GEAP-4053-2

AEC RESEARCH AND
DEVELOPMENT REPORT

July 1, 1962

ACCURATE NUCLEAR FUEL BURNUP ANALYSES

SECOND QUARTERLY PROGRESS REPORT

March - May, 1962

by

B. F. Rider

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SUMMARY

Work has continued on the Development of Accurate Nuclear Fuel Burnup Analysis. The work performed during this quarter is summarized as follows:

1. Approval for irradiation of Capsule GEV-1 was obtained assigning it to the MTR.

2. Design of capsule GEV-1 was completed.

3. The chemistry of molybdenum was studied using Mo-99 as a tracer. Several possible solvent extraction procedures were studied as a function of acidity, time of contacting and reagent concentration.

4. Mass spectrometric behavior of molybdenum was investigated and found to be somewhat difficult. A method of carbon reduction of molybdenum oxide to metal on the filament was developed which greatly enhanced sensitivity for this stable fission product.

PROGRAM PROGRESS

Considerable progress has been made on the program during this quarter in three major areas. These include design of capsule GEV-1 for irradiation in MTR, chemical separation studies on molybdenum, and mass spectrometric behavior of stable molybdenum isotopes.

Design of GEV-1 Irradiation Assembly

This assembly consists of a capsule holder approximately 8" in length which contains three 2-1/2" capsules each containing one target foil. The capsule holder is a 1-1/8" O.D. aluminum tube into which twelve slots are cut to permit cooling of the outside surfaces of three foil-containing capsules.

The three foil capsules are fabricated from 0.937 O.D. stainless steel tubing with stainless end caps fusion welded to each end of the tubing. The foils are inserted between segments of aluminum machined to fit snugly between the foils and the I.D. of the stainless steel tubing. A stainless steel spring is placed at one end of each capsule to place a force against the aluminum segments assuring assuring close contact between all heat transfer surfaces. Flux wires of nickel, cobalt, iron, rhodium, and manganese are placed in holes within the aluminum segments.

The end caps are welded to the capsule within a helium atmosphere. After welding, the capsules were leak-checked, using mass spectrometric techniques, and no leak was detectable.

Details of the foil capsule design can be seen in drawing 117B1068, and the capsule holder in drawing 117B1076.

The capsules contain 1.5 gm. of U-233, 0.6 gm. of U-235 and 1.0 gm. of Pu-239, and are to be irradiated in a thermal flux of 2×10^{13} nv for three MTR cycles.

DRAWING NO 117B1068 CONT ON SHEET SH NO

GENERAL ELECTRIC

117B1068 CONT ON SHEET SH NO

UNLESS OTHERWISE SPECIFIED USE THE FOLLOWING:—

APPLIED PRACTICES	SURFACES	TOLERANCES ON MACHINED DIMENSIONS: FRACTIONS	DECIMALS	ANGLES
	32	± 1/64	±.002	±0°-30'

