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MASTER

SNAP PROGRAMS
Quarterly Progress Report No. 6
Task 5
Thermionic Isotopic Power Systems
January 1 through March 31, 1961
MND-P-3014-II

Approved by

Wilfred L. Kershaw
Project Engineer

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FOREWORD

This is the sixth unclassified SNAP Quarterly Progress Report prepared by The Martin Company. It covers the work performed during the period January 1, 1961, through March 31, 1961, under Contract AT(30-3)-217, Task 5, Thermionic Isotopic Power Systems, for the U. S. Atomic Energy Commission.

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SUMMARY

The Martin Company is currently under AEC contract to design, fabricate and test power supplies utilizing radioisotopes as heat sources. The present systems rely on thermoelectric generators to convert the thermal energy produced by the isotope heat source into useful electrical power. The isotopes utilized in these systems are, generally, those readily available today. Many future missions can best be satisfied through the use of isotope heat sources not generally available today and by employing thermionic energy conversion.

The purpose of Tasks 5 and 6 of Contract AT(30-3)-217 is to carry out the development required before thermionic conversion can be utilized. The thermionic technology is incorporated in Task 5, and the fuel technology, in Task 6. This report covers only the work on Task 5; Task 6 is reviewed in a separate report.

Since the last quarterly report, work has been initiated on two new subtasks under Task 5. Subtask 5.2, Cesium Diode Technology, has been reinstated. The purpose of this work is to develop a cesium thermionic system that can perform satisfactorily at relatively low temperatures (1600° to 1800° K). Work is advancing in the following three areas:

- (1) Parametric study.
- (2) Design, fabrication and testing of Test Units 1 and 2 to verify the parametric study.
- (3) Design, fabrication and life testing of Test Unit 3.

Subtask 5.4, Curium Fuel Capsule for Vacuum Diode Generator, was also initiated this quarter. Its ultimate objective is to fabricate a suitable curium fuel capsule for the generator being developed under Subtask 5.3. Effort this quarter consisted principally of:

- (1) Materials survey.
- (2) Thermal analysis.
- (3) Preliminary design of the capsule.

Work was continued on Subtask 5.3, Vacuum Diode Generator, with the emphasis on the development of Generator 2B. Several significant design changes were effected this period. The sapphire spacer rods were replaced by a peripheral ring support to provide the desired inter-electrode spacing. Preliminary tests indicate that the use of an oxidized

molybdenum collector may be an effective way of attaining and maintaining a collector work function less than 1.9 volts. Further testing is required before this idea can be incorporated into Generator 2B. This requirement has delayed the completion of Generator 2B; however, the anticipated improvement in performance offsets the delay.

Work during this quarter on Subtask 5.3 has also included:

- (1) Investigation of other electrode materials.
- (2) Study of the effects of patchy electrode surfaces on the performance of a close-spaced diode.
- (3) Preparations for the dynamic testing of Generator 2B.

I. INTRODUCTION

This is the third Quarterly Progress Report required under Contract AT(30-3)-217 for Fiscal Year 1961, including an appraisal of work on Task 5, Thermionic Isotopic Power Systems (SNAP-TIP Systems).

The SNAP programs covered by The Martin Company are divided into separate tasks and subtasks, each task being separately administered. Task 5 is directed by R. J. Harvey, who prepared this report, based on principal contributions by:

- (1) Thermo Electron Engineering Corporation.
- (2) Dr. J. Gumnick, RIAS and Loyola College.
- (3) Dr. B. Weigman, Loyola College.
- (4) G. Storrs, T. Bustard, J. Kane and B. Callender of The Martin Company.

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II. TASK 5--THERMIONIC ISOTOPIC POWER SYSTEMS

Presently, there are three active subtasks under Task 5:

- (1) 5.2--Cesium Diode Generator Development.
- (2) 5.3--Vacuum Diode Generator Development.
- (3) 5.4--Curium Fuel Capsule Development.

A. SUBTASK 5.2--CESIUM DIODE GENERATOR DEVELOPMENT

1. Objectives

The objectives of this subtask are to investigate, both analytically and experimentally, the characteristics of cesium-filled thermionic converters operating in the low temperature region (1600° to 2000° K). This work is being performed by Thermo Electron Engineering Corporation (TEEC) of Waltham, Massachusetts, as a subcontractor to The Martin Company.

2. Scope of Work

Subtask 5.2 consists of three sub-subtasks. During the quarter, work was performed on each of the sub-subtasks:

- (1) Parametric study (5.2.1).
- (2) Design, fabrication and testing of Units 1 and 2 (5.2.2).
- (3) Design, fabrication and life testing of Unit 3 (5.2.3).

Work on all three sub-subtasks is continuing into the following quarter.

3. Technical Discussion

a. Parametric study (5.2.1)

The purpose of the parametric study is to investigate the effects of various parameters on the performance of cesium-filled thermionic generators. This investigation is analytical and will result in the compilation of a number of tables showing the interdependence of the various converter parameters. During this quarter two rather extensive tables have been constructed (Tables 1 and 2).

