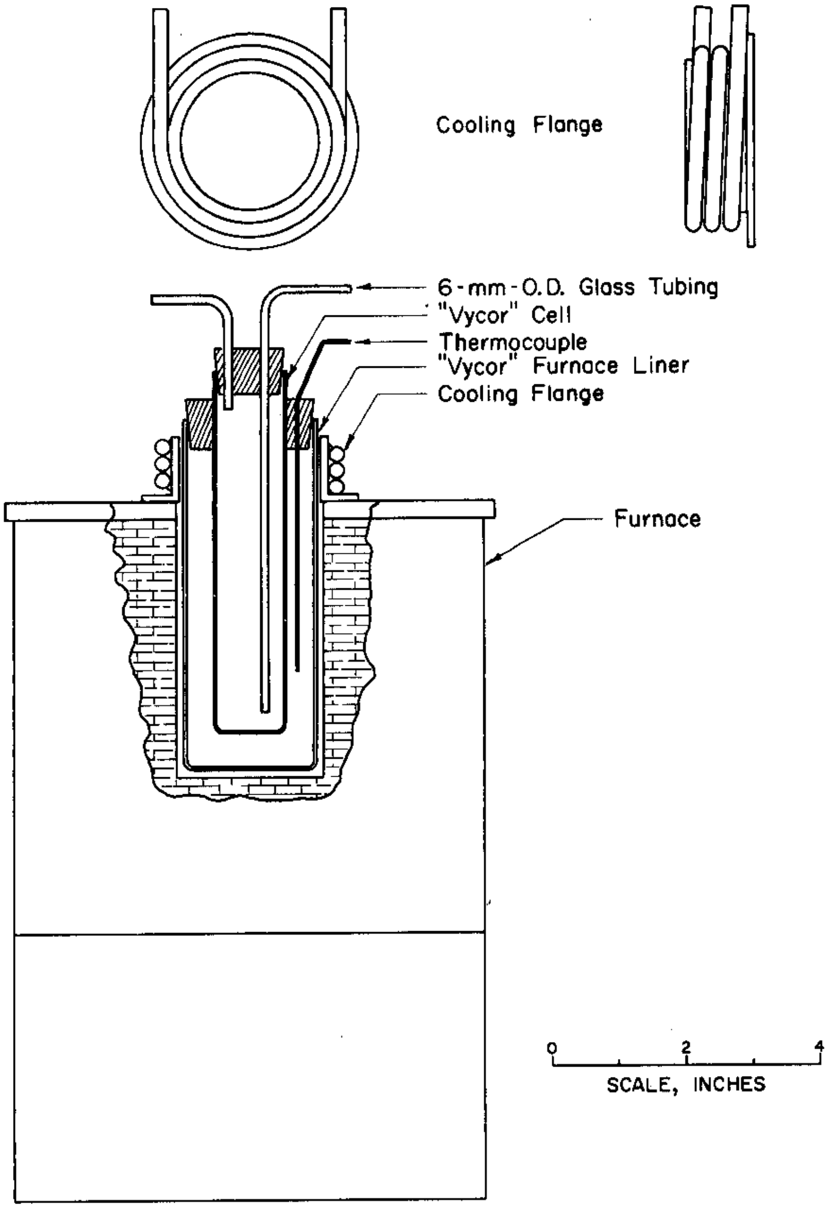
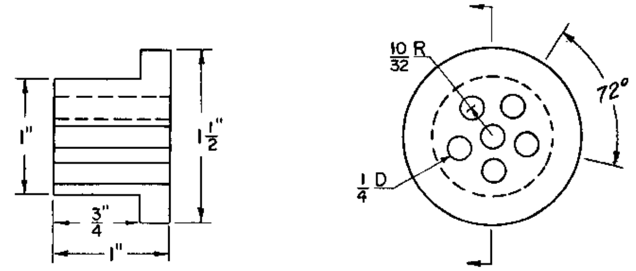
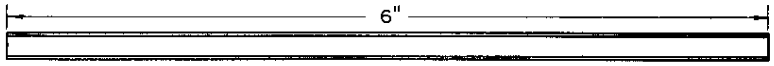


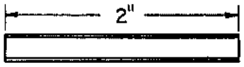
FIG. 1 FURNACE AND COOLING FLANGE ASSEMBLY



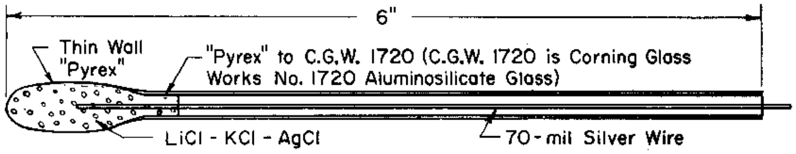
"TEFLON" CELL CAP



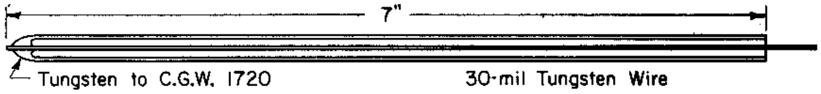
SPARGE TUBE (6-MM C.G.W. No. 1720)



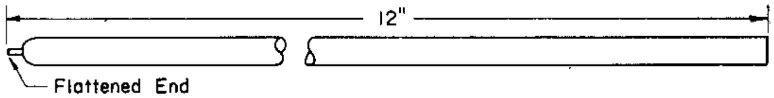
GAS EXIT TUBE – 6-MM "PYREX" ("PYREX" IS A TRADEMARK OF CORNING GLASS WORKS)



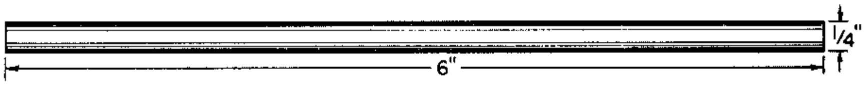
SILVER REFERENCE ELECTRODE – 6-MM DIA.



BISMUTH CONTACT – 6-MM C.G.W. 1720



STIRRER – 6-MM "VYCOR"



CARBON ELECTRODE

FIG. 2 COMPONENTS OF ELECTRODE AND CAP ASSEMBLY

Stirring was provided by a 100-rpm motor (Bodine Electric Co. #B8124E-18B). For the dehydration-melting step, the cell was fitted with a rubber stopper containing two 6-mm glass tubes as shown in Figure 1. For the electrolysis, this closure was replaced by the electrode assembly.

The complete electrolysis assembly including power supply, vacuum-tube voltmeter and gas distribution system is shown in Figure 3. The power supply was a Lambda Electronics Corporation Model LH 124, and the vacuum tube voltmeter was a Senior "Voltohmyst."* A three electrode system was utilized. A constant current was applied between the bismuth cathode and carbon rod anode; the electrolysis potential was measured between the silver reference electrode (SRE) and the bismuth cathode. The electrolysis potential was controlled manually.

Procedure

A detailed remote operation procedure is given in Appendix B; only those details pertinent to the tracer studies are given here. All of the usual techniques except vacuum dehydration were used to minimize the deleterious effects of moisture.

For a typical run, the electrodes, stirring rod, and gas inlet and outlet tubes were assembled in the "Teflon" cap and held in position by rubber "O" rings. The reference electrode had previously been charged in a dry box. The electrode assembly was then inserted into a clean cell and positioned in Furnace 1. A second cell containing a weighed (±0.1 g) Bi wafer, 40.0 g of ($CsCl-LaCl_3$ + 0.5% $ThCl_4$) and 0.500 g of the ^{228}Th tracer was positioned in Furnace 2. This cell was fitted with a rubber closure as shown in Figure 1, and the gas inlet tube was connected to the HCl-helium gas supply. The cell off-gas was vented to the hood exhaust. Both furnaces were turned on, and the limit controls were set for 400°C and 600°C, respectively. While the electrode assembly was preheating, the salts were slowly heated to melting in a copious stream of HCl-helium. When the melt was clear, the rubber stopper was quickly replaced by the preheated electrode assembly. The gas and electrical connections were now changed to conform to Figure 3, and the stirring rod was connected. The solution was then electrolyzed for two hours to deposit the thorium.

The bismuth button and salt were dissolved separately and analyzed for ^{228}Th by a combination of electrodeposition[3] and alpha pulse height analysis with gross alpha counting.