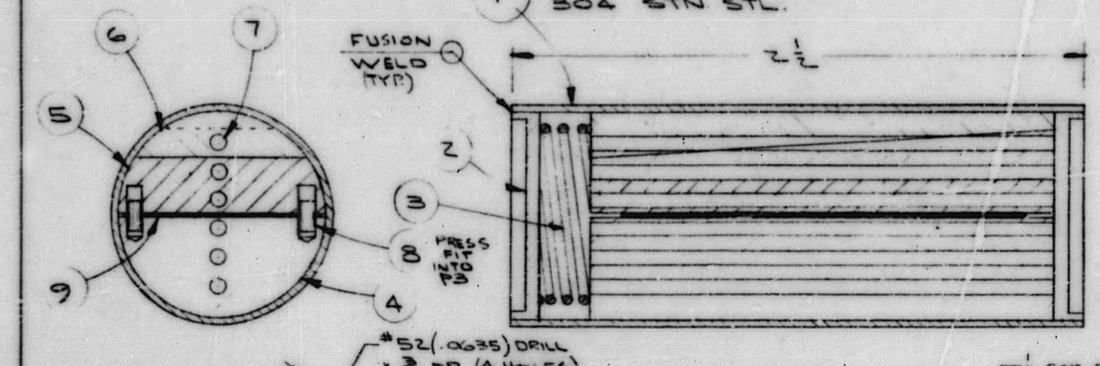
REV. NO. 1

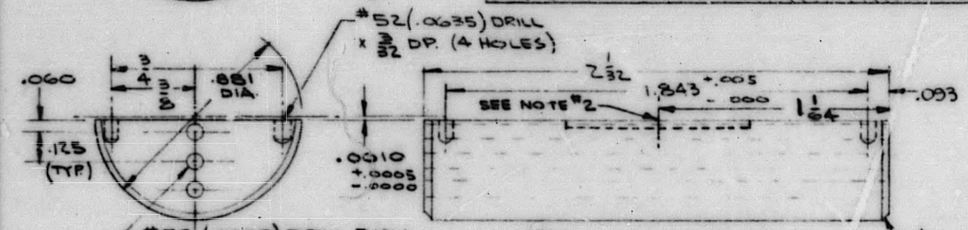
117B1068 CONT OF SHEET SH NO

TITLE: CHEMISTRY FOIL CAPSULE

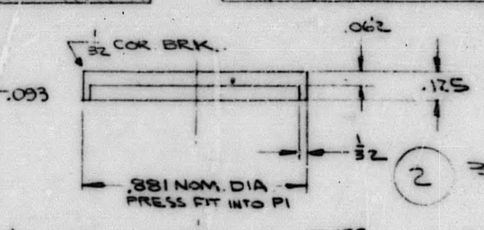
FIRST MADE FOR

QUAN. PT. G1	PT. NO.	NAME	DESCRIPTION
1	1	TUBE	SEE DETAIL
2	2	END CAP	
1	3	SPRING	
1	4	SEGMENT	
1	5	WEDGE	
1	6	WEDGE	SEE DETAIL
6	7	FLUX WIRE	.062 DIA. x 2" Lg.
4	8	ROLL PIN	1/16 DIA x 1/8 Lg STN. STL.
1	9	FOIL	a) U233: 0.875" x 1.75" x 0.0030" b) U235: 0.750" x 1.75" x 0.0012" c) PU: 0.812" DIA. x 0.022" - 10 MILS AL CLAD OVER 0.0010" x 0.750" AL/Pu ALLOY

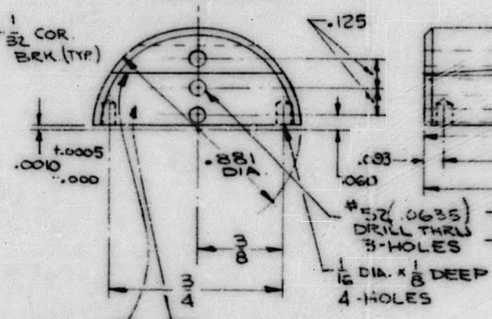




4 6061-T6 ALUM.



2 304 STN. STL.



6 5 6061-T6 ALUM.

3 COMPRESSION SPRING
I.D. = 23/32
WIRE DIA. = .031
NUMBER OF ACTIVE COILS = 3
FREE LENGTH = 9/32
ENDS SQUARED & GROUND
STN. STL.

REVISIONS		PRINTS TO
1	ADDED NOTE #2 REV. P3 DIMS. [illegible] W.E.S. 6/21/62	

△1
2. 0.812" DIA. x 0.010" DP. HOLE FOR Pu FOIL ONLY.
1. DEGREASE AFTER MACHING.
NOTE:

MADE BY	DATE	APPROVALS	DIV OR DEPT	LOCATION	117B1068 CONT ON SHEET	SH NO
T. Beck	MAR. 16, 1962	C.C. [illegible]	APED	VAL		
ISSUED					CHK'D.	

FF-506-P (3-61) REV. PRINTED IN U. S. A.

