~~SECRET~~

664121

DP-1058, Del.

AEC RESEARCH AND DEVELOPMENT REPORT

EXTRACTION OF TRITIUM FROM LITHIUM ALUMINATE TARGETS

A. A. KISHBAUGH

CLASSIFICATION CANCELLED with Deletions
BY AUTHORITY OF
Bram C. Feldman, USERDA Division of Classification
NAME — TITLE — DATE
Jan 10, 1975 - PLM 5-1-75

DU PONT
REG. U.S. PAT. OFF.

Savannah River Laboratory

Aiken, South Carolina

[redacted]

~~SECRET~~

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

664121

[redacted]

DP-1058, Del.

Technology, Tritium Production
(M-3679, 45th Ed.)

[redacted]

EXTRACTION OF TRITIUM FROM LITHIUM ALUMINATE TARGETS

by

Albert A. Kishbaugh

SRL FILE COPY

Approved by

D. S. Webster, Research Manager
Separations Engineering Division

August 1966

[redacted]

[redacted]

CLASSIFICATION CANCELLED with Deletions
BY AUTHORITY OF
Bram C. [illegible], USERDA, Division of Classification
NAME TITLE DATE
Jan 10, 1975, RCM 5-1-75

E. I. DU PONT DE NEMOURS & COMPANY
SAVANNAH RIVER LABORATORY
AIKEN, S. C. 29801

CONTRACT AT(07-2)-1 WITH THE
UNITED STATES ATOMIC ENERGY COMMISSION

BY: SPECIAL REREVIEW
CJ Banick AED Class Off
FINAL DETERMINATION
UNCLASSIFIED
Date: 3/27/86

[redacted]

[redacted]

ABSTRACT

Tritium can be extracted satisfactorily from lithium aluminate targets by heating at 850°C in a vacuum for 10 hours; less than 0.1% of the tritium remains in the residue.

CONTENTS

[redacted]

664121

[redacted]

DP-1058, Del.

Technology, Tritium Production
(M-3679, 45th Ed.)

[redacted]

EXTRACTION OF TRITIUM FROM LITHIUM ALUMINATE TARGETS

by

Albert A. Kishbaugh

Approved by

D. S. Webster, Research Manager
Separations Engineering Division

August 1966

[redacted]

CLASSIFICATION CANCELLED with Deletions
BY AUTHORITY OF

Bram C. Feldman, USERDA, Division of Classification
NAME **TITLE** **DATE**
Jan 10, 1975 PGM 5-1-75

E. I. DU PONT DE NEMOURS & COMPANY
SAVANNAH RIVER LABORATORY
AIKEN, S. C. 29801

CONTRACT AT(07-2)-1 WITH THE
UNITED STATES ATOMIC ENERGY COMMISSION

[redacted]

ABSTRACT

Tritium can be extracted satisfactorily from lithium aluminate targets by heating at 850°C in a vacuum for 10 hours; less than 0.1% of the tritium remains in the residue.

CONTENTS

LIST OF TABLES AND FIGURES

[redacted]

EXTRACTION OF TRITIUM FROM LITHIUM ALUMINATE TARGETS

INTRODUCTION

The New Production Reactor (NPR) at Hanford requires tritium-producing targets that will not melt even in the event of accidents that might raise the temperature of the zirconium-clad targets to an estimated 1100°C. Lithium aluminate was chosen as the most suitable target material (mp = 1900°C) after tests of its irradiation behavior.

Pacific Northwest Laboratory (PNL) proposed extraction of tritium from the de-clad lithium aluminate targets by dissolution in molten sodium tetraborate at 850°C, since preliminary data indicated that thermal extraction of the tritium without the use of a flux would give high losses to the residue. The Savannah River Laboratory (SRL) was asked to evaluate the proposed process for use at the Savannah River Plant (SRP) --- PNL does not have facilities for large scale tests with irradiated targets. Experiments with both irradiated and unirradiated targets, with and without flux, were conducted at SRL to obtain sufficient information to estimate the cost of modifying existing tritium separations equipment for extraction of the NPR ceramic targets. The results of these tests are presented in this report.

SUMMARY

Experiments conducted at SRL to evaluate a process for extracting tritium from lithium aluminate targets showed that:

- The targets do not have to be dissolved in flux to extract the tritium; in fact, the use of flux results in a 1 to 2% loss of tritium to the residue.

- The tritium can be extracted from the targets by heating at 850°C in a vacuum for 11 hours, with less than 0.1% tritium lost to the residue. Use of a lower extraction temperature increases the loss of tritium in the residue; for example, 11 hours at 750°C resulted in a 1.2% loss.

- The isotopic purity of tritium in the feed to the plant thermal diffusion columns is expected to be in the range of 50 to 80%. The impurity is protium derived from H_2O present in the ceramic and on the crucible; the evolved water vapor will be decomposed on an existing uranium bed.

[redacted]

[redacted]

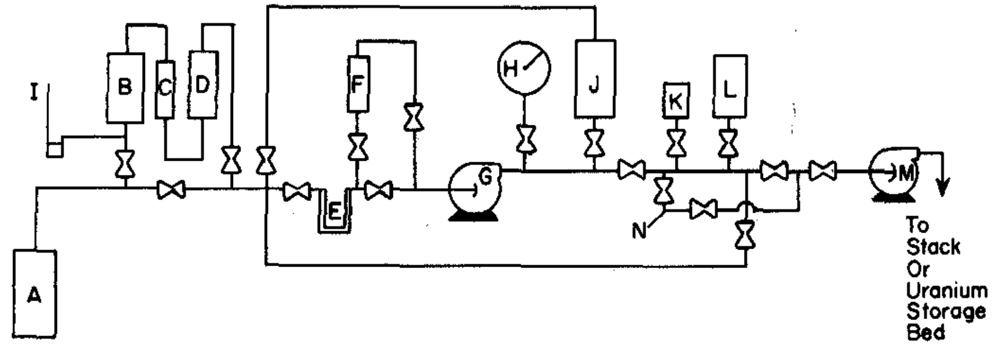
- CO and CO_2 evolved from the ceramic during extraction add a volume equivalent to one-tenth to one-third that of the tritium.

- The hydrogen isotopes can be separated from most of the CO and CO_2, if necessary, with Hopcalite* (oxidizer) and zeolite (water absorber) beds in series. This treatment will be used only if current studies show that the effectiveness of the plant uranium decomposer and/or the palladium diffuser is reduced by these gases.

DISCUSSION

EXPERIMENTAL APPARATUS

The pilot-scale tritium extraction equipment used for these tests is illustrated schematically in Figure 1. The apparatus consisted of a vacuum furnace capable of heating a 1-foot target element and the required flux to a temperature of 1000°C, a combination of Hopcalite (oxidizer) and zeolite (water absorber) beds in series for separating the isotopes of hydrogen from the other extraction gases, a uranium bed used for a decomposer, a specially built vacuum pump with a limited collection volume on the discharge side (to permit accurate measurements of small gas volumes), and gas collection vessels. Pressures were measured



A - Extraction Furnace
B - "Hopcalite" Bed
C - Cooling Water Jacket
D - Zeolite Bed
E - Cold Trap

F - Uranium Bed Decomposer
G - Special Vacuum Pump
H - 0-150 cm Hg Ashcroft Cage
I - Closed End Manometer
J - 8339-cc Collection Vessel

K - 33.2-cc Collection Vessel
L - 77.6-cc Collection Vessel
M - Modified Welch "Duo-Seal" Vacuum Pump
N - Sample Point

FIG. 1 SCHEMATIC DIAGRAM OF EXTRACTION FACILITY

---

* Hopcalite is the registered trademark of Mine Safety Appliances Co. for chemical materials for use as a catalyst.

[redacted]

with an 8-inch-diameter 0-150 cm Hg, pressure-vacuum gage. A trap cooled with liquid nitrogen was included between the furnace and the pump to collect all gases condensable at -77°C. A Welch "Duo-Seal" vacuum pump, modified by W. M. Welch Scientific Company to be leaktight on both the vacuum and discharge sides, was used to transfer gases from the collectors to either the stack or a uranium storage bed. All apparatus was of metal except for the glass cold trap and sample bulbs. The furnace was shielded to protect personnel against radiation, and all equipment was enclosed in a ventilated glovebox. Figure 2 is a photograph of the complete facility.

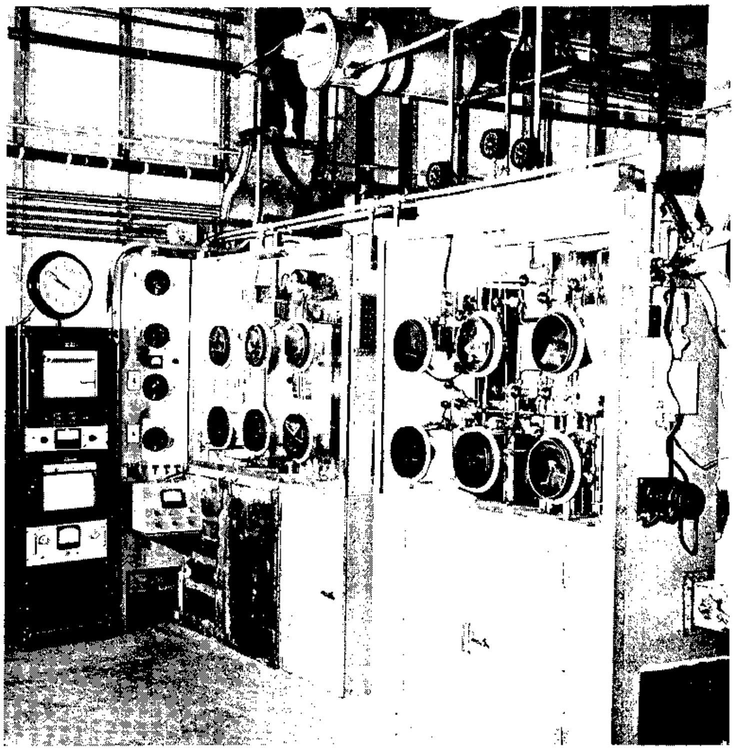


FIG. 2 PHOTOGRAPH OF EXTRACTION FACILITY

EXPERIMENTAL PROCEDURE

Extraction Cycle

The extractions were made with various charges to the furnace to study the effects of the following variables: ratio of flux to target, pretreatment of flux, location of target in flux, material of construction of crucible, and time for extraction at various temperatures. After loading the furnace and pumping the system down to less than 50 microns, a normal extraction cycle started with a 2-hour furnace heatup period to the desired extraction temperature. The gases evolved were passed through one or more of the following systems, depending on the desired results: Hopcalite-zeolite beds, cold trap, and uranium decomposer. All gases were collected in calibrated volumes at known temperature and pressure, and were analyzed by mass spectrometry.