Parametric Study of the Cesium Thermionic Diode with a Tantalum Emitter and no Back Emission

Coverage fraction, θ	Work function, ϕ (volts)	Dependent parameters	Emitter Temperatures, T_E ($^{\circ}\text{K}$)																				
			1500	1550	1600	1650	1700	1750	1800	1850	1900	1950	2000	2050	2100	2150	2200	2250	2300	2350	2400	2450	2500
0.05	3.78	μ_{Cs} } for attainment of T_{Cs} } work function reduction μ_{Cs} } for space charge neu- T_{Cs} } tralization for given current I_s } saturation current λ } mean free path for coverage pressure							—	—	1.42×10^{19} 482 2.77×10^{15} 324 0.0403 1.3×10^{-2}	2.2×10^{19} 495 3.8×10^{15} 328 0.0745 8.4×10^{-3}	3.1×10^{19} 505 7.26×10^{15} 336 0.142 6×10^{-3}	4.5×10^{19} 516 1.3×10^{16} 345 0.258 4.4×10^{-3}	6.4×10^{19} 528 2.3×10^{16} 352 0.46 2.9×10^{-3}	8.8×10^{19} 539 4.05×10^{16} 360 0.805 2.1×10^{-3}	1.2×10^{20} 550 6.5×10^{16} 368 1.3 1.5×10^{-3}	1.6×10^{20} 560 1.1×10^{17} 376 2.16 1.2×10^{-3}	2.1×10^{20} 571 1.7×10^{17} 384 3.35 8.8×10^{-4}	2.8×10^{20} 582 2.6×10^{17} 293 5.18 6.5×10^{-4}	3.6×10^{20} 592 4.1×10^{17} 400 8.2 5×10^{-4}	4.6×10^{20} 603 6×10^{17} 408 12 4×10^{-4}	5.9×10^{20} 612 9×10^{17} 417 17.9 3×10^{-4}
0.10	3.46	μ_{Cs} (atoms/cm ² sec) T_{Cs} ($^{\circ}\text{K}$) μ_{Cs} (atoms/cm ² sec) T_{Cs} ($^{\circ}\text{K}$) I_s (amp) λ (cm)							1.21×10^{19} 477 2.67×10^{16} 355 0.0725 16×10^{-2}	1.85×10^{19} 490 4.56×10^{16} 363 0.155 1×10^{-2}	3×10^{19} 503 6.9×10^{16} 374 0.279 6.4×10^{-3}	4.4×10^{19} 515 1.46×10^{17} 381 0.498 3.8×10^{-3}	6.6×10^{19} 530 2.42×10^{17} 390 0.902 2.7×10^{-3}	8.1×10^{19} 536 4×10^{17} 400 1.6 2.3×10^{-3}	1.15×10^{20} 548 6.6×10^{17} 410 2.73 1.6×10^{-3}	1.6×10^{20} 560 1.02×10^{18} 420 4.45 1.2×10^{-3}	2.2×10^{20} 572 1.5×10^{18} 430 6.9 8.6×10^{-4}	3.1×10^{20} 585 1.9×10^{18} 435 11 6.2×10^{-4}	4×10^{20} 596 3.2×10^{18} 444 16.4 4.7×10^{-4}	5.3×10^{20} 608 4.6×10^{18} 451 24.5 3.5×10^{-4}	7.1×10^{20} 620 6.8×10^{18} 461 38.3 2.6×10^{-4}	9.1×10^{20} 632 9.6×10^{18} 470 55 2×10^{-4}	1.2×10^{21} 645 1.35×10^{19} 480 79 1.5×10^{-4}
0.15	3.17	μ_{Cs} T_{Cs} μ_{Cs} T_{Cs} I_s λ					1.19×10^{19} 478 4.1×10^{17} 400 0.136 1.5×10^{-2}	1.19×10^{19} 490 7.1×10^{17} 410 0.279 1×10^{-2}	3×10^{19} 504 1.21×10^{18} 425 0.526 6×10^{-3}	4.8×10^{19} 520 1.9×10^{18} 434 0.961 3.7×10^{-3}	1.1×10^{20} 532 3.04×10^{18} 443 1.7 2.5×10^{-3}	1.51×10^{20} 558 4.69×10^{18} 464 4.96 1.7×10^{-3}	2.2×10^{20} 570 7.28×10^{18} 475 8.16 1.3×10^{-3}	3.1×10^{20} 585 1.1×10^{19} 486 13.5 9×10^{-4}	4.4×10^{20} 600 2.5×10^{19} 498 21.7 6×10^{-4}	5.9×10^{20} 612 3.4×10^{19} 508 32.2 4×10^{-4}	7.7×10^{20} 624 4.8×10^{19} 518 49.5 3.1×10^{-4}	1.04×10^{21} 637 6.7×10^{19} 530 73 2.4×10^{-4}	1.4×10^{21} 653 8.8×10^{19} 540 103.5 1.3×10^{-4}	1.8×10^{21} 667 1.3×10^{20} 553 162 9.6×10^{-5}	2.3×10^{21} 680 1.6×10^{20} 560 218 7.5×10^{-5}	2.9×10^{21} 695 2.1×10^{20} 570 304 5.5×10^{-5}	