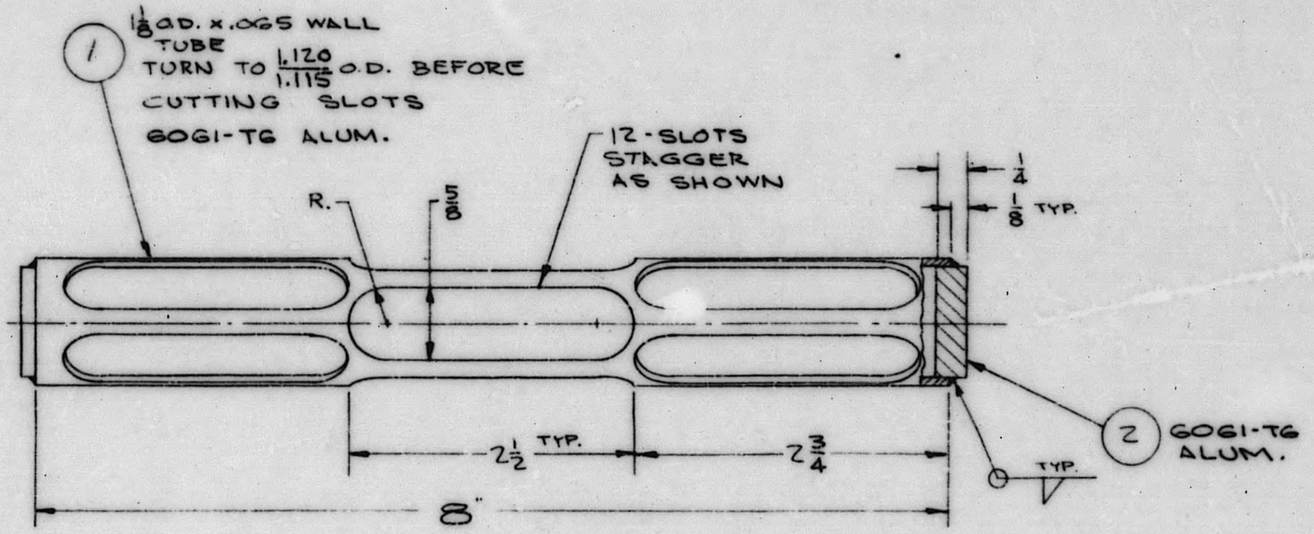
DRAWING NO. 117B1076 SH NO. CONT ON SHEET

GENERAL ELECTRIC

117 B 1076 CONT ON SHEET SH NO.

UNLESS OTHERWISE SPECIFIED USE THE FOLLOWING:—					REV NO. 0	TITLE
APPLIED PRACTICES	SURFACES	TOLERANCES ON MACHINED DIMENSIONS			117 B 1076	MTR CAPSULE HOLDER
		FRACTIONS	DECIMALS	ANGLES	CONT ON SHEET SH NO.	FIRST MADE FOR
	✓	±	±	±		

QUAN	PT NO	NAME	DESCRIPTION
1	1	TUBE	SEE DETAIL
2	2	PLUG	SEE DETAIL



REVISIONS	PRINTS TO

MADE BY E W Mahn April 17-62 ISSUED 7-6-62

APPROVALS 4/19/62

APED DIV OR DEPT

VAL LOCATION

117 B 1076 CONT ON SHEET SH NO.

CANCELLED

FF-005-P (3-61) REV. PRINTED IN U. S. A.

Pressure stresses are calculated not to exceed 17,900 psi and thermal stress not to exceed 7,500 psi. Allowed total stress for 304 stainless @ 200°F is 25,000 psi. Heat transfer calculations for the U-233, U-235 and Pu-239 capsules show capsule surface temperatures of 134°F, 125°F, and 152°F while fuel temperatures are 361°F, 240°F, and 729°F, respectively. The fissionable material is calculated to be 0.60, 0.24 and 0.40 gms/inch which will generate 317, 160, and 300 watts per inch at heat flux ratings of 65,000, 32,800 and 124,000 BTU/hr-ft^2, respectively.

The thermal and hydraulic calculations have been made assuming a conservative (maximum) gap between other foils, aluminum segments, and stainless steel tubing. The stress calculations have been made assuming a conservative (minimum) gap between the same components.