When the cold trap was used, it was warmed to room temperature at the completion of the extraction cycle and the gases evolved were collected and sampled for analysis. Further heating of the trap, to about 100°C, evolved the water vapors (H_2O, HTO, T_2O), which were pumped through the uranium decomposer at 550°C, collected, and analyzed.

At the completion of each run with irradiated targets the equipment was rinsed with hydrogen at elevated temperatures to remove tritium absorbed on the walls. The tritium collected in the rinse was added to that collected during the run.

Hopcalite-zeolite System

The Hopcalite (88 g of an 80/20 mixture of manganese dioxide and cupric oxide) and zeolite (61 g of Linde Molecular Sieve Type 3A) beds were used during two runs to demonstrate a method of separating the hydrogen isotopes from the majority of the other gases. The system is illustrated schematically in Figure 3.

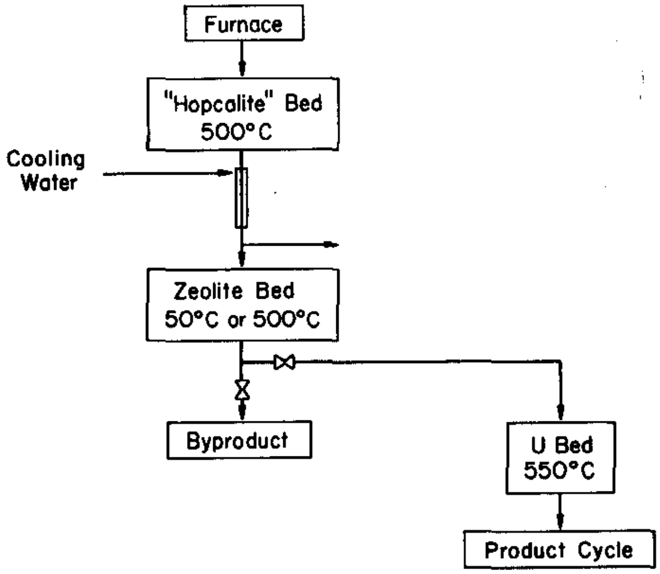


FIG. 3 PROCEDURE FOR REMOVAL OF $CO + CO_2$ FROM PRODUCT

The gases evolved from the furnace were passed through the Hopcalite bed at 500°C to oxidize the hydrogen isotopes. The vapors were then sorbed on the zeolite bed, which was maintained at 50°C during the extraction cycle, while the remaining gases (mainly He, CO, and CO_2) were collected, sampled, and discharged to the stack. At the completion of the extraction cycle, the zeolite bed was heated to 500°C for desorption, and the water vapors were passed through a uranium (185 g U) bed decomposer at 550°C to produce the gaseous hydrogen isotopes.

Residue Analysis

Several methods were used to measure the tritium content of the residues. The residue from runs made using flux was sampled at various locations in the crucible; each sample was dissolved in boiling 5M HCl in a closed system. Gases evolved during the dissolution were sampled and analyzed by mass spectrometry. The HCl solution was decomposed with calcium metal and the resulting hydrogen was analyzed for tritium by an ion chamber.

Residues from the runs in which no flux was used were analyzed by two methods: (1) A 4 to 6 g sample of the residue was dissolved in a closed system by a 50-50 mixture of concentrated H_2SO_4 and concentrated H_3PO_4; the gas phase was analyzed by mass spectrometry and the liquid phase was diluted, decomposed with calcium, and the gases were analyzed for tritium by ion chamber. (2) A 0.5 to 0.9 g sample of the residue was heated to 1400°C in a closed system under vacuum, and the evolved gases were collected and analyzed directly in the ion chamber. Results by the second method of analysis showed significant variation between several samples from one pellet, whereas good agreement was obtained using the first method. The necessity of using small samples for the second method probably prevented representative sampling. Only analyses obtained by the first method are presented in this report.

CRITERION FOR SATISFACTORY EXTRACTION

Product loss in the residue was used as the sole criterion for satisfactory extraction of tritium during a run. Normally the tritium content of a target at the time of extraction is calculated from the helium quantity: $\text{cc } T_2 = 1/2\ (\text{cc } ^4\text{He} - \text{cc } ^3\text{He})$. However, the targets used in these studies were vacuum-tested for cladding leaks after fabrication by introducing ^{4}He into the target through a small hole in the end cap, and sealing the hole. The excess ^{4}He varied from target to target thus making it impossible to obtain the normal material balance after extraction.

[redacted]

Estimates of the degree of lithium burnup in the reactor similarly were not precise enough to provide a reliable indication of recovery performance. Further error was introduced by adsorption of tritium on the walls of the equipment, an effect that was accentuated by the very low tritium content of the targets (average T_2 GVR = 0.63 STP cc/cc ceramic). In contrast, the tritium content of the residue can be measured accurately to less than 0.1% of the total tritium.

TARGET DESCRIPTION

The unirradiated targets used for these tests consisted of an aluminum housing tube, 13 inches long by 1-1/4 inches OD, containing pellets of lithium aluminate 2 to 2-1/2 inches long and 1-3/32 inches in diameter. The density of the pellets was 2.33 g/cc (89% of theoretical).

The irradiated targets were similar to the unirradiated targets except that they were only 12 inches long and the density of the pellets averaged about 2.07 g/cc (79.3% of theoretical). The zirconium jackets that were present over the aluminum during irradiation were removed at Hanford before shipping.

RESULTS

The data for all of the runs are summarized in Table IV of Appendix A, which gives the conditions for each run, the overall gas composition in terms of the major gases evolved, and the tritium content of the residues. Tables V through XIV of Appendix A list in detail the composition of the gases evolved at various times during extraction of the irradiated NPR ceramic targets. Observations made during the tests are discussed below.

Elimination of Flux

Before the irradiated targets were received, a fairly extensive preparatory study was made of the dissolution of lithium aluminate in sodium tetraborate, in accordance with the original process concept. Subsequent tests made with irradiated targets showed that flux was not needed to recover the tritium, so the remainder of the program was concentrated on the thermal extraction of tritium without the use of flux; only these nonflux runs will be discussed in the main section of this report. The preliminary studies made with flux and unirradiated targets before receipt of the irradiated targets are presented in Appendix B.

[redacted]

Temperature and Time Requirements

Figure 4 shows the rate at which gases are evolved during a typical extraction (no flux) in a stainless steel crucible (Run 13). As the crucible was heated to the desired temperature for extraction (900°C in Run 13), the aluminum can melted (~660°C) releasing an initial surge of ^{4}He, H_2, T_2, and $CO+CO_2$. After 8 hours of heating, with about 6 hours at extraction temperature, the evolution of ^{4}He stopped and the rate of H_2, T_2, and $CO+CO_2$ evolution decreased rapidly. As indicated by the residue analyses in Table I, more than 99.9% of the product had been extracted from the target in 6 to 11 hours; the H_2 and T_2 recovered after that time is gas that was absorbed on the walls of the metal equipment during the initial surge and was subsequently vacuum outgassed. The CO and CO_2 evolved are from both target and crucible.

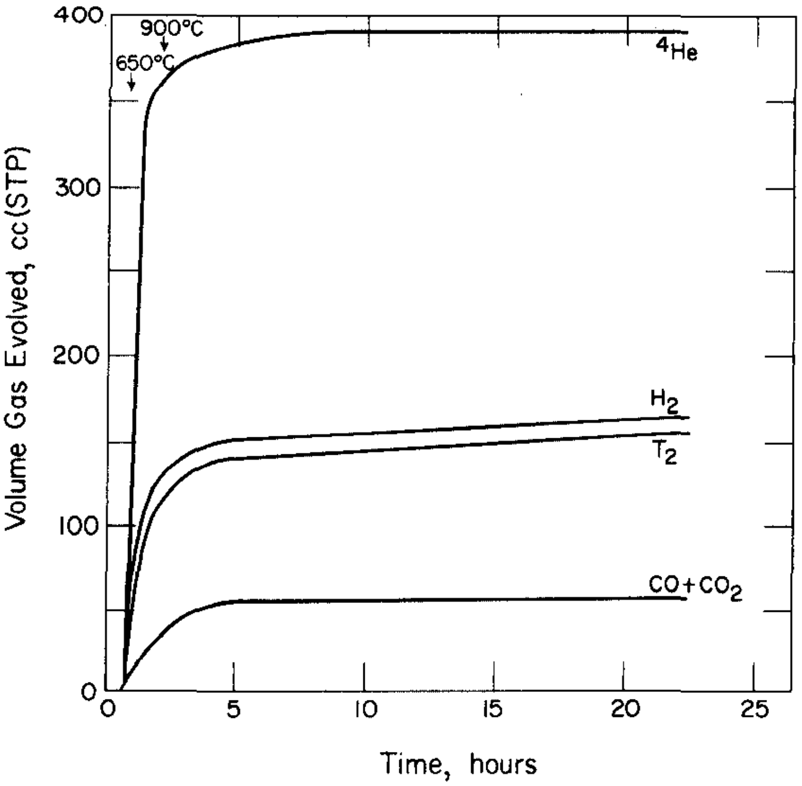


FIG. 4 GASES EVOLVED AT 900°C - NO FLUX (Run 13)

At the completion of a run the pellets were brittle and easily cracked along their axes; higher temperatures of extraction increased the brittleness.