0.20	2.9	μ_{Cs} T_{Cs} μ_{Cs} T_{Cs} I_s λ	4×10^{18} 450 1.83×10^{18} 433 0.0387 4.5×10^{-2}	7.4×10^{18} 465 4.03×10^{18} 449 0.0988 2.5×10^{-2}	1.28×10^{19} 480 6.16×10^{18} 460 0.207 1.5×10^{-2}	2.24×10^{19} 495 1.05×10^{19} 473 0.434 8×10^{-3}	3.7×10^{19} 510 1.78×10^{19} 489 0.814 5×10^{-3}	5.86×10^{19} 526 2.46×10^{19} 498 1.61 3.2×10^{-3}	9.4×10^{19} 542 3.8×10^{19} 512 3 1.9×10^{-3}	1.45×10^{20} 557 5.77×10^{19} 523 5.26 1.3×10^{-3}	2.14×10^{20} 571 8.32×10^{19} 533 9.2 8.8×10^{-4}	3.16×10^{20} 586 1.17×10^{20} 548 14.9 6×10^{-4}	4.45×10^{20} 600 2.4×10^{20} 560 24.8 4.2×10^{-4}	6.5×10^{20} 615 2.4×10^{20} 576 38 2.8×10^{-4}	8.9×10^{20} 630 3.2×10^{20} 586 58 2.1×10^{-4}	1.2×10^{21} 645 4.6×10^{20} 600 92.5 1.5×10^{-4}	1.6×10^{21} 660 5.5×10^{20} 610 135 1.1×10^{-4}	2.2×10^{21} 678 7.5×10^{20} 623 199 7.8×10^{-5}	2.9×10^{21} 695 9.5×10^{20} 635 275 5.6×10^{-5}	3.7×10^{21} 710 1.2×10^{21} 645 390 4.2×10^{-5}	4.8×10^{21} 728 1.6×10^{21} 660 580 2.8×10^{-5}	6×10^{21} 740 2.05×10^{21} 670 783 2.3×10^{-5}	
0.25	2.65	μ_{Cs} T_{Cs} μ_{Cs} T_{Cs} I_s λ	1.23×10^{19} 477 9.43×10^{19} 542 0.279 1.6×10^{-2}	2.25×10^{19} 495 1.82×10^{20} 566 0.663 8×10^{-3}	3.9×10^{19} 512 2.79×10^{20} 582 1.31 4.5×10^{-3}	6.45×10^{19} 529 3.57×10^{20} 591 2.6 2.9×10^{-3}	1.11×10^{20} 547 5.35×10^{20} 608 4.65 1.7×10^{-3}	1.65×10^{20} 563 6.97×10^{20} 620 8.68 1.1×10^{-3}	2.62×10^{20} 580 9.88×10^{20} 636 15.5 7×10^{-4}	4×10^{20} 596 1.36×10^{21} 652 25.8 4.6×10^{-4}	5.9×10^{20} 613 1.74×10^{21} 665 43.4 3×10^{-4}	8.7×10^{20} 630 2.42×10^{21} 681 66.9 2.2×10^{-4}	1.23×10^{21} 647 3.24×10^{21} 702 104 1.5×10^{-4}	1.65×10^{21} 665 4.1×10^{21} 715 153 10^{-4}	2.3×10^{21} 682 5.5×10^{21} 735 232 7×10^{-5}	3.2×10^{21} 700 7.4×10^{21} 753 358 5×10^{-5}	4.2×10^{21} 720 8.3×10^{21} 762 500 3.5×10^{-5}	5.6×10^{21} 736 1.08×10^{22} 780 730 2.5×10^{-5}					
0.30	2.43	μ_{Cs} T_{Cs} μ_{Cs} T_{Cs} I_s λ	3.6×10^{19} 510 3.33×10^{21} 705 1.8 5×10^{-3}	6.45×10^{19} 528 5.42×10^{21} 734 3.92 2.9×10^{-3}	1.1×10^{20} 546 6.72×10^{21} 748 7.14 1.7×10^{-3}	1.86×10^{20} 566 9.23×10^{21} 770 13.4 1×10^{-3}	2.94×10^{20} 584 1.2×10^{22} 788 22.1 6.2×10^{-4}	4.55×10^{20} 601 1.43×10^{22} 800 39.2 4×10^{-4}	7.05×10^{20} 620 1.79×10^{22} 820 65.1 2.6×10^{-4}	1.05×10^{21} 657 2.29×10^{22} 838 104 1.7×10^{-4}	1.51×10^{21} 676 2.87×10^{22} 860 164 1.2×10^{-4}	2.18×10^{21} 698 3.39×10^{22} 875 242 8×10^{-5}	3.1×10^{21} 716 4.06×10^{22} 890 358 5.2×10^{-5}	4.3×10^{21} 740 5.2×10^{22} 915 537 3.8×10^{-5}	5.9×10^{21} 760 6.5×10^{22} 940 779 2.3×10^{-5}								
0.35	2.23	μ_{Cs} T_{Cs} μ_{Cs} T_{Cs} I_s λ	9.55×10^{19} 543 5.84×10^{22} 930 8.34 1.7×10^{-3}	1.66×10^{20} 562 1.025×10^{23} 995 16.7 1.1×10^{-3}	2.75×10^{20} 582 1.345×10^{23} 1035 30 6.8×10^{-4}	4.56×10^{20} 600 1.51×10^{23} 1070 52.6 4.2×10^{-4}	7.2×10^{20} 621 1.77×10^{23} 1095 85.2 2.5×10^{-4}	1.11×10^{21} 641 2.05×10^{23} 1155 145 1.7×10^{-4}	1.7×10^{21} 665 2.3×10^{23} 1170 232 1×10^{-4}	2.5×10^{21} 685 2.95×10^{23} 1190 356 6.8×10^{-5}	3.6×10^{21} 708 3.3×10^{23} 1210 576 4.4×10^{-5}												
0.40	2.07	μ_{Cs} T_{Cs} μ_{Cs} T_{Cs} I_s λ	2.57×10^{20} 577 8.91×10^{23} $T > 1400$ 30.4 7.2×10^{-4}	4.46×10^{20} 599 1.06×10^{24} 57.3 4.2×10^{-4}	6.9×10^{20} 620 1.23×10^{24} 106 2.6×10^{-4}	1.17×10^{21} 645 1.3×10^{24} 161 1.5×10^{-4}	1.86×10^{21} 660 1.48×10^{24} 260 1.1×10^{-4}	2.9×10^{21} 680 1.61×10^{24} 420 6×10^{-5}															