This assembly will be irradiated in a highly thermal flux in the beryllium reflector of the MTR and cadmium ratio for the position will be furnished by the MTR staff.

Chemistry of Molybdenum

Concurrent with the design of the capsule chemical studies on some of the most promising stable fission products have been carried on. Studies on the solvent extraction behavior of molybdenum in alpha benzoin oxime, bromo-8-hydroxyquinoline, and acetyl acetone were conducted.

Alpha benzoin oxime reagent was prepared by dissolving 2 grams of the reagent in 100 ml of ethanol. To ten milliliter samples containing Mo-99 tracers and various nitric acid molarities were added 150 microliters of reagent and a few crystals of $KBrO_3$ to insure that molybdenum would attain a valence of six. An equal volume of chloroform was added and the phases contacted for various times.

Figure 1 shows recovery of molybdenum as a function of nitric acid molarity and time of contacting. Recoveries are good up to 0.5 Molar nitric acid but fall off at molarities as high as 2 M. Recovery improved with time of contacting up to 10 minutes.

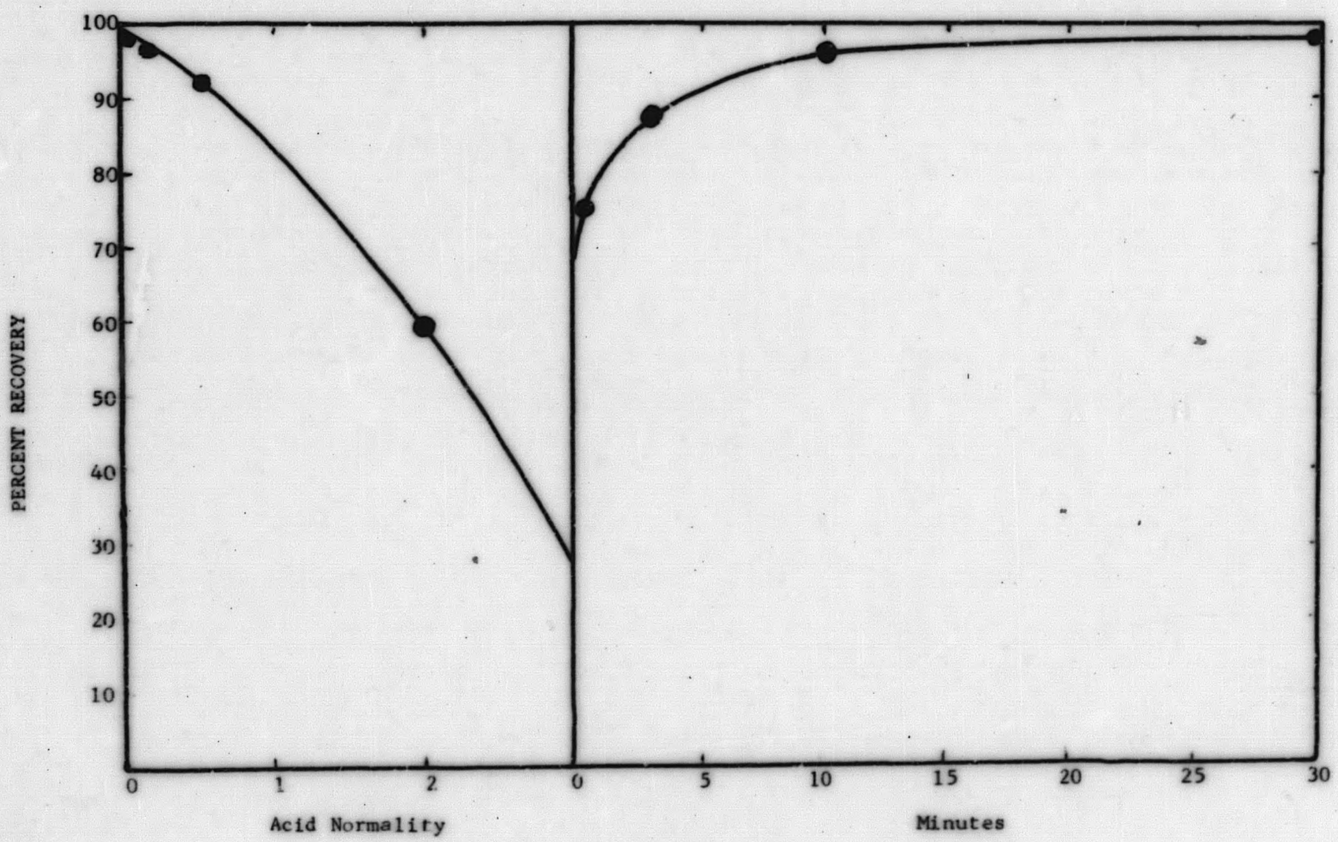


FIGURE 1. MOLYBDENUM EXTRACTION BY α BENZOIN OXIME

While tracer Mo-99 was available, other extractants were also investigated. Diethyl ether was found to have low extraction efficiency. Distribution ratios between ether and hydrochloric acid solutions were found to be 0.06 at 4N, 0.7 at 6N, and 0.4 at 9N. On the other hand, a 1% solution of 8-hydroxyquinoline in chloroform was found to extract better than 98% of molybdenum from an equal volume of aqueous phase containing up to 0.2N HCl but the recovery of Mo fell to 80% at 1N HCl. Even better extraction was found to be obtainable by use of bromo-8-hydroxyquinoline.