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*Trademark of Radio Corporation of America

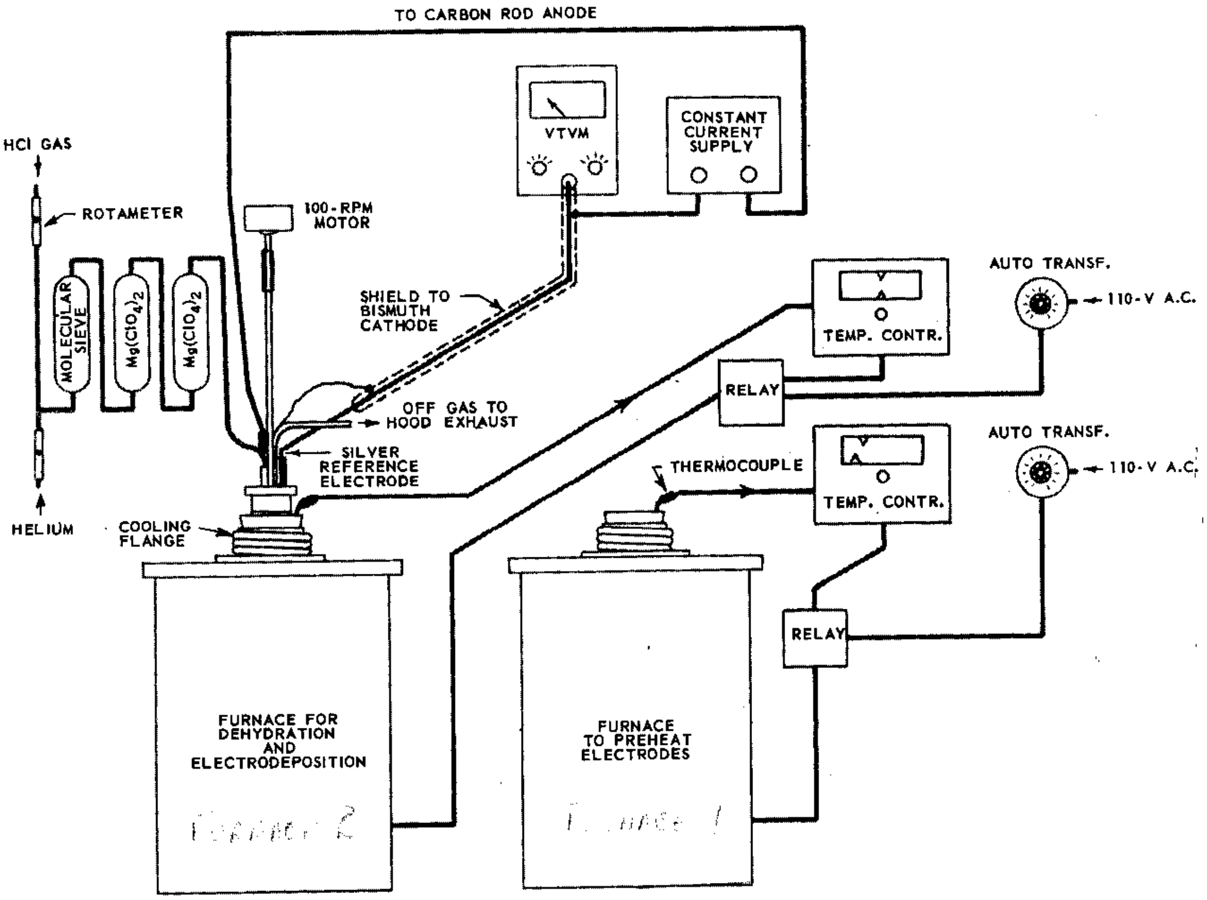

FIG. 3 ELECTRODEPOSITION EQUIPMENT

The procedure for voltammetric studies was the same except that in the case of the tungsten wire electrode, the bismuth wafer was omitted and the bismuth contact served as a tungsten wire electrode. Voltammograms were recorded with a fast-scan polarograph.

RESULTS

Voltammetric Studies

The electrodeposition behavior of thorium at the tungsten wire and bismuth pool electrodes was determined by voltammetry. Current-voltage curves for the deposition-stripping of thorium at the tungsten wire electrode exhibited a deposition wave at -1.0 v and a stripping peak at -0.85 v vs. SRE. No waves for thorium were observed at the bismuth electrode. When the potential of the tungsten wire electrode was scanned into the limiting current region, electrolyte decomposition was observed at -1.3 v vs. SRE, and a stripping peak due to the dissolution of cesium and/or lanthanum appeared at -1.2 v vs. SRE.

These results do not indicate an optimum potential for the electrodeposition of thorium, but the absence of a thorium wave at the bismuth cathode indicated that the deposition potential exceeds -1.0 v, at which considerable cesium and/or lanthanum will probably be codeposited.

^{228}Th Tracer Studies

Because the voltammetric results were inconclusive, the deposition of ^{228}Th from molten $CsCl-LaCl_3$ eutectic into liquid bismuth was studied as a function of potential, temperature, and sparge gas. Eight depositions were made, and the yields were computed from the amount of ^{228}Th tracer remaining in the salt upon completion of the electrolysis.

A working range from -1.0 to -1.5 v vs. SRE was selected for study. Deposition yields were 60 to 75% between -1.0 and -1.2 v. The one run at -1.5 v gave a yield of 95%; however, at this potential, considerable codeposition of lanthanum and/or cesium is expected. The optimum electrolysis potential is probably $\sim$ -1.2 v.

Studies of the effect of temperature on the electrolysis were confined to visual estimates of the amount of deposit that formed between the bismuth cathode and the glass cell. Excessive temperature appeared to aggravate this condition.

The nature of the sparge gas had no apparent effect on the electrolysis current for a given titration, and helium and HCl could be used interchangeably. However, a cloudy melt indicating the presence of moisture always produced excessive electrolysis currents, increased deposits at the metal-glass interface, and caused excessive pitting of the glass. Cloudy melts could be cleared by extensive purging with HCl-helium. Melts purged with the mixed gas cleared more rapidly than those purged with HCl alone; melts purged with helium only did not clear. Depositions from clear melts produced bright bismuth wafers with a minimum deposit at the metal-glass interface.

The results of the tracer studies are summarized in Table I. Run 9 was an unsuccessful attempt to rinse the ^{228}Th from the shipping container as received (a platinum crucible) by the melt; this would have eliminated a dissolution step in the remote facility.

TABLE I

Summary of ^{228}Th Tracer Studies

Run	Temp, °C	Sparge Gas	Electrolysis Potential, volts	Deposition Efficiency, %	Remarks
1	650	HCl	-1.2	43.6	Electrolysis terminated before 2-hr limit
2	650	HCl	-1.2	76	Negligible activity at bismuth-glass interface(a)
3	700	HCl	-1.5	97	Remelted wafer with Cd and Sn. Activity follows dross.(a)
4	680	HCl	-1.2	(b)	Wafer <10 ppm Cl^- by activation analysis
5	620	HCl	-1.1	62	
6	620	HCl	-1.0	89	(a)(c)
7	620	HCl	-1.2	96	(a)(d)
8	620	He	-1.2	74	Wafer analyzed by spark source mass spectrometer.
9	700	He	-1.2	32	(e)

(a) Surface of bismuth wafer mottled.
(b) No ^{228}Th tracer added.
(c) Additional thorium added to cell after 1.5 hr to scavenge the activity. Total electrolysis time: ~3 hr.
(d) Melt cloudy, black deposit at bismuth-glass interface.
(e) 4.5 g platinum added to cell charge. On melting, the platinum dissolved in the salt and caused salt to solidify.

Table II gives the mass spectrometric results for the wafer from Run 8 versus the starting material. These results show that significant amounts of cesium and lanthanum are deposited at -1.2 v vs. SRE. The high concentration of chlorine in the bismuth was surprising in view of the activation analysis result for Run 4. However, the estimated neutron background for a bismuth wafer containing 2 x 10^3 ppm Cl (atomic) was only 2% that for an evaporated ^{228}Th source (as the nitrate) of 5.5 x 10^4 n/(sec)(Ci).

TABLE II

Major Impurities in Bismuth Metal Before and After Electrodeposition

Impurity	Concentration, ppm atomic Before	 After
La	10	5 x 10^4
Cs	10^3	2 x 10^4
Cu	10^2	7
Ni	10^3	20
Ti	40	23
Cl	1.3 x 10^2	2 x 10^3
Si	2.1 x 10^2	3.5 x 10^2
B	10	20

Full-Scale Electrodeposition

A 4.3-curie source was prepared in equipment described in Appendix A. The deposition efficiency for the process as determined calorimetrically[4] was 71.5%, and the neutron background as determined by a manganese bath[5] was 3.3 x 10^3 n/(sec)(Ci). This represents a 96% reduction in the neutron background over a corresponding dry salt source. A preliminary evaluation of the source indicates that it can be used to determine deuterium to ±0.001% at the 0.015% level. The complete electrodeposition procedure is given in Appendix B.

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3. M. Y. Donnan and E. K. Dukes. "Carrier Technique for Quantitative Electrodeposition of Actinides." Anal. Chem. 36, 392 (1964).

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APPENDIX A

REMOTE ELECTRODEPOSITION EQUIPMENT

The equipment described in this section is designed for remote operation in High Level Caves* with master slave manipulators. The ^{228}Th is leached from its shipping container into the electrolysis cell by a Soxhlet extraction with HCl. After the evaporation, the extractor is removed and replaced by the electrode assembly. The ^{228}Th is dehydrated, the eutectic salt and the bismuth metal added, and the ^{228}Th electrodeposited into a liquid bismuth cathode. The electrical equipment for this deposition was that used in the tracer studies. The equipment consists of: (1) a gas inlet system, (2) furnace assembly, (3) crucible leacher, (4) electrolysis cap, and (5) an off-gas scrubber. All connections, except electrical, are made via polyethylene § 12/5 spherical joints.

Gas Inlet System

The gas inlet system is assembled from commercially available components and polyethylene tubing. A schematic diagram of this system is shown in Figure A-1. This equipment is located outside the shielded cell in the operating area, and connections to the equipment in the cell are made through service plugs. The unit is designed so that the individual gas flow rates are monitored on separate rotameters and then mixed before entering the drying towers. This arrangement permits the magnesium perchlorate towers to be flushed with argon at the completion of the run. (The pressure relief valve provides a safe exit for the HCl if a tower becomes plugged.) The direct argon line provides a copious flow of argon to speed the evaporation and to assure a positive pressure on the cell during the electrolysis.