TABLE 2

Operational Characteristics in the
Collision-Free Region*

$$T_E = 1500^\circ \text{K}$$

Energy Losses Q (watts/cm ²)	Output Voltage V (volts)	Output Current I (amp/cm ²)	Output Power P (watts/cm ²)	Efficiency η^* (%)
5	0.468	6.3	2.7	12.8
10	0.406	10.3	4.2	11.8
15	0.371	13.1	4.9	10.4
20	0.346	16.5	5.7	9.5
25	0.328	18.7	6.15	8.8
50	0.276	28.5	7.85	6.7

$$T_E = 1550^\circ \text{K}$$

5	0.537	7	3.7	15.9
10	0.468	12	5.6	13.8
15	0.423	17	7.2	12.5
20	0.403	20	8.06	11.6
25	0.383	23	8.8	10.8
50	0.334	32	10.7	8.4

* Calculations for maximum efficiency. Collector work function
 ϕ_C constant at 1.81 volts.

$$* \eta = P / [I (\phi_C + V + 2kT_E) + Q]$$

TABLE 2 (continued)

 $T_E = 1600^\circ \text{ K}$

Energy Losses Q (watts/cm ²)	Output Voltage V (volts)	Output Current I (amp/cm ²)	Output Power P (watts/cm ²)	Efficiency η^* (%)
5	0.601	8.2	4.95	19.9
10	0.529	12.5	6.6	15.4
15	0.488	18	8.8	14.1
20	0.46	21	9.65	13
25	0.438	25	11	12.2
50	0.376	36.5	13.7	9.6
75	0.34	54	16	8.7

 $T_E = 1650^\circ \text{ K}$

5	0.674	8.4	5.7	20.1
10	0.596	15	9	17.8
15	0.552	21	11.6	16.5
20	0.523	26	13.6	15.9
25	0.500	30	15	14.6
50	0.431	49	21	12.1
75	0.391	64	25	10.7

TABLE 2 (continued)

 $T_E = 1700^\circ \text{ K}$

Energy Losses Q (watts/cm ²)	Output Voltage V (volts)	Output Current I (amp/cm ²)	Output Power P (watts/cm ²)	Efficiency η^* (%)
5	0.740	9.9	7.3	22
10	0.659	16.7	11	19.6
15	0.616	23	14.2	18.3
20	0.581	29	16.8	17.2
25	0.560	33	18.5	16.4
50	0.487	54	26.3	13.9
75	0.446	72	32	12.3
100	0.425	84	35.6	11.3

 $T_E = 1750^\circ \text{ K}$

5	0.810	10.9	8.9	24.1
10	0.730	18.2	13.3	21.5
15	0.680	25	17	20
20	0.650	30	19.5	19
25	0.623	37	23	18.2
50	0.543	63	34.2	15.8
100	0.471	100	47.1	13.3
200	0.403	160	64.5	10.6

TABLE 2 (continued)

 $T_E = 1800^\circ \text{K}$

Energy Losses Q (watts/cm ²)	Output Voltage V (volts)	Output Current I (amp/cm ²)	Output Power P (watts/cm ²)	Efficiency η * (%)
5	0.885	11.6	10.2	25.5
10	0.800	19.9	15.9	23.2
15	0.748	27.9	20.9	22
20	0.71	34	24.2	20.6
25	0.686	40.4	27.7	20
50	0.603	69	41.6	17.5
100	0.525	115	60.5	15
200	0.453	178	81	12.2

 $T_E = 1850^\circ \text{K}$

5	0.960	11.7	11.2	28.3
10	0.810	20.9	18.2	25
15	0.815	29	23.6	23.5
20	0.780	37.3	29.1	22.7
25	0.752	45	33.8	21.8
50	0.673	71.5	48	19.2
100	0.583	124	72.4	16.6
200	0.506	204	103	14