TABLE I

Tritium Loss to the Residue

Run	Condition	Tritium Loss to Residue,[a] %
13	24 hours at 900°C	0.004
9	21 hours at 850°C	0.012
14	11 hours at 850°C	0.008[b]
16	6 hours at 850°C	0.36[c]
15	11 hours at 750°C	0.91
21	11 hours at 750°C	1.5
20	Residue from Run 15, 6 hours at 850°C	0.012
23	Residue from Run 21, 6 hours at 850°C	0.008

(a) Based on tritium recovered.

(b) Believed to be low by a factor of 10.

(c) Average of 0.41, 0.32, and 0.36%, the analyses obtained from three different pellets.

The residue analyses for the runs using irradiated targets and no flux, summarized in Table I, show that tritium can be satisfactorily extracted from lithium aluminate targets in, at most, 11 hours at 850°C (Runs 14 and 16). In runs 15 and 21 the target was heated to only 750°C for 11 hours; the resulting average loss of 1.2% is considered too high for normal plant production runs (<0.1%), but should be low enough to permit a full-scale extraction test in the separations plant.

Isotopic Purity

The capacity of the plant thermal diffusion columns for separating hydrogen from tritium decreases considerably as the tritium concentration in the feed decreases, as shown in Table II. The isotopic purity of the extraction gases during the nonflux runs varied from 15 to 46%, with 15 to 20% of the tritium present as HTO and T_2O (Runs 9, 16, 21). As noted in the next section, most of the protium comes from the crucibles. Furthermore, all of these runs were made with tritium GVR's (ratio, volume of gas at STP to volume of target) varying from 0.25 to 0.95, whereas the expected production targets for the plant will have a tritium GVR of about 1.2. Because the hydrogen content (water) of the production targets is expected to be lower than that of the targets used in this study, the isotopic purity for the plant feed to the thermal diffusion columns should be in excess of 50 to 60%. (The water content of targets currently produced at the NPR corresponds to a feed of 75 to 80% T_2.) At this concentration, one thermal column will be sufficient to process the predicted load of 6 kg/yr from the NPR.

TABLE II

DELETED

Material of Construction for Crucible

Tests comparing mild steel and stainless steel crucibles showed that the use of 304L stainless steel crucibles in the plant should be continued, since stainless evolves significantly less $CO+CO_2$ than does mild steel, as shown in Table III. The expected $CO+CO_2$ GVR for typical plant operation would be less than the 0.4 observed in Run 16, since the much higher ratio of target to crucible mass in the plant will decrease the relative contribution of $CO+CO_2$ by the crucible; the empty crucible in Run 22 evolved $CO+CO_2$ equivalent to 0.32 GVR, implying a contribution of only 0.1 GVR from ceramic in Runs 16, 17, and 21. The data in

TABLE III

Gases Evolved from Crucibles of Stainless Steel and Carbon Steel

Run	Crucible Steel	Contents	Temp, °C	Time, hr	H_2, GVR	$CO+CO_2$, GVR
15	304L	Irr'd. Target	750	11	0.66	0.80
21	304L	Irr'd. Target	750	11	1.06	0.42
16	304L	Irr'd. Target	850	6	0.73	0.42
17	304L	Unirr'd. Target	850	4	0.70	0.40
22	304L	Empty	850	4	0.74(a)	0.32(a)
18	Carbon	Unirr'd. Target	850	4	0.73	2.22
19	Carbon	Empty	850	4	0.51(a)	2.50(a)

(a) Calculated as though a target were present.

Table III also indicate that most of the H_2 evolved during an extraction comes from the crucible (Runs 22 and 19) and not the target; again, the relative contribution will be diminished in the plant.

Removal of CO and CO_2

The CO and CO_2 evolved during the extraction of tritium from the ceramic targets may have detrimental effects in the plant on the performance of the uranium bed decomposer (reduces H_2O, HTO, and T_2O) and the palladium diffuser (separates hydrogen isotopes from other components of the process gases). A portion of the CO and CO_2 will be reduced by the uranium, an action which adds to the consumption of uranium and may lead to inactivation by a surface layer of carbon. Decomposition of methane on the palladium diffuser can decrease its performance sufficiently to require oxidative regeneration; fortunately, methane was not produced when mixtures of T_2, CO, and CO_2 were passed through the hot uranium decomposer (Run 13). Because of these uncertainties regarding the effects of $CO+CO_2$, an additional purification step using a combination of Hopcalite and zeolite beds in series was developed for the plant in case it should prove necessary to prevent the $CO+CO_2$ from passing through the uranium decomposer and/or the palladium diffuser. This process is described in the section under Experimental Procedure and is shown in Figure 3.

Eighty to ninety percent of the $CO+CO_2$ was separated from the tritium stream by the Hopcalite-zeolite system (Runs 14 and 15), with no tritium lost to the byproduct stream of He and $CO+CO_2$.

FUTURE WORK

The extraction process will be confirmed with additional irradiated targets from the NPR. These targets, which are more representative of the expected production targets, will have an average ceramic density of 88.6% of theoretical and an average tritium GVR of 1.4, compared to 79.3% and 0.6 for the targets already investigated. The effects of $CO+CO_2$ on the uranium decomposer and palladium diffuser will be determined and the ability to extract tritium at 750 and 850°C will be verified. This evaluation should eliminate the need for a large-scale demonstration at the Semiworks, and permit a full-scale extraction test to be conducted at 750°C in the plant without equipment modification.

APPENDIX A

DETAILED SUMMARY OF DATA OBTAINED

Table IV summarizes all of the runs made. Tables V through XIV give the detailed composition of the gases evolved at various times during all of the tests with irradiated targets, and during the tests to determine the material of construction for the crucible.

[redacted]

TABLE IV

Summary of Tests

Run	Furnace Charge	Crucible	Temp-Time, °C-hr	H_2 - Total cc (STP)	H_2 - Total GVR(a)	H_2 as H_2O, %
1-a	79g tgt (no Al) 674g flux	Carbon Steel	800-1 750-4	710	22.6	87
1-b	same	Carbon Steel	780-3	34	1.1	0
2-a	1270g flux (powder)	Carbon Steel	600-5	10,400	81.3	95
2-b	same	Carbon Steel	850-4	<1,000	<8.1	<60
2-c	same	Carbon Steel	850-4	213	1.7	0
2-d	same	Carbon Steel	1,000-11(f)	276	2.2	0
3-a	500g tgt	Carbon Steel	550-4½	<615	<3.1	<97
3-b	same	Carbon Steel	850-6	50	0.3	68
4	500g tgt,1900g flux	Carbon Steel	850-10	893	4.5	32
5	same as 4 except tgt suspended & no Al	Carbon Steel	850-10	543	2.7	35
6	500g tgt, 1070g flux	304 LC Stainless Steel	850-10	888	4.4	54
7	1070g flux	304 ELC Stainless Steel	850-7	2,500	21.8	87
8	410g tgt suspended 1980g flux	Carbon Steel	850-20	1,670	9.0	83
9	410g tgt	Carbon Steel	850-21	405	2.2	67
10	1230g flux	Carbon Steel	850-11	2,150	7.0	55
11	410g tgt(h) suspended 1980g pretreated flux	Carbon Steel	850-20	275	1.5	53
12	410g tgt suspended 1980g pretreated flux	Carbon Steel	850-24	242	1.4	?(i)
13	410g tgt	304L Stainless Steel	900-24	266	1.5	?(i)
14	410g tgt	304L Stainless Steel	850-11	445	2.4	?(i)
15	410g tgt	304L Stainless Steel	750-11	122	0.7	?(j)
16	410g tgt	304L Stainless Steel	850-6	134	0.7	35
17	500g tgt	304L Stainless Steel	850-4	139	0.7	46
18	500g tgt	Carbon Steel	850-4	146	0.7	29
19	Carbon Steel Crucible	Carbon Steel	850-4	94	0.5(l)	30
20	Residue from Run 15	304L Stainless Steel	850-6	254(m)	1.4	?(i)
21	410g tgt	304L Stainless Steel	750-11	196	1.1	35
22	304L Crucible	304L Stainless Steel	850-4	136	0.7(l)	16
23	Residue from Run 21	304L Stainless	850-6	1110(n)	6.0	?(i)

(a) GVR -- ratio, volume of gas at STP to volume of target. If flux is present but no target material, a 4/1 flux to charge weight ratio is assumed and GVR is expressed as if tgt were present.

(b) Some helium in target prior to irradiation.

(c) Irradiated targets used in Runs 8, 9, 11, 12 through 16, 20, and 21.