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* See A. L. Coogler, G. J. Deily, and R. J. Hale. _Evolution of the High Level Caves at the Savannah River Laboratory_. Proceedings of the 13th Conference on Remote Systems Technology, American Nuclear Society, Hinsdale, Illinois (1965).

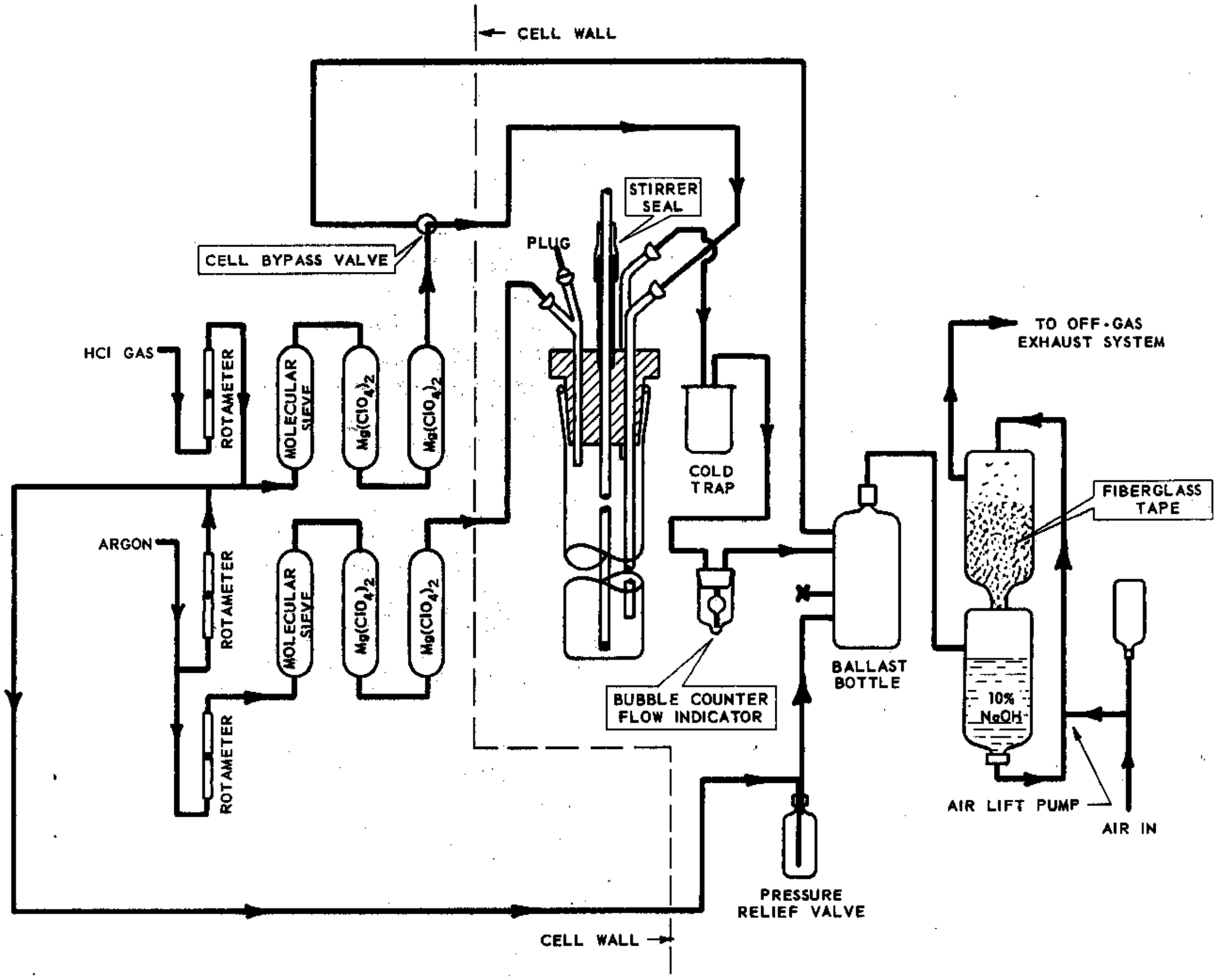


FIG. A-1 GAS INLET SYSTEM EQUIPMENT CONNECTED FOR DEHYDRATION

Why [illegible]
relief
systems?

Furnace Assembly

The furnace assembly is shown in Figure A-2. The furnace, a Hoskins Mfg. Co. Type FH 305 (large tube), is modified for vertical operation and then mounted on a stand by means of pulleys and counterweights. The counterweights are adjusted so that a slight pressure will raise or lower the furnace.

The cooling flange for the electrolysis cell is affixed to the stand by sliding clamps. Thus, the electrolysis cell and cooling flange can also be raised and lowered. The stand ensures accurate alignment between the electrolysis cell and the leacher and electrolysis heads when the racks on which these components are mounted are locked in position on the furnace stand. The guides were necessary to ensure smooth operation of the furnace. The furnace liner protects the furnace in the event that the electrolysis cell is broken. The furnace controls consist of a variable transformer in series with a "Pyr-O-Vane" furnace temperature control. The variable transformer serves to limit the heating rate (power input to the furnace). These controls (with the exception of the thermocouple) are located in the operating area.

Crucible Leacher

The crucible leacher is shown in Figure A-3. This device is designed as a Soxhlet extractor with the electrolysis cell as the boiler. This unit is mounted on a rack designed to lock onto the furnace stand.

The crucible holder subassembly fits into the leacher and consists of a cold-finger, condensate return tube and crucible holder fabricated as a unit. The crucible holder slides freely on the condensate return tube. Thus, when the subassembly is lifted out of the leacher, the crucible can be positioned in the holder; and as the subassembly is lowered into the leacher, the condensate return tube is automatically positioned inside the crucible. As a result, during the extraction, the crucible is continuously flushed with fresh solvent.

What is all this? Compare descriptions with figures.

A "Teflon" sleeve (Kontes Glass Co., Vineland, N. J.) over the ₮ 29/42 inner joint of the leacher prevents this joint from freezing together. Before use, the leacher is heavily insulated.