TABLE 2 (continued)

 $T_E = 1900^\circ \text{K}$

Energy Losses Q (watts/cm ²)	Output Voltage V (volts)	Output Current I (amp/cm ²)	Output Power P (watts/cm ²)	Efficiency η * (%)
15	0.891	31.2	27.8	25.3
20	0.851	39.3	33.5	24.3
25	0.821	47	38.6	23.5
50	0.731	80.6	59	21
100	0.643	138	88.8	18.3
200	0.559	226	126	15.5

 $T_E = 1950^\circ \text{K}$

15	0.953	32.5	31	26.9
20	0.918	40.7	37.3	25.7
25	0.887	50	44.4	25.2
50	0.791	87	69	22.6
100	0.701	149	104.5	19.8
200	0.615	250	153.5	17.2

TABLE 2 (continued)

$$T_E = 2000^\circ \text{ K}$$

Energy Losses Q (watts/cm ²)	Output Voltage V (volts)	Output Current I (amp/cm ²)	Output Power P (watts/cm ²)	Efficiency η * (%)
15	1.03	34.3	35.4	28.5
20	0.987	44	43.5	27.5
25	0.958	53	50.7	26.7
50	0.861	92.5	79.5	24.2
100	0.765	159	126.5	21.5
200	0.678	272	184	19

Table 1 gives emitter temperature from 1500° to 2500° K in 50-degree increments and the fractional coverage of cesium on tantalum from 0.05 to 0.40 in increments of 0.05. The emitter work function is considered a function of coverage only, independent of temperature. The various values are obtained with the aid of a semiempirical equation derived in Ref. 1:

$$\begin{aligned} \phi_A = & \phi_{Ta} - (\phi_{Ta} - \phi_{Cs}) \left[1 - (1 - 3\theta^2 + 2\theta^3) \right. \\ & + 3.12 \left(\frac{r_W}{r_{Ta}} \right)^2 (1 - 3\theta^2 + 2\theta^3) \theta \\ & \left. - 4.87 \left(\frac{\theta_{Ta} - \phi_{Cs}}{\theta_W - \phi_{Cs}} \right) \frac{r_W}{r_{Ta}} \theta^2 (1 - \theta)^2 \right] \end{aligned} \quad (1)$$

where

ϕ_A = apparent work function

$\phi_{Ta}, \phi_W, \phi_{Cs}$ = work functions of tantalum, tungsten and cesium, respectively

r_{Ta}, r_W = lattice spacings of tantalum and tungsten, respectively

θ = coverage.

Equation (1) seems to be the most satisfactory representation of experimental data obtained to date. Its derivation was presented at the March 1961 Conference of Physical Electronics at The Massachusetts Institute of Technology.

The saturation current of the emitter was calculated with the aid of the Richardson equation, assuming A , the thermionic emission constant, equal to $120 \text{ amp/cm}^2 \cdot \text{K}^2$.

The cesium pressure and corresponding temperature for the attainment of work function were tabulated for various emitter temperatures. The relation of atomic and ionic emission to particle arrival, as tabulated in Ref. 2, was used to determine the rate of atom arrival at the cesium bath. The corresponding cesium bath temperature was then obtained by means of Nottingham's equation (Ref. 3).

The atomic mean free path corresponding to each temperature was calculated by methods proposed by Nottingham. The mean free path, when related to the interelectrode spacing, produces a useful ratio which determines the region in which the converter operates. Generally, at large spacings, the converter will operate in the collision region; but if the spacing is smaller than 10 mean free paths, the converter will probably operate in the collision-free region.

In Table 1 a second cesium pressure has been tabulated. This is the pressure necessary to provide complete neutralization of the electron space charge. It is obtained from the Saha-Langmuir equation for the probability of ionization of cesium particles impinging on a metallic surface. These values, initially expressed in atom arrival rates, can be translated into temperatures and can be compared with those of the work function reduction.

Both of these cesium pressures are critical. The higher of the two must be used for efficient operation since only it will provide both work function reduction and space charge neutralization.

Table 2 lists the performance characteristics of cesium converters in the collision-free region. These characteristics have been arrived at by optimization of the efficiency of the converter for various fixed design losses independent of the electron cooling (i.e., radiation between emitter and collector, stray radiation and the various conduction losses due to the structural supports).

The efficiency can be expressed by the equation

$$\eta = \frac{VI}{I(V + \phi_C + 2kT) + Q} \quad (2)$$

where

- η = converter efficiency
- V = output voltage
- I = current
- ϕ_C = collector work function
- k = Boltzmann constant
- T = emitter temperature
- Q = design losses.