[redacted]

TABLE IV (Continued)

CO + CO_2		H_e[b]	T_2[c]		T_2 as HTO and T_2O, %	Isotopic Purity, % T_2	T_2 Remaining in Residue,[d] %
cc (STP)	GVR[a]	cc (STP)	cc (STP)	GVR[a]			
-	-	5.6	-	-	-	-	-
-	-	2.3	-	-	-	-	-
-	-	5.7	-	-	-	-	-
277[e]	2.2	0	-	-	-	-	-
189[e]	1.5	0	-	-	-	-	-
1,020[e]	8.0	0	-	-	-	-	-
8	-	0	-	-	-	-	-
50	0.2	4.3	-	-	-	-	-
687	3.4	5.1	-	-	-	-	-
921	4.6	3.0	-	-	-	-	-
47	>0.2[g]	3.2	-	-	-	-	-
128	1.1	0	-	-	-	-	-
2,220	12.0	412	172	0.92	75	9	1.4
1,420	7.6	331	129	0.70	17	24	0.012
1,250	4.0	-	-	-	-	-	-
1,440	7.8	400	123	0.67	40	31	1.2
1,220	6.7	259	87	0.47	-	27	1.7
59	0.3	381	175	0.95	-	40	0.004
9 (93)[k]	0.05 (0.5)[k]	296	92	0.49	-	15	0.008
33 (148)[k]	0.18 (0.8)[k]	349	46	0.25	-	27	0.91
77	0.4	279	117	0.63	20	46	0.36
80	0.4	4.7	-	-	-	-	-
445	2.2	3.5	-	-	-	-	-
462	2.5[l]	1.6	-	-	-	-	-
204	1.1	3.4	2.4	-	-	-	-
77	0.4	3.5	84	0.46	15	30	1.5
59	0.3[l]	0	-	-	-	-	-
0.5	0.03	1.2	2.1	-	-	-	-

(d) Based on T_2 recovered.

(e) Minimum value, not all was collected.

(f) Flux decomposed.

(g) Gas was trapped in the borax, probably CO+CO_2.

(h) Not all of the $LiAlC_2$ was in the flux.

(i) All the gases were passed directly through a uranium decomposer at 550°C.

(j) All the gases were passed directly through a Hopcalite and zeolite bed.

(k) Value in parentheses based on total CO+CO_2 evolved, not just that evolved with the T_2 from zeolite bed.

(l) Based on 410g target.

(m) Residue probably absorbed moisture while exposed to air for several days.

(n) Residue probably absorbed H_2 during exposure to an H_2 rinse.

TABLE V

Run 8: Gases Evolved from Irradiated Target in Flux at 850°C for 20 Hours - Carbon Steel Crucible

		Total cc (STP)												
Date	Time, min	Total Vol[a]	H_2	^3He	^4He	HT	T_2	Total T_2[b]	CO	N_2	O_2	CO_2	Ar	CH_4
2/14/66	47	154	5.68	0.54	131	1.11	4.90	5.73	10.4	-	-	0.32	-	0.03
"	52	170	1.72	0.75	153	-	1.00	1.38	13.0	-	0.12	0.46	-	0.03
"	67	96.7	3.06	0.69	84.8	-	0.18	0.53	1.26	5.01	0.12	0.49	-	0.07
"	115	155	18.2	0.60	33.7	8.67	3.05	7.68	86.7	2.34	0.08	1.42	-	0.32
"	161	178	34.6	-	7.41	8.66	0.60	4.93	126	-	0.02	0.35	-	0.05
"	210	160	39.0	-	0.35	9.57	0.62	5.50	110	-	0.02	0.45	-	0.08
"	285	171	37.7	-	-	6.20	0.38	3.48	126	-	0.07	0.41	-	0.07
"	375	170	25.7	-	0.07	5.64	0.34	3.16	138	-	0.20	0.39	-	-
"	480	164	18.3	-	-	4.03	0.20	2.21	141	-	0.21	0.08	-	-
"	600	165	15.4	-	-	3.52	0.23	1.99	146	-	0.15	0.11	-	-
"	730	161	12.3	-	-	2.92	0.19	1.65	145	-	0.16	0.15	-	0.05
"	(c)	419	0.5	-	1.22	0.21	0.21	0.32	4.74	1.22	0.17	411	-	0.16
"	(d)	1530	1220	-	-	227	10.2	124	14.0	10.7	-	42.7	-	0.46
Total		3690	1430	2.58	412	278	22.1	163	1060	19.2	1.32	458	-	1.32
2/15/66	-	151	5.78	-	-	1.15	0.08	0.66	13.3	98.1	31.4	0.23	1.26	-
"	160	162	18.7	-	-	3.48	0.18	1.92	123	12.7	2.96	0.26	0.15	0.05
"	295	154	8.22	-	-	1.68	0.09	0.93	143	0.37	0.66	0.22	0.02	0.03
"	460	157	6.12	-	-	1.25	0.08	0.70	149	-	0.17	0.27	0.02	0.03
"	535	65.9	2.16	-	-	0.42	0.03	0.24	62.1	0.57	0.34	0.18	0.01	0.01
"	625	74.6	2.28	-	-	0.44	0.03	0.25	71.8	-	0.06	0.03	-	0.01
"	(c)	135	0.03	-	-	-	0.03	0.03	0.18	-	-	134	-	0.07
"	(d)	59.6	41.3	-	-	7.52	0.35	4.01	9.20	0.11	0.04	1.04	-	0.05
Total		959	84.6	-	-	15.9	0.87	8.74	572	112	35.6	136	1.47	0.25
Grand Total		4650	1520	2.58	412	294	23.0	172	1630	131	37.0	594	1.47	1.57
Product retained in equipment								?						
Product retained in residue								2.34						
Total product								174						

(a) Column 9 not included.

(b) Includes 1/2 the ^3He found.

(c) Gases evolved from cold trap while thawing.

(d) Gases evolved from heating cold trap and passing gases through a uranium decomposer at 550°C.

TABLE VI

Run 9: Gases Evolved from Irradiated Target at 850°C for 21 Hours - Carbon Steel Crucible

		Total cc (STP)												
Date	Time, min	Total Vol[(a)]	H_2	^3He	^4He	HT	T_2	Total T_2[(b)]	CO	N_2	CO_2	Ar	CH_4	O_2
2/17/66	29	169	19.1	0.47	128	1.35	2.75	3.67	3.06	13.0	0.22	0.35	0.41	-
"	35	155	6.41	0.93	127	5.62	1.49	4.76	2.93	10.2	0.39	0.26	0.08	-
"	49	159	15.1	0.57	50.5	15.6	5.06	13.2	56.6	15.2	0.57	0.08	0.10	-
"	60	163	4.72	-	6.03	15.7	15.7	23.5	110	9.64	1.24	-	0.03	-
"	74	168	3.07	-	5.21	11.0	10.7	16.2	134	2.75	1.47	-	0.03	0.05
"	81	164	3.43	0.03	5.16	8.28	5.22	9.38	126	15.3	0.86	-	0.03	0.08
"	107	169	4.07	0.02	3.30	7.12	3.18	6.75	142	8.04	0.71	-	0.07	0.02
"	165	177	4.69	-	1.58	5.52	1.75	4.51	138	24.7	0.18	-	0.05	0.14
"	285	167	5.54	-	1.12	4.61	1.02	3.32	154	-	0.22	-	0.03	0.03
"	420	112	4.47	-	0.66	3.07	0.61	2.15	103	-	0.04	-	0.02	0.04
"	540	70.6	3.21	-	0.39	2.15	0.34	1.42	60.4	3.97	0.03	0.01	0.01	0.05
"	630	42.6	2.05	0.02	0.20	1.18	0.19	0.79	35.5	3.42	0.01	-	0.01	0.06
"	720	38.9	2.10	-	0.18	1.15	0.16	0.74	34.0	1.33	-	-	0.01	0.06
"	(c)	94.2	0.01	-	-	0.01	0.01	0.02	-	0.41	93.6	-	0.08	0.06
"	(d)	326	245	-	0.39	39.7	1.73	21.6	-	5.98	32.9	0.07	-	0.56
Total		2180	323	2.04	330	122	49.9	112	1100	114	132	0.77	0.96	1.15
2/18/66	255	59.5	4.22	-	0.19	2.41	0.30	1.50	52.0	0.21	0.03	-	0.13	0.04
"	450	59.0	2.97	-	0.22	1.60	0.19	0.99	50.9	3.07	-	-	0.02	0.04
"	630	48.6	2.44	-	0.16	1.28	0.15	0.79	41.5	2.99	0.01	-	0.01	0.08
"	780	36.4	1.95	-	0.11	0.93	0.09	0.55	30.6	2.69	-	-	0.01	0.05
"	(e)	10.0	5.61	-	-	1.09	0.05	0.59	-	0.23	2.99	0.01	-	0.02
Total		214	17.2	-	0.68	7.31	0.78	4.42	175	9.19	3.03	0.01	0.17	0.23
Grand Total		2390	340	2.04	331	129	50.7	116	1280	123	135	0.78	1.13	1.38
Product retained in equipment								12.5						
Product retained in residue								.015						
Total product								129						

(a) Column 9 not included.
(b) Includes 1/2 the ^3He found.
(c) Gases evolved from cold trap while thawing.
(d) Gases evolved from heating cold trap and passing gases through a uranium bed decomposer at 550°C.
(e) Combination of (c) and (d).