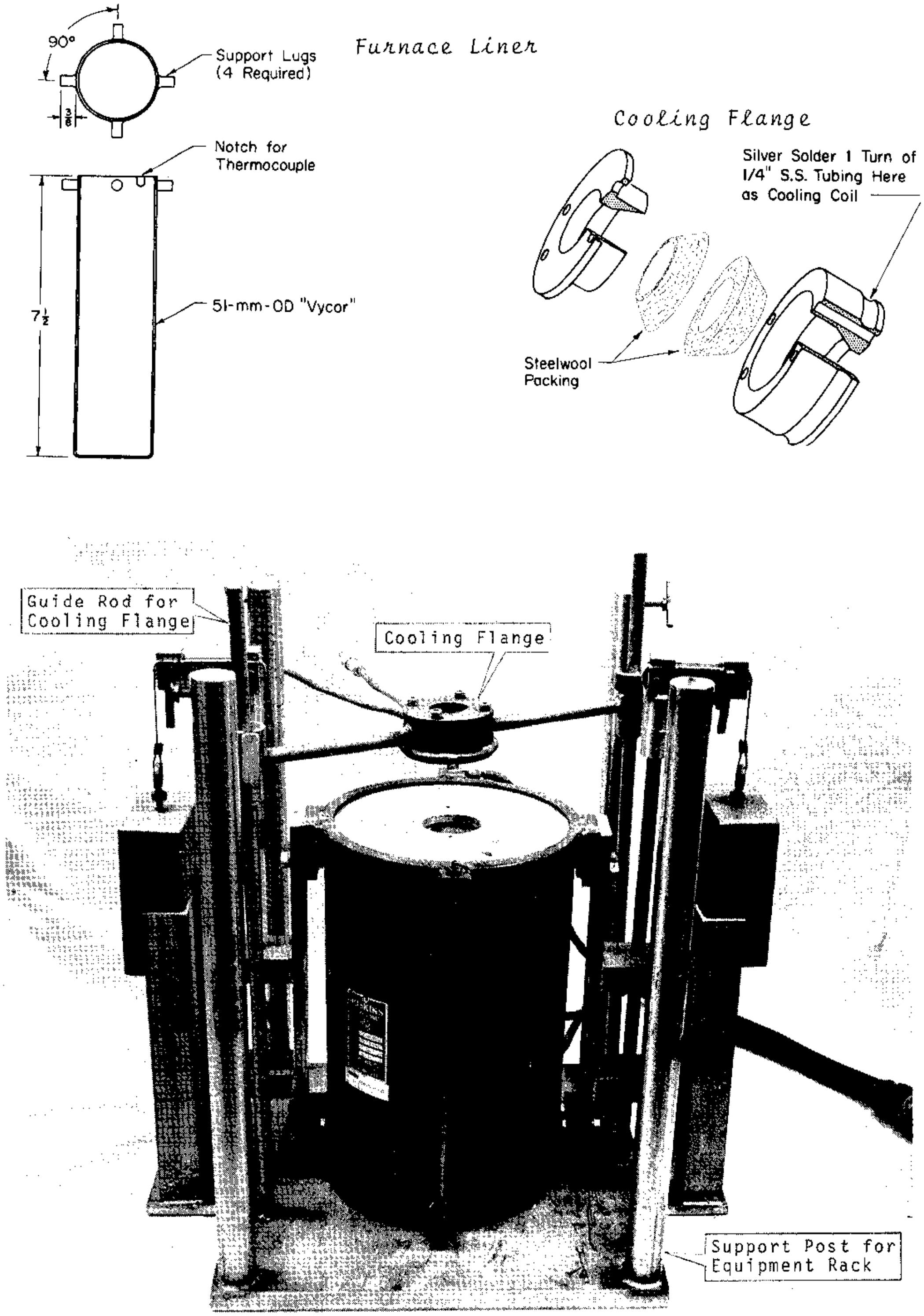


FIG. A-2 FURNACE ASSEMBLY

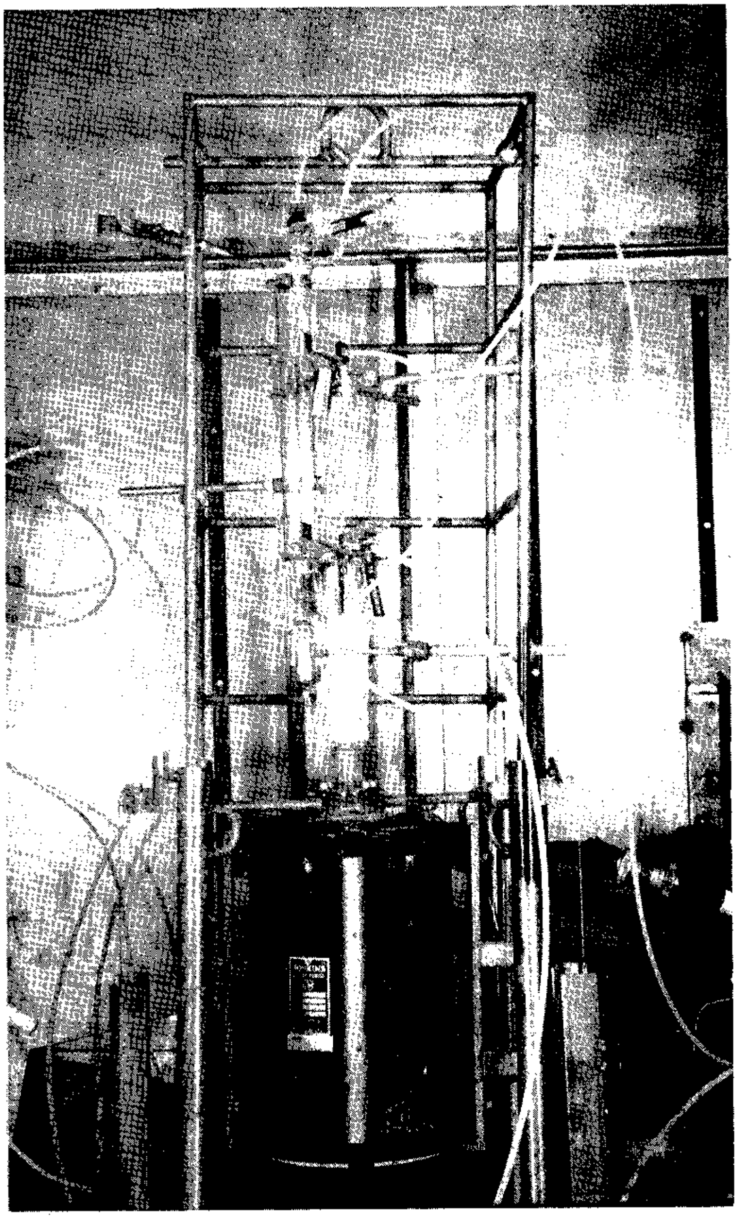
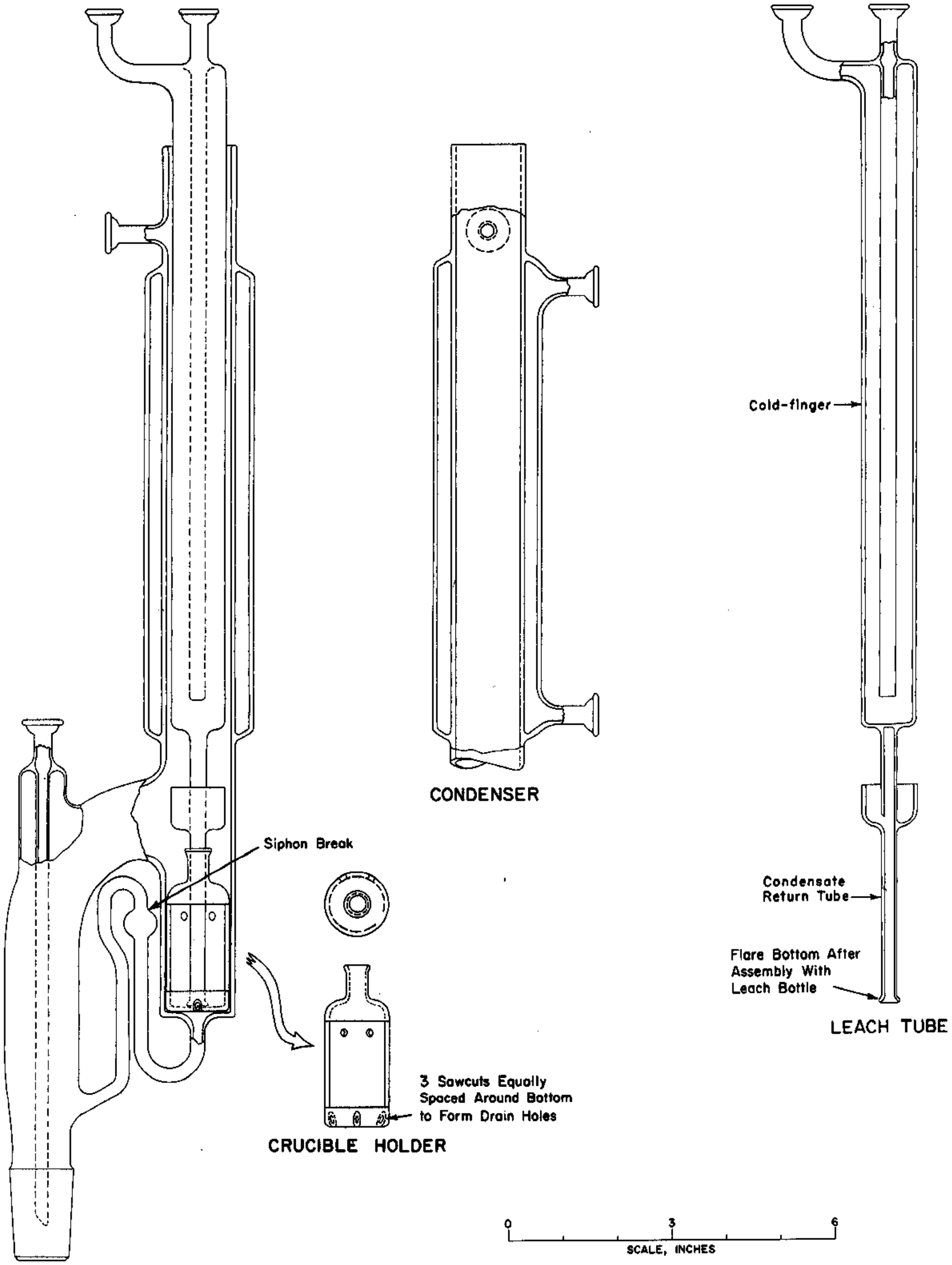


FIG. A-3 LEACHING HEAD ASSEMBLY
Leacher Mounted on Furnace



Leacher Components

Electrolysis Head

The electrolysis head is also mounted on a rack. This unit consists of a 100-rpm motor (Bodine Electric Co. #B 8124E-18B), stirring rod, cell cap and stirrer seal, electrodes, gas inlet, outlet and sparge tubes, and the loading head subassembly. This subassembly is used to charge the eutectic and bismuth to the cell via the cell cap. Figure A-4 shows this complete assembly.

A length of 1/4-inch-ID plastic tubing, stiffened by inserting a shorter length of 1/4-inch-OD stainless steel rod serves as the flexible connection between the motor and stirring rod. The stirrer enters the cell through a stainless steel tube. A short length of greased rubber tubing serves as a seal between the stainless steel tubing and the stirrer shaft.

The cell cap and electrode assembly are shown in Figure A-5. The cap is machined from fiberglass-filled epoxy bar stock. To ensure a gastight fit, the electrodes and other components are sealed into the cap with epoxy resin.

In preparation for charging the silver electrode, the cap assembly is placed in the dry box, a 12-inch length of 70-mil silver wire is placed in the electrode, and sufficient LiCl-KCl + 8.75 wt % AgCl is added to fill the electrode to a depth of 1.5 inches. The top of the electrode is now sealed with "Silastic"* 731 RTV, and the completed cell cap assembly is installed on the rack.

The loading head and transfer rod are shown in Figure A-6. The transfer rod is designed to assist in the transfer of solids from the loading head to the electrolysis cell. The transfer rod is attached to the loading head by means of a vinyl plastic sleeve. The loading head is attached to the rack by a bracket to position accurately the solids loading assembly on the rack with mechanical manipulators. This feature is necessary because the loading head is kept in the dry box until minutes before use.