Differentiating this efficiency with respect to output voltage and setting the result equal to zero produce the output voltage for optimum efficiency as a function of design losses Q :

$$\frac{V}{I} \left(\frac{1}{kT} \right) - \frac{1}{I} - \frac{2kT + \phi_C}{Q} = 0 \quad (3a)$$

where

$$I = AT^2 \exp \left(- \frac{\phi_C + V}{kT} \right) \quad (3b)$$

The significance of this result is that the selection of the optimum design point depends on the losses incurred across the diode. If the radiation losses per square centimeter of emitter area were quite high, then the optimum performance point would be at a higher current and lower voltage than for the case of lower radiation losses. Essentially, the electron cooling term and power output must be increased in order to mask out the higher radiation loss. For example, with an operating temperature of 1500° K, design losses may be increased from 5 to 50 watts/cm² (an increase of one order of magnitude) while the efficiency decreases by less than half. The design of cesium converters is flexible in that high design losses can be offset to some degree by shifting the operating point.

b. Test Units 1 and 2 (5.2.2)

The design of Units 1 and 2 has started and, with the exception of a diode section, the test setup has been selected. It is shown in Fig. 1. The test setup for Units 1 and 2 is similar to that used for the life test unit, Unit 3. The basic differences among Units 1, 2 and 3 lie in the interelectrode spacing and the emitter material. After several attempts 1-mil spacing has been achieved in Unit 3. It is anticipated that the designs of Units 1 and 2 will be completed early in the next quarter.

c. Life Test Unit 3 (5.2.3)

Unit 3 was designed and constructed during this quarter, and preliminary testing will begin in April. A cross section of the diode assembly is shown in Fig. 2, and the assembled test setup is shown in Fig. 1. Figures 3 and 4 show the test unit during construction.

The emitter is a pure tantalum disc, and the collector, a pure molybdenum disc. Interelectrode spacing is achieved by means of a 0.003-inch thick tantalum cylinder and is of the order of 0.005 inch. The cylinder is electron-beam welded to the emitter and brazed to the base of the collector, which is electrically insulated by a ceramic braze. The diode assembly is brazed to the copper housing, which carries lead-throughs, an evacuation tube and a cesium reservoir tube. The bottom

224 018

Fig. 1. Typical Test Unit

224 016

Scale 5 x 1

Fig. 2. Life Test Unit 3

224 330

Fig. 3. Test Section Parts

224 021

Fig. 4. Test Setup

224 022

of the housing is connected by three copper rods to a cooling coil, with water as the coolant. For better control of the diode housing temperature, a heating coil is wrapped around its lateral surface. The cesium reservoir is also connected to the cooling fixture by a copper strap, and a heating coil is wrapped around the reservoir. The cesium temperature may be varied from 50° to 350° C.

The emitter is heated by electron bombardment from a tungsten filament suspended above the unit. Through the use of an optical pyrometer, the emitter temperature is determined by measuring the temperature of a cavity provided in the side of the emitter, to eliminate the effect of reflections from the bombardment filament. The collector temperature is measured by a thermocouple placed in the copper housing, close to the collector base, and the cesium reservoir temperature is measured by a thermocouple attached to the reservoir.

This unit will operate and be tested in a bell-jar vacuum system, as originally planned.

4. Conclusions

The cesium thermionic work has been initiated and is progressing on schedule in spite of a late start. Because of the common design features of Test Units 1, 2 and 3 and because of other similar contract work at TEECo, the present scope of work can be expanded within the original estimated cost. Additional life test units originally planned for Fiscal Year 1962 can be fabricated this fiscal year.

B. SUBTASK 5.3--VACUUM DIODE GENERATOR

1. Objectives

The objectives of this subtask are to design, develop, fabricate and test an operational power system coupling a thermionic converter device to a radioisotope heat source. The thermionic development is being performed by Thermo Electron Engineering Corporation. The Martin Company carries out environmental testing, heat source development and other systems work.

2. Scope of Work

The following sub-subtasks were included in the work program for this quarter:

- (1) Evaluation of effects of fueling procedure on generator performance (TEECO) (5.3.2).

- (2) Dynamic analysis of Generator 2B (TEECo) (5.3.3).
- (3) Design and development of Generator 2B (TEECo) (5.3.4).
- (4) Fabrication and testing of Unit 2B (5.3.5).
- (5) Determination of the effect of patchy electrodes on generator performance (5.3.14).

3. Technical Discussion

a. Evaluation of fueling procedure (5.3.2)

The fueling procedure submitted by The Martin Company in Ref. 4 was evaluated by TEECo. Subsequent basic changes in the design of Generators 2B and 2C have created a need for a re-evaluation of anticipated fueling procedures. Therefore, this sub-subtask, though completed as originally planned, will still require the evaluation of new fueling procedures.

b. Dynamic analysis of Generator 2B (5.3.3)

A topical report of the dynamic analysis of Unit 2B has been completed. This report covers analytical results based on dynamic test requirements specified by the Jet Propulsion Laboratory, and submitted to TEECo by The Martin Company. Since the report was completed, there were some design changes in Unit 2B; specifically, the diode assembly was changed. A new dynamic analysis of the diode assembly will be prepared and a supplement to the topical report submitted in the next quarter.

c. Design of Generator 2B (5.3.4)

(1) Diode assembly

The previous quarterly report reported the design of Unit 2B as having sapphire rod spacers and spring retainment mechanisms to prevent separation of the emitter during dynamic testing. A dynamic analysis indicated that the sapphire rods would not withstand impact testing. This indication, along with previous expansion tests on sapphire rods, led to the conclusion that sapphire rods were not the ideal spacers for the next test unit.