In this case, the bromo-8-hydroxyquinoline reagent was prepared by dissolving 0.1 gm. of reagent in 100 ml of chloroform. One volume of reagent was contacted with one volume of aqueous sample made 0.1, 0.5, 1, and 2N in HCl or HNO_3. Recoveries in all cases were greater than 90% in 5 minutes and 95% in 10 minutes. Only at higher acidities of 3N and 4N did recoveries fall to 80% and 40%, respectively.

The last reagent tested was acetyl acetone in chloroform. This extractant was found to be sensitive to time of extraction, percent by volume of reagent in chloroform and acidity of the sample. Figure 2 shows how the recovery increases with time of contacting from 40% recovery within 30 seconds, increasing to 90% at 30 minutes, and 98% in 60 minutes from 6M H_2SO_4 using a 1:1 acetyl acetone to chloroform organic phase.

Decreasing the reagent to chloroform ratio to 1:4 reduces the ultimate recovery to about 90% and a 1:10 ratio of reagent to chloroform further reduces recovery to 60%. Compromising at 30 minutes as a practical contacting time, a study of H_2SO_4 normality on recovery showed 98% recovery at 3N, 90% at 6N, and 3% at 9N. In all cases, a few crystals of $KBrO_3$ was added.

From this work, acetyl acetone diluted 1:1 with chloroform appears to be one of the best extractants for molybdenum because it is effective at acidities as high as 6N H_2SO_4 where specificity is greatest. Adequate