The gas scrubber system (Figure A-7) absorbs corrosive HCl vapors in 10% NaOH solution. Except for the cold trap insert and bubble counter, the device is fabricated from commercial polyethylene components.

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* Trademark of Dow Corning for their silicone resin.

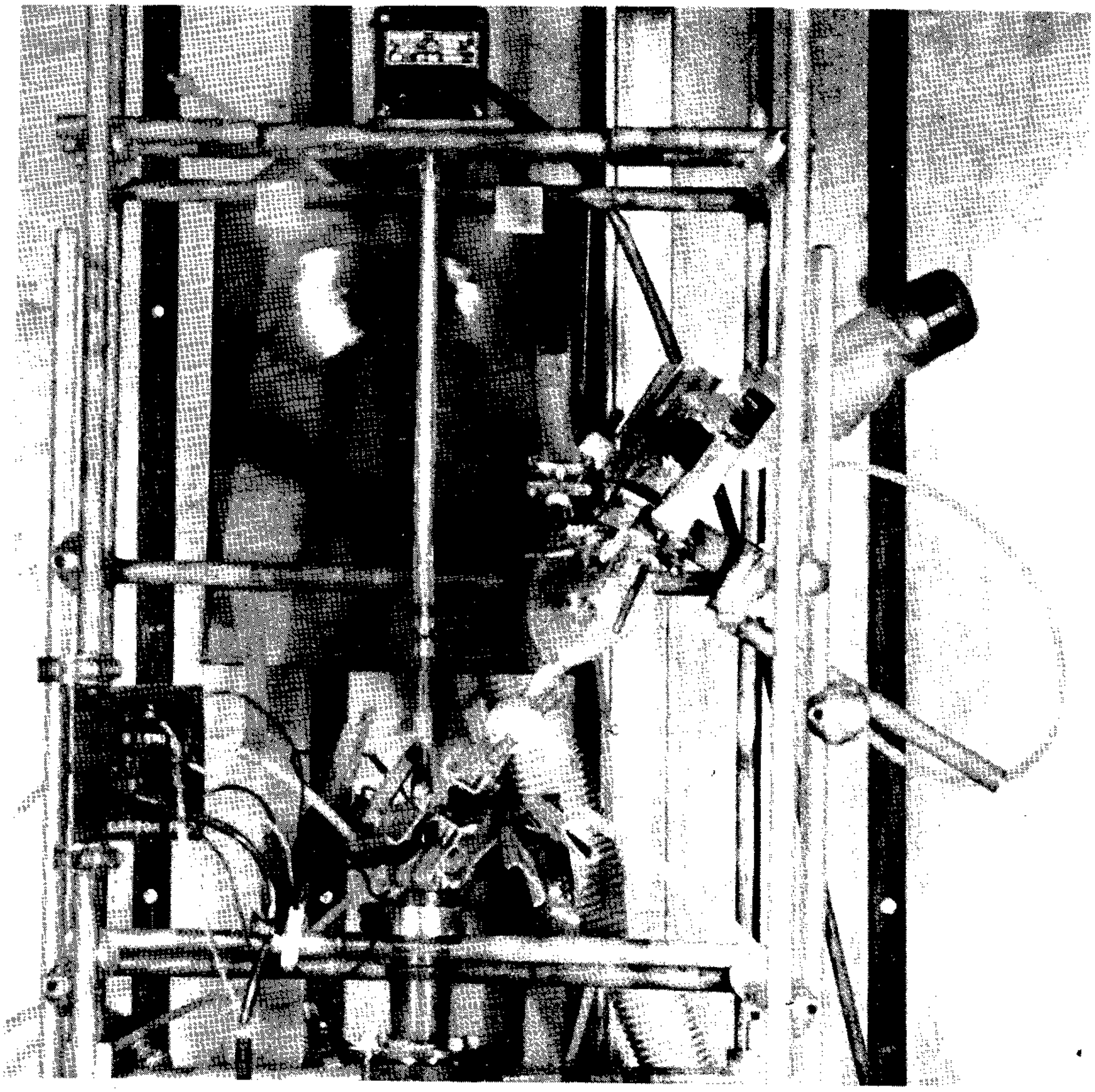


FIG. A-4 ELECTROLYSIS HEAD ASSEMBLY

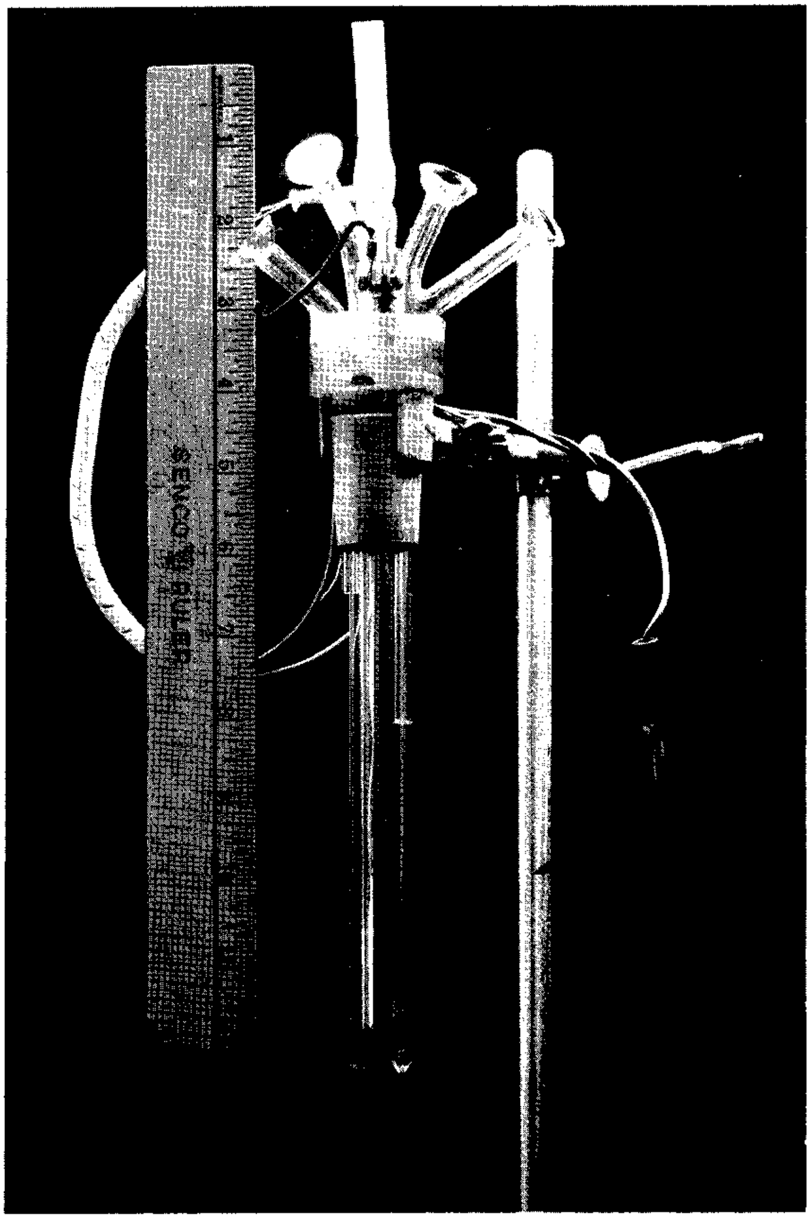
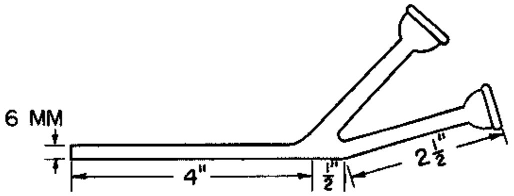
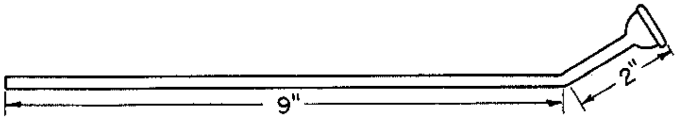


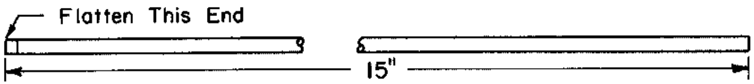
FIG. A-5 CELL CAP AND ELECTRODE ASSEMBLY



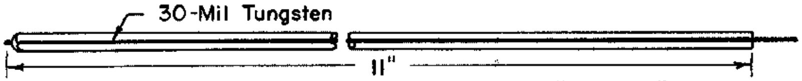
ADDITION TUBE (BOROSILICATE)



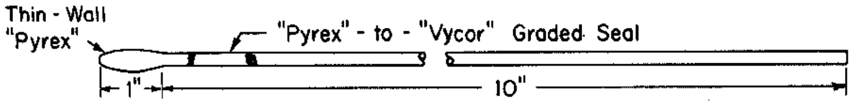
SPARGE TUBE - 6 MM "VYCOR"



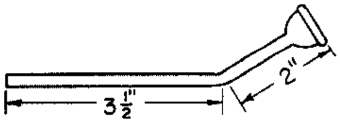
STIRRER — 6-MM "VYCOR"



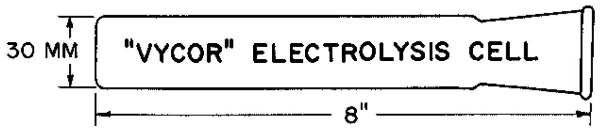
BISMUTH CONTACT — 6-MM-OD "VYCOR"