TABLE VII

Run 11: Gases Evolved from Irradiated Target in Pre-Treated Flux* at 850°C for 20 Hours - Carbon Steel Crucible

		Total cc (STP)												
Date	Time, min	Total Vol$^{(a)}$	H_2	^3He	^4He	HT	T_2	Total $T_2$$^{(b)}$	CO	N_2	CO_2	Ar	CH_4	O_2
2/24/66	70	176	9.48	0.63	149	1.87	4.78	6.03	1.87	7.58	0.12	0.26	0.02	0.48
"	75	170	2.65	0.76	151	4.63	2.32	5.02	0.95	6.83	0.12	0.22	-	0.05
"	120	146	8.79	1.18	60.8	11.2	4.18	10.4	46.4	12.2	0.41	0.07	0.04	0.44
"	150	148	7.83	0.36	28.8	24.6	8.23	20.7	68.6	8.70	0.83	-	0.07	-
"	210	145	27.9	-	6.64	17.4	3.04	11.7	79.1	10.3	0.14	-	0.03	0.16
"	330	143	13.7	-	2.48	9.26	1.71	6.34	114	1.22	0.37	0.04	0.03	0.42
"	480	134	6.71	-	0.94	4.41	0.83	3.03	120	0.28	0.28	0.04	0.01	0.55
"	600	89.0	2.97	-	0.08	1.84	0.33	1.25	82.9	-	0.36	0.03	0.01	0.44
"	720	86.6	2.09	-	0.08	1.34	0.23	0.90	81.3	-	1.01	0.03	0.02	0.46
"	(c)	259	3.24	-	-	0.05	0.05	0.08	-	-	256	-	-	-
"	(d)	190	93.0	-	-	64.7	13.5	45.9	-	2.48	15.3	-	0.06	0.59
Total		1690	178	2.93	400	141	39.2	111	595	49.6	275	0.69	0.29	3.59
2/25/66	210	144	4.47	-	0.07	2.26	0.27	1.40	134	1.87	0.22	0.03	0.03	0.42
"	390	111	2.23	-	0.04	1.17	0.16	0.76	106	1.39	0.12	0.02	0.01	0.29
"	570	105	1.76	-	0.03	0.93	0.12	0.69	102	-	0.04	0.01	0.01	0.25
"	750	100	1.60	-	0.03	1.67	0.10	0.99	96.3	0.06	0.06	0.02	0.01	0.21
"	(c)	125	0.01	-	-	-	0.02	0.02	-	0.30	125	-	-	0.05
"	(d)	26.4	10.6	-	-	4.83	0.52	2.42	4.99	2.72	2.69	0.01	0.01	0.06
Total		611	21.6	-	0.17	10.9	1.19	6.28	443	6.34	128	0.09	0.07	1.28
Grand Total		2300	199	2.93	400	152	40.4	117	1040	56.0	403	0.78	0.36	4.87
Product retained in equipment								4.8						
Product retained in residue								1.5						
Total product								123						

---

* Vacuum dried at 850°C for 11 hours prior to run.

(a) Column 9 not included.
(b) Includes 1/2 the ^3He found.
(c) Gases evolved from cold trap while thawing.
(d) Gases evolved from heating cold trap and passing gases through a uranium bed decomposer at 550°C.

TABLE VIII

Run 12: Gases Evolved from Irradiated Target in Pre-Treated* Flux at 850°C for 24 Hours - Carbon Steel Crucible

		Total cc (STP)												
Date	Time, min	Total Vol[a]	H_2	^3He	^4He	HT	T_2	Total T_2[b]	CO	N_2	CO_2	Ar	CH_4	O_2
3/7/66	136	547	91.7	2.19	249	51.0	13.1	39.7	122	11.0	4.77	1.70	0.44	0.33
"	185	135	21.2	0.01	8.55	20.2	4.24	14.3	76.4	-	3.98	0.01	0.03	0.01
"	290	157	20.6	-	1.38	16.0	2.97	11.0	110	-	6.40	-	0.03	0.02
"	440	156	15.6	-	0.45	10.0	1.58	6.60	122	-	6.44	0.03	0.03	0.11
"	620	145	9.77	-	0.03	5.69	0.78	3.63	122	-	6.16	0.01	0.03	0.07
"	800	123	6.47	-	-	3.58	0.47	2.26	107	-	5.50	-	0.02	0.06
Total		1263	165	2.20	259	106	23.1	77.5	659	11.0	33.3	1.75	0.58	0.60
3/8/66	210	129	7.99	-	-	3.67	0.37	2.21	108	0.53	8.20	0.01	0.01	0.01
"	360	118	3.22	-	-	1.54	0.18	0.95	104	-	8.84	-	0.01	0.12
"	570	142	3.47	-	-	1.63	0.19	1.01	126	-	10.8	-	0.01	0.03
"	870	175	4.28	-	-	1.85	0.21	1.13	156	-	12.3	-	0.02	0.07
Total		564	19.0	-	259	8.70	0.95	5.30	494	0.53	40.1	0.01	0.05	0.23
Grand Total		1830	184	2.20		115	24.1	82.8	1150	11.5	73.4	1.76	0.63	0.83
Product retained in equipment								3.9						
Product retained in residue								1.5						
Total product								88.2						

---

* Vacuum dried at 850°C for 11 hours prior to run.

(a) Column 9 not included; all of the gases passed directly through uranium bed decomposer at 550°C during the run.

(b) Includes 1/2 the ^3He found.

TABLE IX

Run 13: Gases Evolved from Irradiated Target at 900°C for 24 Hours - 304L Stainless Steel Crucible

		Total cc (STP)												
Date	Time, min	Total Vol[(a)]	H_2	^3He	^4He	HT	T_2	Total T_2[(b)]	CO	N_2	CO_2	Ar	CH_4	O_2
3/10/66	95	689	118	2.96	361	110	45.0	102	24.4	23.2	3.31	0.75	0.48	0.20
"	230	121	23.9	0.02	16.3	37.9	16.2	35.1	24.5	1.38	0.92	0.01	0.02	0.01
"	800	55.0	23.6	-	2.33	18.1	4.16	13.2	5.12	1.38	0.11	0.04	0.03	0.11
Total		865	166	2.98	380	166	65.4	150	54.0	26.0	4.34	0.80	0.53	0.32
3/11/66	300	11.1	5.79	-	0.18	3.82	0.55	2.46	0.27	0.34	0.02	0.03	0.05	0.05
"	870	12.6	7.20	-	0.34	4.04	0.50	2.52	0.20	0.23	0.02	0.02	0.01	0.04
Total		23.7	13.0	-	0.52	7,86	1.05	4.98	0.47	0.57	0.04	0.05	0.06	0.09
Grand Total		889	179	2.98	381	174	66.5	155	54.5	26.6	4.38	0.85	0.59	0.41
Product retained in equipment								20.0						
Product retained in residue								0.007						
Total product								175						

(a) Column 9 not included; all of the gases passed directly through uranium bed decomposer at 550°C during the run.

(b) Includes 1/2 the ^3He found.

TABLE X

Run 14: Gases Evolved from Irradiated Target at 850°C for 11 Hours - 304L Stainless Steel Crucible

Date	Time, min	Total cc (STP)												
		Total Vol[(a)]	H_2	^{3}He	^{4}He	HT	T_2	Total T_2[(b)]	CO	N_2	CO_2	Ar	CH_4	O_2
3/22/66	62	175	0.02	0.70	111	-	-	0.35	-	14.2	7.17	0.35	0.28	41.3
"	72	177	-	1.89	141	-	-	0.94	-	12.5	12.3	0.30	0.23	8.48
"	370	153	0.06	0.70	42.3	-	-	0.35	-	4.04	51.2	0.02	0.08	54.1
"	760	38.6	0.06	0.03	1.41	-	-	0.01	-	0.90	12.8	-	0.02	23.4
Total		544	0.14	3.32	296	-	-	1.65	-	31.6	83.5	0.67	0.61	127
3/22/66	(c)	515	363	-	-	130	12.2	77.1	5.56	1.03	2.73	-	0.05	0.10
3/23/66	(c)	124	14.1	-	-	4.53	0.43	2.69	0.43	102	0.41	1.64	0.01	0.04
Total		639	377	-	-	135	12.6	79.8	5.99	103	3.14	1.64	0.06	0.14
Grand Total		1180	377	3.32	296	135	12.6	81.5	5.99	135	86.6	2.31	0.67	127
Product retained in equipment								10.7						
Product retained in residue								0.007						
Total product								92.2						

(a) Column 9 not included; all of the gases passed directly through Hopcalite and zeolite beds.

(b) Includes 1/2 the ^{3}He found.

(c) Gases evolved after heating zeolite bed to 500°C.

TABLE XI

Run 15: Gases Evolved from Irradiated Target at 750°C for 11 Hours - 304L Stainless Steel Crucible

Date	Time, min	Total cc (STP) Total Vol[(a)]	H_2	^3He	^4He	HT	T_2	Total T_2[(b)]	CO	N_2	CO_2	Ar	CH_4	O_2
3/28/66	67	175	-	1.01	142	-	-	0.50	-	14.8	10.1	0.35	0.26	5.93
"	76	179	-	2.09	156	-	-	1.04	-	9.56	10.7	0.32	0.25	0.11
"	280	129	0.03	1.10	49.3	-	-	0.55	-	13.2	58.1	0.14	0.09	7.17
"	775	62.1	0.05	0.04	1.53	-	0.01	0.02	-	15.6	36.5	0.20	0.02	8.10
Total		545	0.08	4.24	349	-	0.01	2.11	-	53.2	115	1.01	0.62	21.3
3/28/66	(c)	153	78.7	-	-	35.4	4.49	22.2	-	12.4	21.4	0.10	0.03	0.15
3/29/66	(c)	76.6	20.2	-	-	10.7	1.39	6.73	9.80	32.6	1.31	0.60	0.02	0.04
Total		230	98.9	-	- 349	46.1	5.88	28.9	9.80	45.0	22.7	0.70	0.05	0.19
Grand Total		775	99.0	4.24	349	46.1	5.89	31.0	9.80	98.2	138	1.71	0.67	21.5
Product retained in equipment								15.0						
Product retained in residue								0.42						
Total product								46.4						

(a) Column 9 not included; all of the gases passed directly through Hopcalite and zeolite beds.