A new spacing approach was undertaken during the latter part of this period. The new approach involves metallic spacers, as shown in Figs. 5 and 6. They consist of strips brazed to the collector and pinned on the side of the emitter; four strips are used for each diode. Proper spacing is obtained through differential expansion of the three diode

Fig. 5. Assembly of Unit 2B

Fig. 6. Diode Assembly

parts--emitter, collector and spacer. The diode exhibits better structural rigidity when assembled with these metallic strips than when assembled with sapphire rods. Tolerances are more easily attained, and assembly procedures are simpler. The parallelism of the emitter and collector surfaces can be reliably obtained and maintained with metallic strips. A diode using this type of spacer can be independently tested and then assembled in the generator casing. Therefore, time and materials can be saved in case of diode failure.

The metallic spacers can be designed and made so that thermal losses from the emitter are kept at the same level as, or lower than, when sapphire rods are used. When the dynamic requirements of the units are eased, the thermal losses can be reduced considerably. In the present design the estimated losses through the spacers are from 9 to 12 watts per diode.

(2) Emitter warpage

During the quarter of this report, an extensive test on emitter warpage as a function of collector contamination was completed. This test consisted of brazing a nonimpregnated emitter to a dense tungsten pellet with a molybdenum shim. This braze was made while the emitter and the tungsten pellet were under pressure so that the emitter would remain flat at high temperatures.

The brazed assembly was sent to Philips Metalonics for impregnation (type B impregnant). Philips reported that there was no obvious difficulty in impregnating the assembly. The impregnated emitter was inserted in a glass test tube and tested for emission. No defect was observable on the emitter; and, during life testing of this tube, no poisoning of the collector occurred, as did occur with other types of brazes. These results terminated the work on emitter warpage since the emitter was maintained flat within 5×10^{-6} inch.

(3) Collector work function

The design of Unit 2B was based on a work function of less than 2 volts. Such a work function, though easily attainable for brief periods, is exceedingly difficult to attain and maintain for operating periods of more than 100 hours. Therefore, this problem was studied further during the latter part of the quarter, and some extremely interesting results were obtained.

Previous tests indicated that, if free barium were oxidized on the collector surface, low work functions could be obtained, and if this oxidation could occur, during operation, when the barium is deposited on the collector, then a stable low work function could be expected.

This idea, though theoretically sound, is not easy to put into practice. The first approach in the laboratory was to provide an oxide film on the collector surface which could dissociate in the presence of barium. It was anticipated that the liberated oxygen would oxidize the barium and that this process could continue as long as there was oxide to be dissociated. In this first test extremely fine molybdenum oxide was used. Results of the work function test are shown in Fig. 7. Here again, although the work function was low--2 volts, complete stability was not achieved.

The next step with oxidized collectors was to try a thicker oxide film on the collector surface. The test results from this approach are shown in Fig. 8, in which it is obvious that the region of low work function has extended to longer periods than in the first test. This indicates that, with an appropriate amount of oxide, it may be possible to obtain a stable region of low collector work function. These results and the ease of attaining a low collector work function have prompted the use of this process in the fabrication of Unit 2B.

d. Fabrication of Unit 2B (5.3.5)

It was anticipated that Unit 2B would be fabricated and tested during this quarter. However, the unit was not tested due to difficulty in activating the emitters and collectors. The structural assembly of the various components of Unit 2B was considerably easier than that of any previous units fabricated.

Figures 9 through 17 show the subcomponents of Unit 2B. The unit was divided into six subcomponents: the heater, the heater section, top diode, bottom diode, top half-case and bottom half-case. Each of these could be processed and checked independently, permitting a quality control, and then assembled into a unit. During fabrication of the unit, the heater section was made first, along with the heater, and was tested for sufficient input heat as well as for sufficient thermal radiation shielding. The results of the tests were satisfactory; the heater and heater section then comprised an accepted part of the unit.

The second step was to fabricate the diodes. Each of these diodes, as shown in Figs. 9 and 10, consists of seven parts which, when assembled, produce a very compact and rigid diode. The diodes were assembled and then tested in an especially constructed test rig. Of four diodes only two produced any net power, and that was well below the anticipated output. The total output of these two diodes was 0.5 watt; individually, they produced 0.28 and 0.22 watt. The work functions of the collectors exceeded 2.15 volts.

Fig. 7. Collector Work Function Versus Time for Tube 221

Fig. 8. Collector Work Function Versus Time for Tube 231

Fig. 9. Exploded Diode

Fig. 10. Exploded Diode with Collector Assembled

Fig. 11. Heater

Fig. 12. Heater Section

224 030

Fig. 13. Heater Section with Heating Element

Fig. 14. Leadthroughs

Fig. 15. Top Diode with Casing During Activation

Fig. 16. Bottom Diode During Spacing Test

224 935

Section A-A

Fig. 17. Assembly of Heater

To explain these limited emissions without further testing is difficult. It is suspected that the oxidized collector permanently poisoned the emitter when the diode was brazed. This braze took place at 950° C, at which temperature the vapor pressure of molybdenum trioxide is high. This permanent poisoning might not have occurred had the emitters not been preprocessed. The emitters were boiled in distilled water for 10 minutes and then vacuum fired at 1125° C for 85 hours to reduce the high rate of barium evaporation so that any shorting in the diode due to barium deposits could be avoided. However, the firing introduced a porous layer in the emitter which, quite possibly, absorbed molybdenum trioxide vapor during the brazing of the diode assembly, causing contamination of the emitter.