SILVER ELECTRODE BODY — 6-MM-OD "VYCOR"



GAS EXIT TUBE — 6-MM-OD "PYREX"



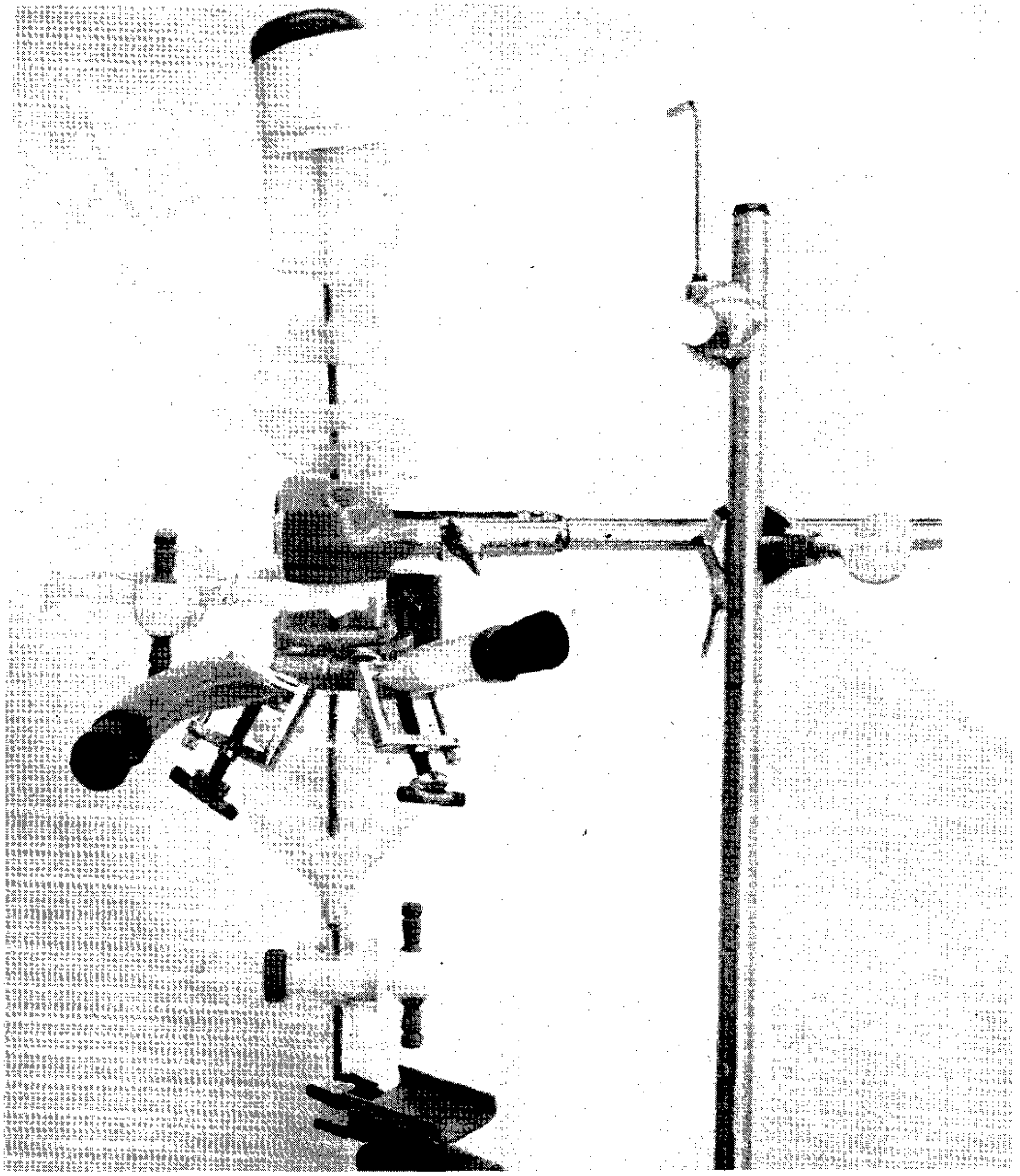
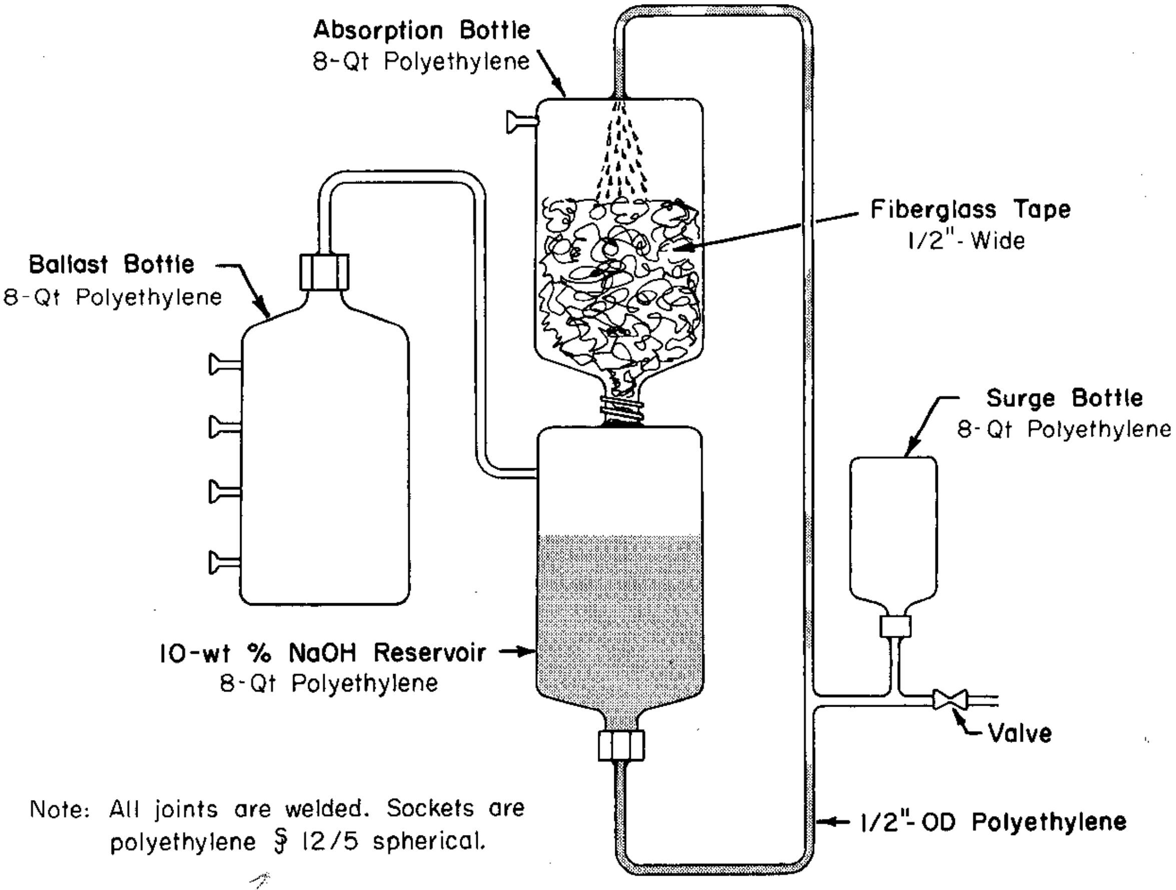


FIG. A-6 COMPLETED SOLIDS LOADING HEAD SUBASSEMBLY



Note: All joints are welded. Sockets are polyethylene § 12/5 spherical.

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FIG. A-7 GAS SCRUBBER

Adjustments

After assembly, the apparatus is adjusted to ensure accurate alignment between the electrolysis cell and the leaching and electrolysis assemblies: The cell is installed in the cooling flange. Steel wool is used as the packing in this operation to provide good thermal contact between the cell and the cooling flange. The leaching head is locked in place on the furnace, and the leacher aligned to fit the cell. The leaching head is then removed and replaced by the electrolysis head. The electrode cap assembly is aligned to fit the cell, and the loading head subassembly is aligned with the addition tube. The final operation is to install a gum rubber sleeve over the stainless steel tube of the stirrer cap to form the stirrer seal. The tubing is lubricated with silicone grease, the stirrer inserted and aligned with the motor. This completes the assembly. After a final check, the furnace assembly, leaching head, and electrolysis head are transferred to the shielded cell in preparation for the electrodeposition.

APPENDIX B

REMOTE ELECTRODEPOSITION PROCEDURE

The remote electrodeposition procedure consists of the following steps: (1) charging the loading head subassembly (a dry box operation), (2) leaching to transfer the ^{228}Th to the electrolysis cell, (3) evaporation and dehydration of the ^{228}Th, and (4) electrodeposition of ^{228}Th into a liquid bismuth cathode. The details of each step are listed sequentially below.