(b) Includes 1/2 the ^3He found.

(c) Gases evolved after heating zeolite bed to 500°C.

TABLE XII

Run 16: Gases Evolved from Irradiated Target at 850°C for 6 Hours - 304L Stainless Steel Crucible

		Total cc (STP)												
Date	Time, min	Total Vol[(a)]	H_2	^3He	^4He	HT	T_2	Total T_2[(b)]	CO	N_2	Ar	CH_4	O_2	CO_2
3/30/66	68	179	18.0	0.77	137	2.63	1.25	2.95	5.02	13.6	0.41	0.11	0.27	0.13
"	90	185	22.5	2.13	110	24.4	13.1	26.4	4.30	8.40	0.19	0.09	0.07	0.11
"	460	151	16.1	0.32	32.2	34.0	24.4	41.5	43.0	0.23	-	0.06	0.05	0.48
Total		515	56.6	3.22	279	61.0	38.8	70.9	52.3	22.2	0.60	0.26	0.39	0.72
3/30/66	(c)	13.0	0.01	-	-	-	-	-	0.21	0.16	0.02	0.01	0.06	12.5
"	(d)	67.9	39.7	-	-	14.3	1.94	9.10	3.34	0.24	-	0.06	0.03	8.26
Total		80.9	39.7	-	-	14.3	1.94	9.10	3.55	0.40	0.02	0.07	0.09	20.8
Grand Total		596	96.3	3.22	279	75.3	40.7	80.0	55.9	22.7	0.62	0.33	0.48	21.5
Product retained by equipment								36.7						
Product retained in residue								0.42						
Total product								117						

(a) Column 9 not included.
(b) Includes 1/2 the ^3He found.
(c) Gases evolved from cold trap due to thawing.
(d) Gases evolved from heating cold trap and passing gases through a uranium decomposer at 550°C.

TABLE XIII

Runs 17, 18, 19, and 22: Gases Evolved from Carbon and Stainless Steel Crucibles at 850°C

			Total cc (STP)													
Run	Date	Time, min	Total Vol(a)	H_2	^3He	^4He	HT	T_2	Total T_2	CO	N_2	Ar	CH_4	O_2	CO_2	CO_2+CO
17	4/1/66	330	142	75.6	-	4.65	2.47	0.01	1.24	40.5	17.9	0.44	0.27	-	0.28	40.7
17A(b)	4/1/66	-	26.5	0.02	-	-	-	-	-	-	0.65	0.03	0.10	0.05	25.6	25.6
17B(c)	4/1/66	-	78.7	60.2	-	-	4.00	0.07	2.07	3.01	0.21	-	0.21	-	11.0	14.0
	Total		247	136	-	4.65	6.47	0.08	3.31	43.5	18.8	0.47	0.58	0.05	36.9	80.4
18	4/4/66	290	565	104	-	3.45	1.19	0.11	0.70	408	45.8	0.40	0.62	0.11	0.85	409
18A(b)	4/4/66	-	28.1	0.02	-	-	-	0.01	0.01	0.32	0.31	0.05	0.04	-	27.3	27.7
18B(c)	4/4/66	-	50.6	41.2	0.03	0.04	1.03	0.01	0.52	1.52	0.14	-	0.07	0.02	6.50	8.02
	Total		644	145	0.03	3.49	2.22	0.13	1.23	410	46.3	0.45	0.73	0.13	34.7	445
19	4/5/66	270	555	65.5	-	1.55	0.50	0.17	0.42	438	48.2	-	0.61	0.61	0.22	438
19A(b)	4/5/66	-	20.7	0.05	-	-	-	0.01	0.01	0.59	0.36	0.05	0.06	0.06	19.5	20.1
19B(c)	4/5/66	-	32.3	27.9	-	-	0.60	-	0.30	0.66	0.21	0.01	0.05	0.06	2.80	3.46
	Total		608	93.5	-	1.55	1.10	0.18	0.73	439	48.8	0.06	0.72	0.73	22.5	462
22	4/19/66	325	158	114	-	-	0.44	-	0.22	42.8	-	-	0.16	0.06	0.13	42.9
22A(b)	4/19/66	-	12.8	0.07	-	-	-	-	-	0.33	0.20	0.01	0.06	0.01	12.1	12.4
22B(c)	4/19/66	-	26.3	22.1	-	0.01	0.29	-	0.15	0.94	0.34	-	0.07	-	2.55	3.49
	Total		197	136	-	0.01	0.73	-	0.37	44.1	0.54	0.01	0.29	0.07	14.8	58.8

Run 17 - 304L crucible containing unirradiated target.
Run 18 - Carbon steel crucible containing unirradiated target.
Run 19 - Empty carbon steel crucible.
Run 22 - Empty 304L crucible.

(a) Columns 10 and 17 not included.
(b) Gas evolved from trap when thawed to room temperature.
(c) Gas passing through uranium decomposer while heating trap to about 100°C.

TABLE XIV

Run 21: Gases Evolved from Irradiated Target at 750°C for 11 Hours - 304L Stainless Steel Crucible

		Total cc (STP)												
Date	Time, min	Total Vol[a]	H_2	^3He	^4He	HT	T_2	Total T_2[b]	CO	N_2	CO_2	Ar	Ch_4	O_2
4/14/66	62	188	35.5	1.03	128	2.13	0.60	2.18	3.93	15.9	0.17	0.60	0.23	0.30
"	85	175	28.5	2.21	106	18.4	4.56	14.8	8.62	6.33	0.21	0.32	0.12	0.07
"	745	150	29.8	0.24	26.0	46.3	23.3	46.6	21.3	2.09	0.04	-	0.09	0.13
Total		513	93.8	3.48	260	66.8	28.5	63.6	33.9	24.3	0.42	0.92	0.44	0.50
(c)		20.6	0.01	-	0.01	0.02	0.01	0.02	0.49	0.24	19.6	0.04	0.02	0.11
(d)		95.6	66.6	-	-	5.33	0.12	2.78	3.67	0.60	19.1	-	0.09	0.11
Total		116	66.6	-	0.01	5.35	0.13	2.80	4.16	0.84	38.7	0.04	0.11	0.22
Grand Total		629	160	3.48	260	72.1	28.6	66.4	38.0	25.2	39.1	0.96	0.55	0.72
Product retained in equipment								17.8						
Product retained in residue								1.28						
Total product								85.5						

(a) Column 9 not included.
(b) Includes 1/2 the ^3He found.
(c) Gases evolved from cold trap due to thawing.
(d) Gases evolved from heating cold trap and passing gases through a uranium decomposer at 550°C.

[redacted]

[redacted]

[redacted]

APPENDIX B

EXTRACTION OF TRITIUM FROM LITHIUM ALUMINATE TARGETS USING FLUX

A considerable amount of work was done at the Savannah River Laboratory to evaluate the process proposed by Pacific Northwest Laboratory for the extraction of tritium from lithium aluminate targets by dissolution in molten $Na_2B_4O_7$ at 850°C. Although a flux was found not to be necessary, this work is of interest and is summarized in this Appendix.

In the proposed fluxing process, shown in Figure 5, the aluminum-clad targets are charged to the furnace with dehydrated sodium tetraborate in a weight ratio of 20% targets to 80% flux.

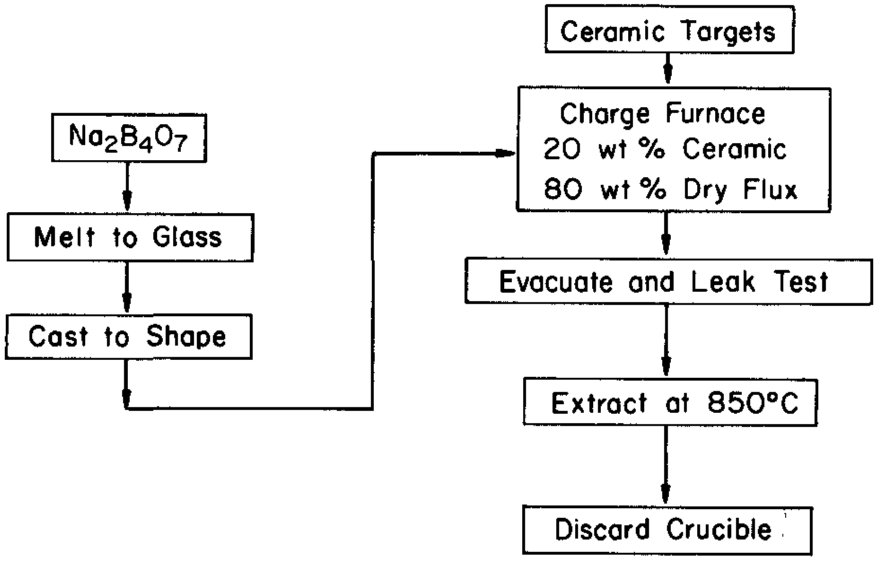


FIG. 5 PROPOSED FLOWSHEET FOR EXTRACTION PROCESS WITH FLUX

As the temperature of the evacuated furnace is increased to 850°C, some of the tritium is evolved when the cladding melts; this is elemental tritium which is released even at room temperature if the cladding is punctured. The remainder of the tritium slowly evolves as the flux dissolves the target. The crucible containing the residue is then buried. Significant information obtained from experiments with both irradiated and unirradiated targets to demonstrate the proposed process is discussed below.