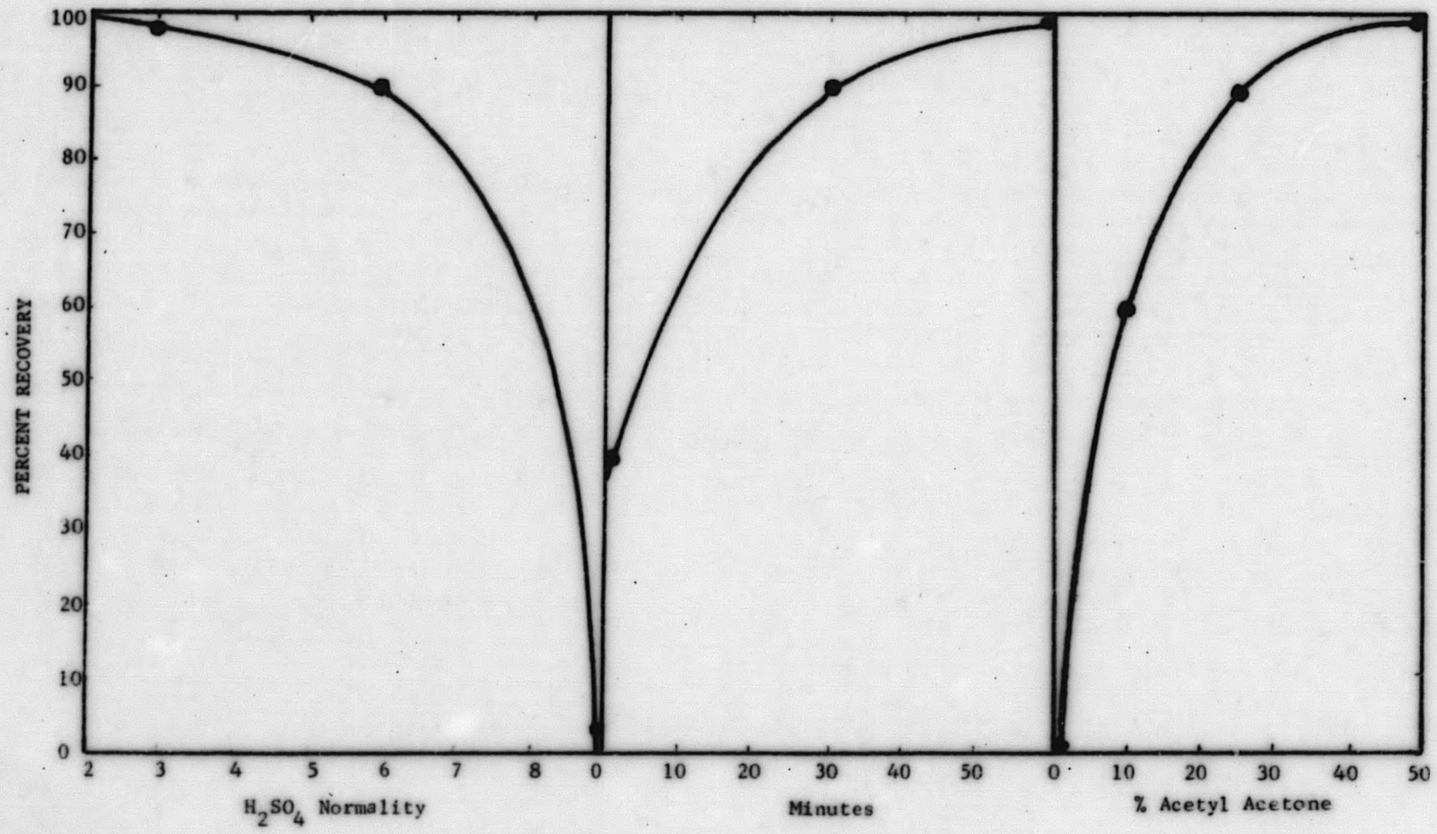


FIGURE 2. MOLYBDENUM EXTRACTION WITH ACETYL ACETONE

recoveries can be made in a few minutes, but increase if somewhat longer contacting times are allowed.

More work is planned for alpha benzoin oxime and acetyl acetone, especially in the presence of radioactive fission products.

Mass Spectrometry of Molybdenum

Mass spectrometry of molybdenum was found to be much poorer than for neodymium, for instance. Beam currents were small and fell off rapidly. As a result very large samples, in excess of 50 nanograms were initially required and emission was achieved only at very high filament temperatures, over 2000°C. It was reasoned that this difficulty arises from the fact that nitric acid samples of molybdates loaded onto filaments are converted to MoO_3 during initial heating of the filaments. Attempts to reduce the MoO_3 to metal by addition of colloidal carbon to the filament were not at first effective, presumably because the volatile MoO_3 sublimed from the filament before it was reduced to metal. However, heating the sample plus carbon on the filament in the instrument at 500°C overnight before increasing the temperature was successful in achieving adequately long and steady runs on as little as 5 nanograms of molybdenum.

Work on mass spectrometry will continue with initial emphasis on molybdenum and neodymium because of the freedom of radiation hazard on these elemental fractions.

END