Solids Loading Subassembly - Charging

1. Place a pinch clamp (modified for cave operation) on each of two 5-inch lengths of 1/2-inch-ID plastic tubing and stopper the open ends. Allow enough room between the end of the tube and clamp to attach tube to the loading head.
2. Place tubing in dry box with the loading head. Open stopcocks and remove stoppers from plastic tubes.
3. Wait 2 hours for system to desiccate.
4. Weigh 45 ±1 g of ($CsCl-LaCl_3$ + 0.5% $ThCl_4$) in dry box.
5. Grind to a fine powder. Avoid lumps that will not pass through stopcock of loading head; sieve through 100-200 mesh screen if necessary.
6. Insert the transfer rod through the bore of the stopcock of the loading head until the bead seats against the bore.
7. Turn the stopcock to hold the rod in place.
8. Place a socket cap over the ball joint at end of loading head and clamp in place. The cap prevents moisture from entering the loading assembly while in transit to the cave.
9. Charge the ($CsCl-LaCl_3$ + 0.5% $ThCl_4$) powder to the main compartment of the loading head via a side arm.
10. Crush (but do not pulverize) about 50 g of bismuth. Screen and collect 20-60 mesh material.
11. Weigh 35 ±1 g of the 20-60 mesh bismuth and charge to one of the plastic tubes.
12. Stopper the tube and attach the clamp end to one side arm of the loading head.

13. Grind 1.5 g of the CsCl-$LaCl_3$ + 10% $ThCl_4$ to a fine powder (100-200 mesh), and charge to remaining plastic tube.

14. Stopper the open end and attach this tube to the loading head as in Step 12.

15. Close the remaining stopcock.

16. The subassembly is stored in the dry box until needed.

Leaching Step - Cave Operation

1. Lower furnace and cooling flange.

2. Install leaching head assembly on furnace.

3. Carefully raise cooling flange so that electrolysis cell is connected to leacher. Inspect this joint for good seal, and make certain the "Teflon" sleeve is in place.

4. Remove cover from crucible containing ^{228}Th.

5. Remove crucible holder from leacher and place crucible containing ^{228}Th in crucible holder.

6. Replace crucible holder assembly in leacher. See that condensate return tube is positioned inside the crucible.

7. Connect the equipment as shown in Figure B-1.

8. Turn on the cooling water to the condenser and cold-finger.

9. With a plastic squeeze bottle, add 75 ml of 6M HCl containing 1 mg/ml $ThCl_4$ to the top of the leaching head. Syphon action should begin as the last of the HCl solution is added.

10. Set the furnace temperature control to 850°F.

11. Starting at 30 volts, slowly increase power input to furnace until solution boils.

12. Continue to raise power input to furnace until syphon action occurs every 30-45 minutes.

13. Lower furnace occasionally during leaching operation and check liquid level in cell at the instant syphon action occurs. Add more 6M HCl if less than 1 inch.

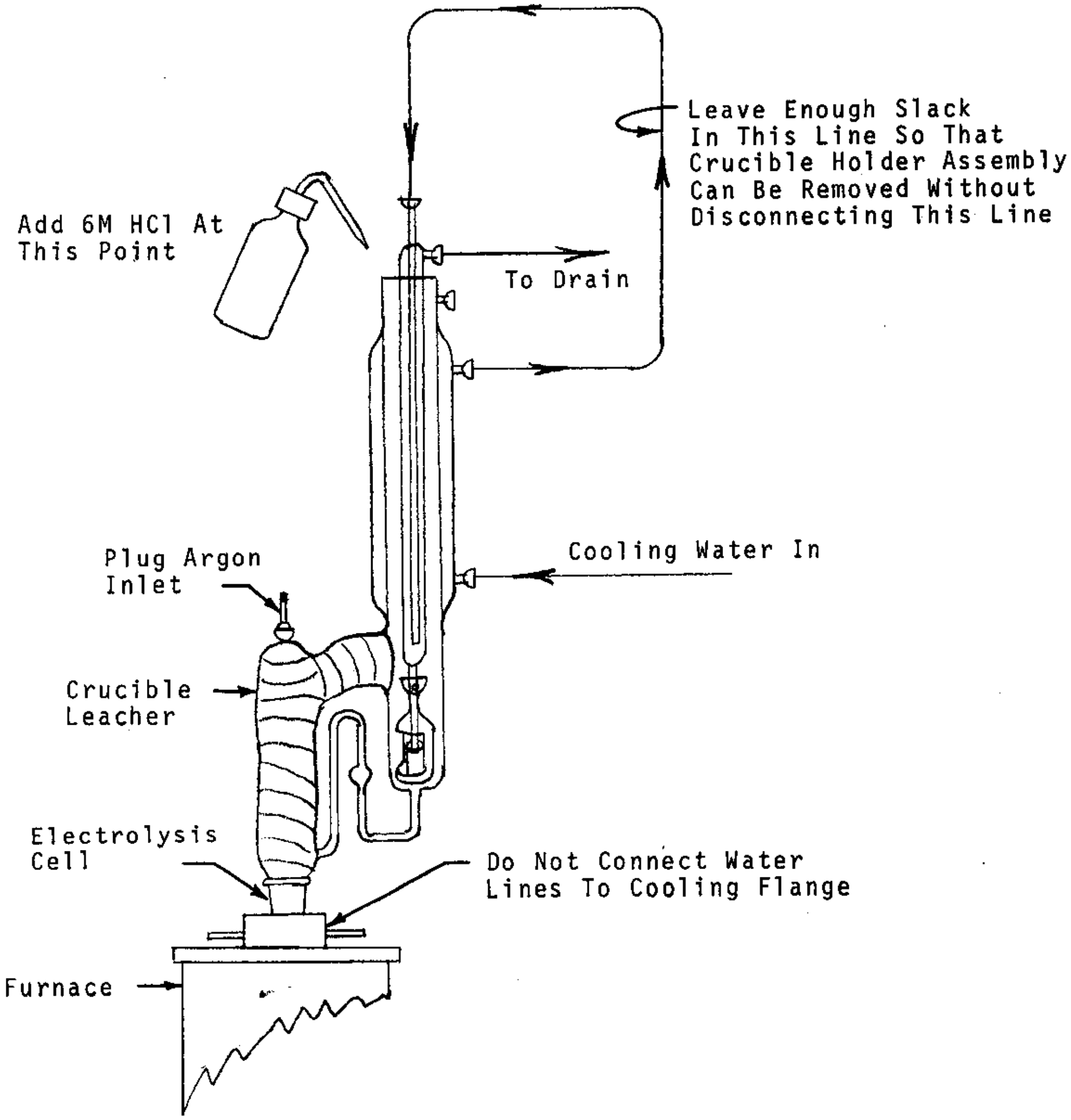

FIG. B-1 EQUIPMENT CONNECTIONS FOR LEACHING OPERATION

14. Leach crucible for a minimum of 12 cycles.

15. When leaching is complete, raise crucible holder assembly and remove crucible. (Do not spill contents.)

16. Pour solution from crucible into leacher.

17. Replace crucible holder assembly in leacher.

18. Continue heating until solution is syphoned into cell.

19. Immediately lower furnace and turn off furnace power. [This completes the leaching operation. The ^{228}Th is now in solution in the electrolysis cell.]

20. Turn off cooling water and drain condenser and cold finger.

21. Remove crucible holder from the leacher.

Evaporation and Dehydration - Cave Operation

1. Change the connections to the leacher as shown in Figure B-2.

2. Connect argon direct line to the insulated side arm.

3. Insert a #7 rubber stopper into top of leaching head.

4. Connect off-gas vent to ballast bottle.

5. Turn on scrubber air lift pump.

6. Check connections to ballast bottle. Plug unused ports.

7. Open argon direct valve. Adjust for a copious flow of argon.

8. Set furnace temperature control for 400°F, and variable transformer to 70 v. Note: The solution does not boil. Vapors are swept from the cell with dry argon gas.

9. As the solution evaporates, moisture will condense in the extraction side of the leacher. Use a heat gun to evaporate this condensate.