[redacted]

Successful Dissolution

Dissolution of a $LiAlO_2$ target in four times its weight of molten sodium tetraborate at 850°C allows extraction of more than 98% of the tritium in 20 hours (Runs 9, 11, and 12). No trace of target material was found when the crucibles containing the residues from these runs were cut at two-inch intervals. Ceramic residue was not found on the bottom of the crucibles when the bottom sections were cut along their axes. A plot of the gases evolved during Run 12, shown in Figure 6, indicates that possibly all of the tritium was released from the target during about six hours at 850°C, as in the case of runs without flux, and that the remaining time was spent vacuum outgassing the tritium from the metal equipment. Since the dissolution of unirradiated ceramic in flux at 850°C was only about 40% complete in 10 hours, as estimated from the crucible sections of Run 5 in Figure 7, irradiated targets apparently dissolve faster than unirradiated targets.

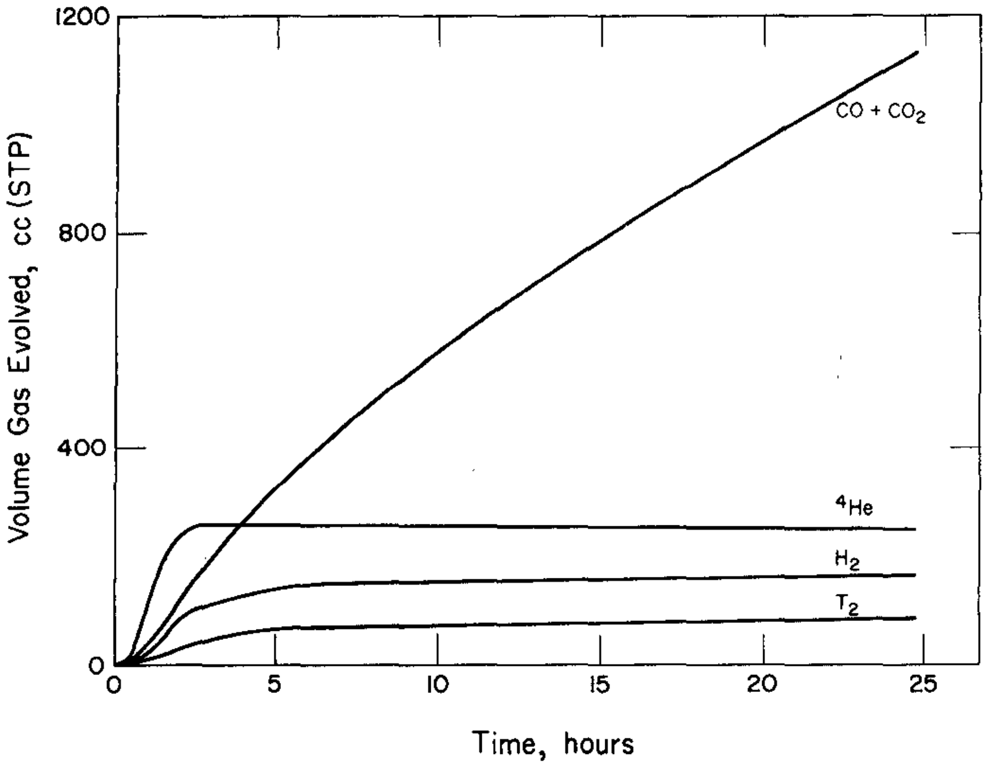
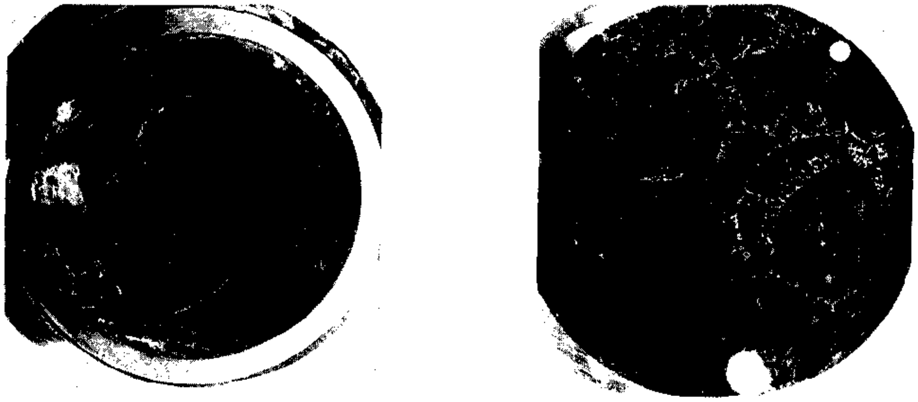
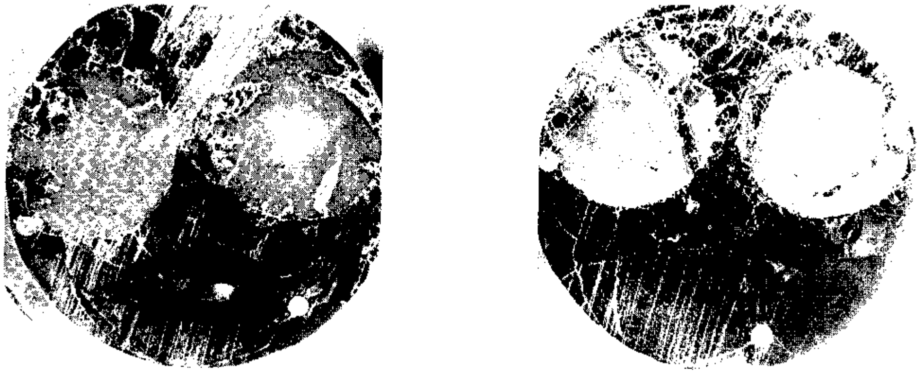


FIG. 6 GASES EVOLVED AT 850°C - WITH FLUX (Run 12)



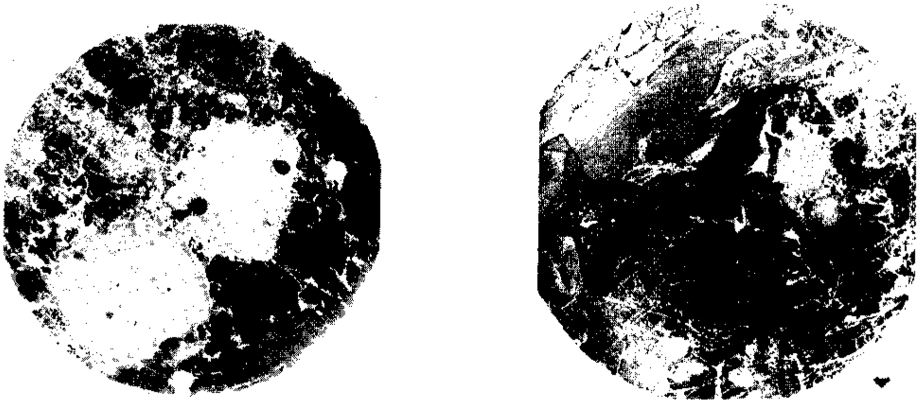
NEG. 66618
2 Inches from Bottom

NEG. 66619
4 Inches from Bottom



NEG. 66620
5 Inches from Bottom

NEG. 66621
6 Inches from Bottom



NEG. 66623
8 Inches from Bottom

NEG. 66624
9 Inches from Bottom

Ceramic Pellets were Placed in the Crucible Side by Side
for a Total of ~5 Inches, Starting from the Bottom.

FIG. 7 EXTENT OF CERAMIC DISSOLUTION AT END OF RUN 5

Losses to the Residue

Analysis of the residues showed that 1 to 2% of the T_2 was retained in the flux and was unextractable (Runs 9, 11, and 12). Although the samples analyzed were taken from various locations in each of the crucibles, they agreed within 1% for any one run, indicating a uniform concentration. The T_2 is probably retained by exchange with the traces of water remaining in the flux.

Suspending Targets

The targets must be suspended off the bottom of the crucible in order to uniformly dissolve the $LiAlO_2$ in flux. Otherwise the dissolution rate of the ceramic on the bottom of the crucible will decrease as the denser flux containing dissolved $LiAlO_2$ settles to the bottom. Figure 8 shows the remains of a target after 10 hours at 850°C in flux (Run 4). The crucible and most of the flux has been removed from the ceramic residue except for the bottom 1 inch. The bottom 4 inches of ceramic dissolved much slower than the remainder of the target. Suspending the target on a grate 4 inches above the bottom of the crucible gave a much more uniform dissolving rate (Figure 7).

NEG. 9522

FIG. 8 EXTENT OF CERAMIC DISSOLUTION AT END OF RUN 4

Evolution of H_2, CO, and CO_2

As shown in Table IV, during the extraction of tritium with untreated flux, 9 GVR of H_2 and 12 GVR of $CO+CO_2$ are also evolved (Run 8). Pretreating the flux at 850°C for 11 hours in a vacuum reduced the H_2 and $CO+CO_2$ evolution to about 1.5 and 7.0 GVR, respectively (Runs 11 and 12). Continued vacuum drying of the flux at 850°C removed very little additional water, as interpreted in Figure 9 from the data of Run 2. Attempts to pretreat at 1000°C caused the flux to decompose (Figure 10). All of these experiments with flux were conducted in carbon steel crucibles.

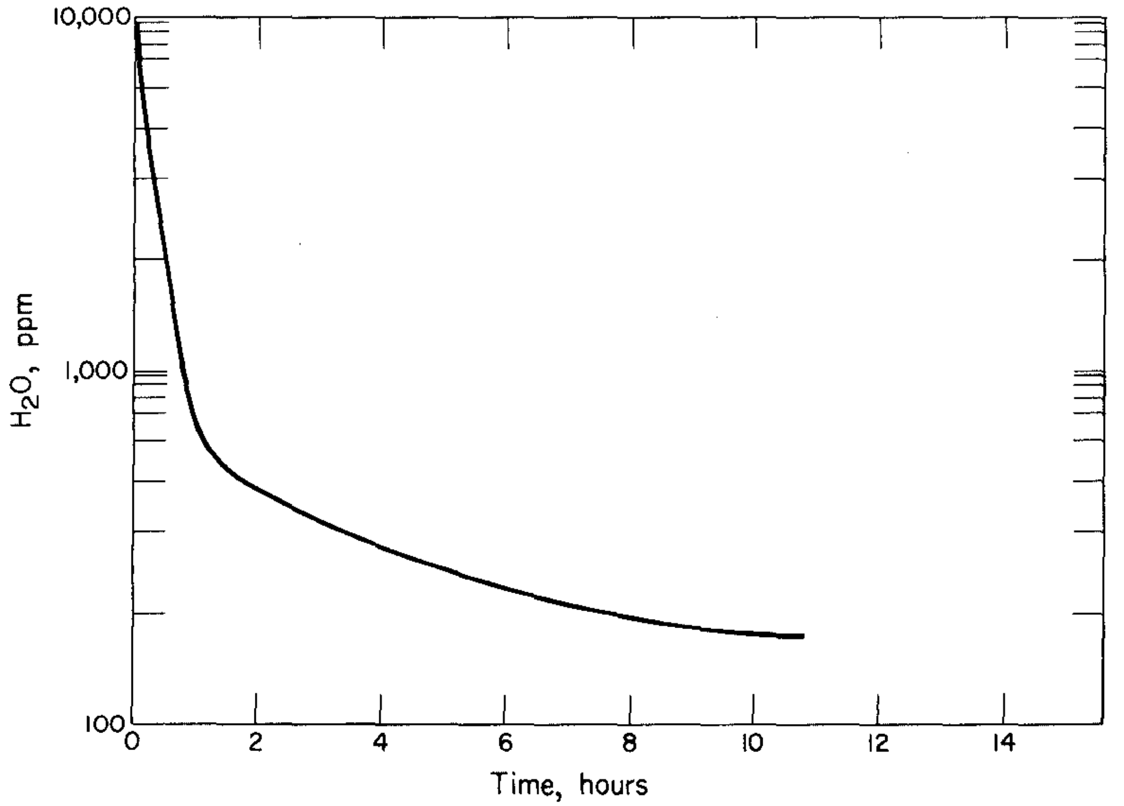
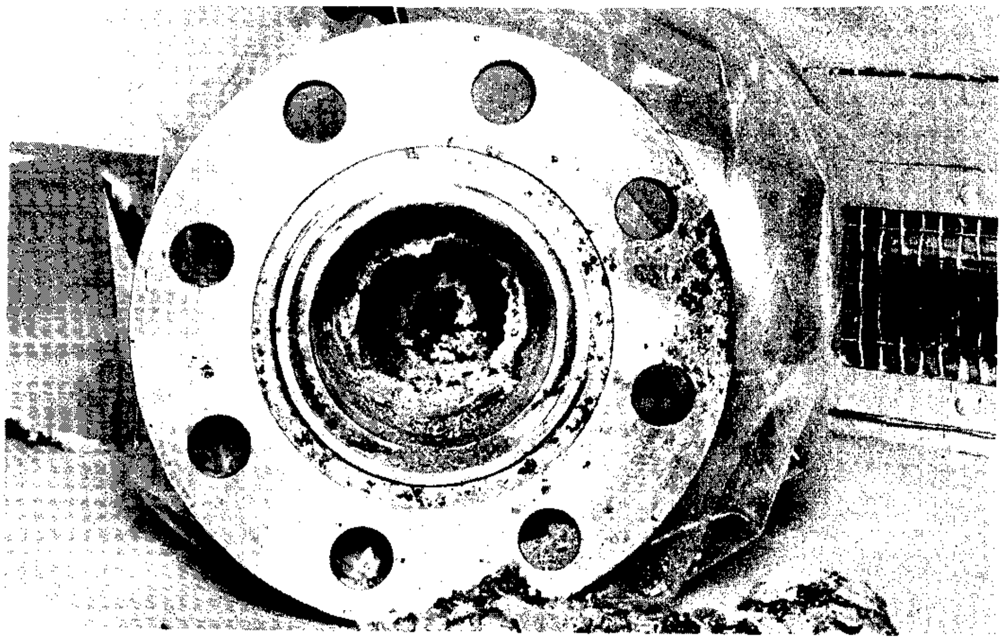


FIG. 9 H_2O LEFT IN $Na_2B_4O_7$ AS A FUNCTION OF TIME AT 850°C IN A VACUUM



NEG. 9460

FIG. 10 INTERNAL VIEW OF FURNACE SHOWING DECOMPOSED $Na_2B_4O_7$

The later realization that the crucibles add both carbon oxides and water in the amounts suggested by Table III has not been included in the data; e.g., approximately 2.5 GVR of CO and CO_2 should be subtracted from the total as the contribution from the carbon steel crucible.

Corrosion

The maximum corrosion rate of both the stainless steel and carbon steel crucibles was about 5 mils in 20 hours, close to that observed by PNL; no localized attack was observed.

HTO AND T_2O

As observed in Runs 8 and 11, 75% of the tritium is evolved as water vapor when untreated flux is used, whereas pretreating the flux reduces the fraction to 40%; this vapor can be readily decomposed by hot uranium chips.

Use of Lower Ratio of Flux to Ceramic

One run was made (Run 6) using a flux-to-ceramic ratio of 2 instead of the usual 4. The amount of ceramic material dissolved or severely attacked by the flux varied considerably depending upon the location of the ceramic in the crucible, as shown in Figure 11. There was a large void around the top three inches of ceramic material thus accounting for the low dissolution rate at that location. The initial release of gases from the target probably acted as an air lift and moved some of the flux into the top, cold, region of the furnace where it solidified. The poor dissolution over the bottom 3 inches indicates again the need for suspending the material off the bottom of the crucible. The results of this run emphasized that a flux-to-ceramic ratio of 4 is necessary to ensure that the targets will be completely surrounded by molten flux.

Analyses of Sodium Tetraborate

A variety of chemical analyses were used to confirm that:

- The decomposition products of the sodium tetraborate which was heated to 1000°C (Run 2d) in vacuum included metallic sodium.

[redacted]

NEG. 66648
1 Inch from Bottom

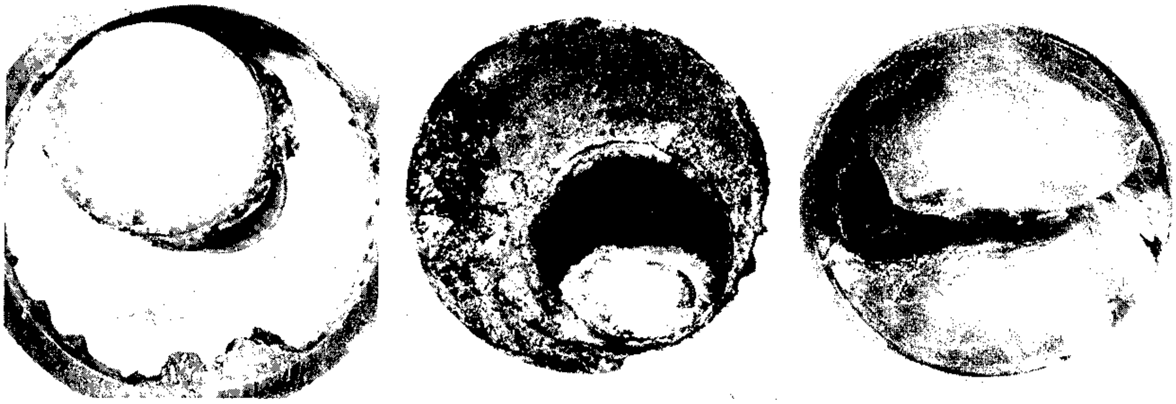
NEG. 66650
3 Inches from Bottom

NEG. 66651
4 Inches from Bottom

NEG. 66653
6 Inches from Bottom

NEG. 66655
8 Inches from Bottom

NEG. 66657
10 Inches from Bottom



NEG. 66659
12 Inches from Bottom

NEG. 66660
13 Inches from Bottom

NEG. 66661
View from Top of Crucible

FIG. 11 EXTENT OF CERAMIC DISSOLUTION AT END OF RUN 6

[redacted]

- The "anhydrous" $Na_2B_4O_7$ contains approximately 1% water that is released between 850 and 1000°C, and contains unstable impurities that liberate significant quantities of CO_2, CO, and H_2.

- There is no selective leaching of lithium from lithium-aluminate targets by the $Na_2B_4O_7$ flux.

- The carbon content of $Na_2B_4O_7$ from several sources varied from 25 to 350 ppm.

Disadvantages of Using Flux

There appear to be no advantages in using flux to extract tritium from lithium aluminate to offset the numerous disadvantages listed below:

- The loss of tritium to the flux will run between 1 and 2% compared to <0.1% extracting without flux.

- $Na_2B_4O_7$ which is low in moisture content may be difficult to obtain; new equipment and techniques would be required for charging the flux to the production furnace.

- The capacity of a furnace charge is reduced markedly by the large volume occupied by the flux.

- The amount of water vapor evolved when using flux is many times greater than without flux, thus requiring additional uranium decomposers.

- The amount of $CO+CO_2$ evolved during the extraction of tritium with flux is at least 10 times greater than without flux.