10. Continue the evaporation, lowering the furnace periodically to check the progress of the evaporation.

11. Evaporate the solution until it thickens. <u>Do Not Bake Salts</u>.

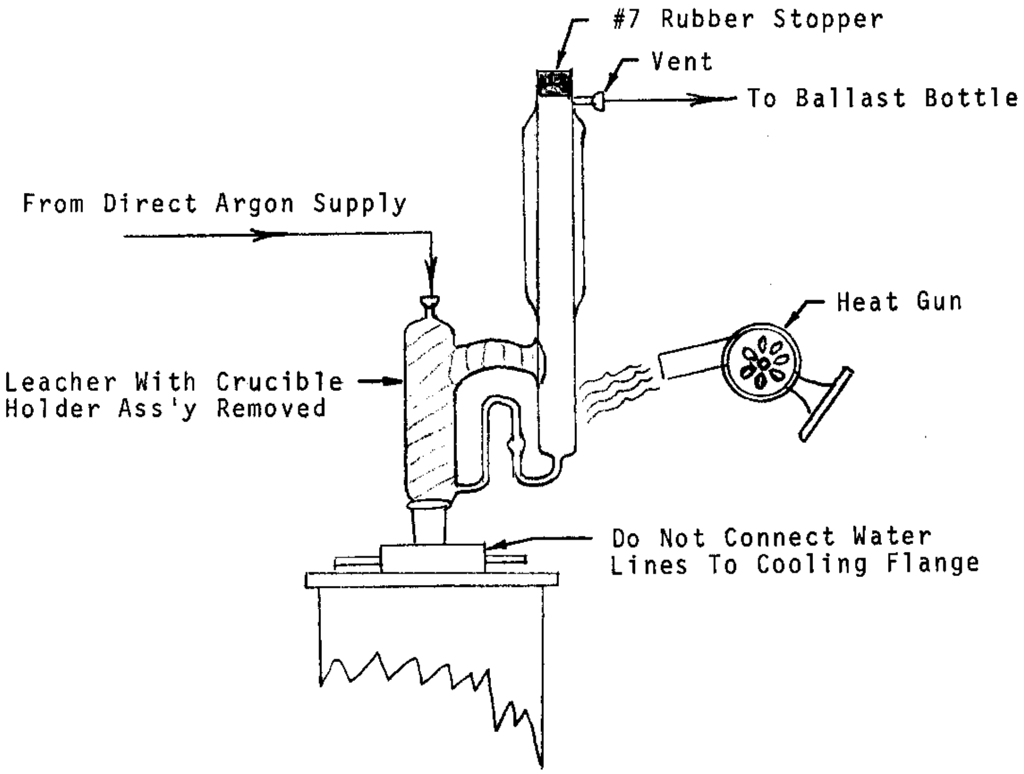

FIG. B-2 EQUIPMENT CONNECTIONS FOR EVAPORATION

12. When the salts reach a thickened dry condition, immediately lower furnace and shut off furnace power.
13. Turn off argon and air to scrubber lift pump.
14. Disconnect lines to the leaching head, and remove leaching head from furnace.
15. Insert a #6 rubber stopper into top of the electrolysis cell.
16. Lower the cooling flange in preparation for the installation of the electrolysis head.
17. Remove the stopper from the electrolysis cell.
18. Install electrolysis head on furnace.
19. Raise cooling flange to attach the electrolysis cell to the electrode assembly. Inspect the joint for a good seal, and make certain the "Teflon" sleeve is in place.

20. Make connections to the electrolysis head as shown in Figure A-1.

21. Connect the argon direct line to the lower inlet of the addition tube and insert a plug to blank of the upper inlet.

22. Connect the argon-HCl gas line to the sparge tube.

23. Connect the exit gas tube to the cold trap.

24. Connect the cold trap to the flow indicator and the flow indicator to the ballast bottle.

25. Plug all unused ports at ballast bottle.

26. Fill cold trap with a "Triclene"* dry ice slurry.

27. Raise furnace.

28. Turn on air to scrubber lift pump.

29. Turn on direct argon; adjust to give a copious flow.

30. Set furnace temperature control to 250°F, and variable transformer to 30 v.

31. Turn on water supply to cooling flange.

32. Turn on HCl supply and adjust flow to 50-100 ml/min.

33. Periodically lower furnace and inspect salts, continue dehydration until salts are dry as ascertained by visual inspection.

34. Proceed immediately with the electrodeposition.

Electrodeposition - Cave Operation

1. Remove the solids loading subassembly from the dry box and transfer immediately to the cave.

2. Turn off HCl.

3. Reduce direct argon flow to about 100 ml/min.

4. Remove blanking plug from upper inlet of addition tube.

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* Trademark of Du Pont

5. Remove plug from bottom of solids loading subassembly; immediately install this assembly in the electrolysis rack.

6. Connect a line from the upper stopcock of the solids loading subassembly to the ballast bottle.

7. Adjust the direct argon valve to give a copious flow of argon.

8. Turn on HCl gas and adjust HCl-argon flow to give about a 1:4 ratio at about 100 ml/min.

9. The connections to the electrolysis cell should now agree with those of Figure B-3.

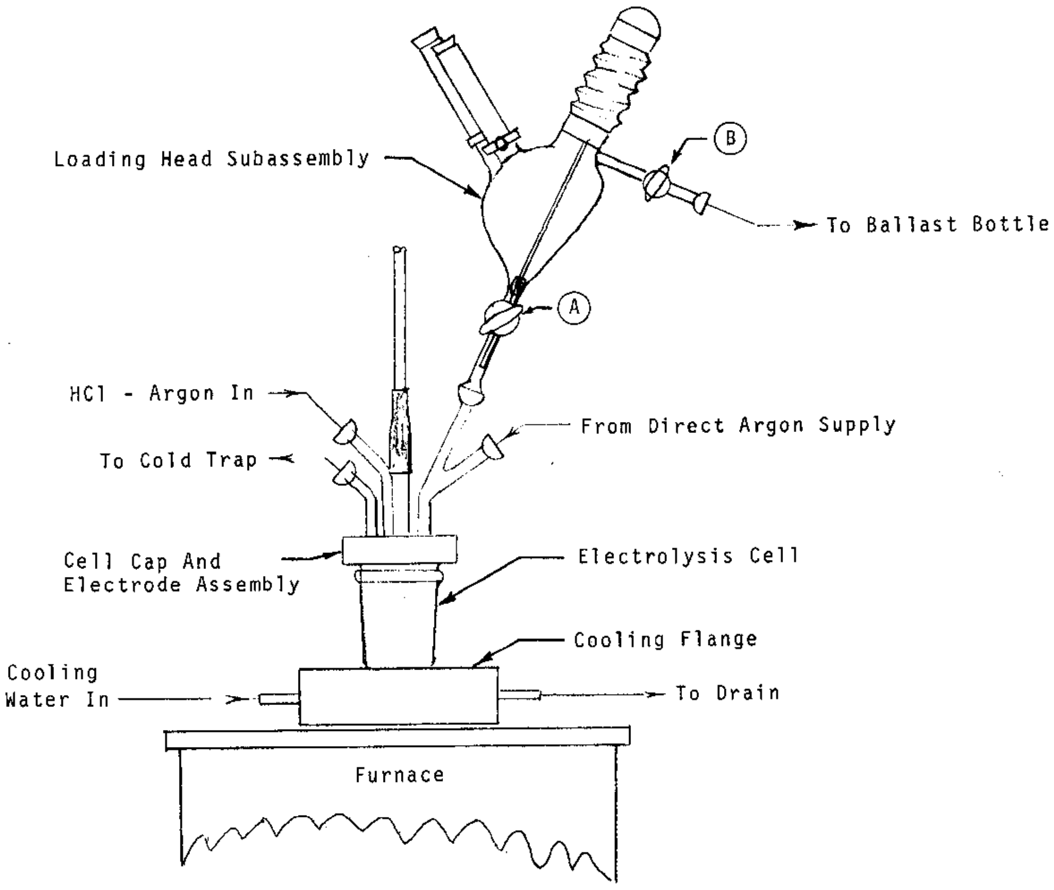

FIG. B-3 EQUIPMENT CONNECTIONS FOR ELECTROLYSIS

10. After 30 minutes, inspect the exit tube and line for evidences of moisture. Continue with the dehydration until the exit line is free of moisture.

11. Open Stopcock B, Figure B-3. Wait 10 minutes.

12. Turn off furnace power, and lower furnace so that the addition of salts can be observed.

13. Close Stopcock B.

14. Open Stopcock A releasing transfer rod.

15. By gentle agitation with the rod, transfer the salts (in the main compartment of the loading subassembly) to the electrolysis head.

16. When the transfer is complete, position the bead on the transfer rod to seal the bore of the stopcock, and turn Stopcock A so that the rod is held in this position.

17. Raise the furnace.

18. Check that the cooling water supply to the cooling flange is on.

19. Set the variable transformer to 70 v.

20. When the furnace temperature reaches the preset 250°F, wait 30 minutes.

21. Now increase the furnace temperature stepwise by 50°F every 15 minutes until 900°F is reached. At this point, check the exit line for signs of moisture. Increase the flow of direct argon if necessary to clear the exit line. Be certain that the exit line is clear before proceeding.

22. When the furnace reaches 900°F, continue as in Step 20 except periodically lower furnace and check for signs of melting.

23. When salt melts, adjust sparge gas to prevent spattering.

24. Salt should be molten before 1250°F is reached. Hold the furnace at 1250°F, and continue sparging until solution clears.

25. Check the electrode connections in preparation for the electrolysis.

26. Turn off HCl, continue sparging with argon.

27. Open pinch clamp on plastic tube containing bismuth granules. This will admit the bismuth to the main compartment of the solids loading assembly.

28. Open Stopcock A and slowly transfer the bismuth to the electrolysis cell.

29. When the transfer is complete, insert the transfer rod into the stopcock to seal the bore, and turn the stopcock to hold the rod in this position.

30. Start stirrer and check operation.

31. Turn on HCl and adjust HCl-argon flow to give good sparging without spattering. The HCl-argon is not critical. A 1:4 ratio is satisfactory.

32. Immediately electrolyze the solution at -1.2 v vs. the silver reference electrode (SRE).

33. Continue the electrolysis for 2 hours.

34. Open the pinch clamp on the "Tygon" tube containing the $CsCl-LaCl_3$ + 10 wt % $ThCl_4$, and transfer the salt to the main compartment of the solids loading subassembly.

35. Open Stopcock A and transfer this salt to the electrolysis cell.

36. Remove the transfer rod from the bore and close Stopcock A.

37. Continue the electrolysis for an additional 2 hours to scavenge the solution of ^{228}Th.

38. The electrolysis step is now complete. Divert the sparge gas to the ballast bottle via the bypass, and turn off the HCl supply.

39. Turn off the furnace power.

40. Immediately remove electrolysis rack from the furnace.

41. Lower furnace and insert a #6 rubber stopper into top of electrolysis cell.

42. Turn off all services.

43. When the salt cools, break the cell; separate the bismuth wafer containing the ^{228}Th.

44. Clean surface of bismuth wafer by gentle abrasion with a wire brush.

45. Transfer wafer to a gastight container.