An encouraging aspect of the fabrication of Unit 2B was the ease in achieving small interelectrode spacings at room temperature. These spacings were measured as 8 microns. The design of the spacers anticipated an additional 3 to 6 microns due to thermal expansion; however, this could not be checked since the emitter was limited and the I-V curves obtained during testing were inaccurate.

e. Investigation of the effect of patchy electrodes (5.3.14)

The Research Institute for Advanced Studies (RIAS), a division of The Martin Company, is conducting a study to determine the effect of nonhomogeneous surfaces on the emission of electrons. The following is a description of the work accomplished this quarter.

The main object of this work is to investigate the effect of nonhomogeneous surfaces on the emission of electrons. It is generally agreed that any surface, except perhaps a plane single crystal, has different work functions for different parts of its surface. These "patches" of different work functions are assumed, however, to generate some sort of average work function which is found experimentally from a Richardson plot. This report concerns some theoretical work on the emission of electrons from a patchy surface.

Instead of assuming that Richardson's equation

$$J = A T^2 \exp\left(-\frac{e\phi}{kT}\right) \quad (4)$$

holds for all surfaces, it has been assumed that it holds only for a portion of the surface having a single work function. The theoretical derivation makes the same assumption.

A real surface with N different patches may be considered as N ideal surfaces, each obeying Richardson's equation. Assigning a different work function to each of the N patches gives a set of N different work functions,

$\phi_1, \phi_2, \dots, \phi_N$. Also, the area of the patch with work function ϕ_1 is given the value a_1 ; the area of the patch with work function ϕ_2 , the value a_2 , and so on. If equilibrium has been attained, all surfaces would be expected to have approximately the same temperature. The current density from patch i would then be

$$J_i = \frac{I_i}{a_i} = AT^2 \exp \left(- \frac{e\phi_i}{kT} \right) \quad (5)$$

Here an A number of 120 is assigned to each surface since it is the theoretical value obtained for a surface with a single work function. The total current I leaving the surface is simply the sum of the currents from each area, shown in the following equations.

$$I = \sum_{i=1}^N I_i \quad (6)$$

$$I = \sum_{i=1}^N Aa_i T^2 \exp \left(- \frac{e\phi_i}{kT} \right) \quad (7)$$

$$\frac{I}{a_T} = AT^2 \sum_{i=1}^N \frac{a_i}{a_T} \exp \left(- \frac{e\phi_i}{kT} \right) \quad (8)$$

$$J = AT^2 \sum_{i=1}^N f_i \exp \left(- \frac{e\phi_i}{kT} \right) \quad (9)$$

where

a_T = total area of the emitting surface

$f_i = \frac{a_i}{a_T}$, the fractional area of the i th patch (9a)

$$J = \frac{I}{a_T}, \text{ the average current density of the emitting surface.} \quad (9b)$$

It is evident that

$$\sum_{i=1}^N f_i = 1 \quad (10)$$

The work function ϕ_i of the i th patch may be written as some average work function $\bar{\phi}$ plus an increment $\Delta\phi_i$ which represents the variation of ϕ_i from $\bar{\phi}$, or

$$\phi_i = \bar{\phi} + \Delta\phi_i \quad (11)$$

Inserting Eq (11) in Eq (9) gives

$$J = AT^2 \sum_{i=1}^N f_i \exp \left(\frac{e\bar{\phi}}{kT} - \frac{e\Delta\phi_i}{kT} \right) \quad (12)$$

$$J = AT^2 \exp \left(-\frac{e\bar{\phi}}{kT} \right) \sum_{i=1}^N f_i \exp \left(-\frac{e\Delta\phi_i}{kT} \right) \quad (13)$$

If the sum in Eq (13) is defined by the symbol F --

$$F = \sum_{i=1}^N f_i \exp -\frac{e\Delta\phi_i}{kT} \quad (13a)$$

then Eq (13) can be rewritten as follows:

$$J = FAT^2 \exp \left(-\frac{e\bar{\phi}}{kT} \right) \quad (14)$$

Equation (14) is the relation sought since it relates the A number and average work function $\bar{\phi}$ with a term F which is definitely a function of the patch structure. Certainly, if F is 1 (true when all $\Delta\phi_i = 0$), Richardson's equation holds for an A number of 120. But this is a perfect surface,

for if $\Delta\phi_i = 0$, then $\phi_i = \bar{\phi}$ for every patch. Equation (14) shows that the values given to $\bar{\phi}$ and to A are quite intimately connected. Richardson's equation shows that a large A number means high electron emission while a large work function means low electron emission. Equations (5) through (14) suggest that the values we choose for $\bar{\phi}$, $\Delta\phi_i$ and, therefore, F have quite a bit to do with the apparent A number. The question which is immediately raised is this:

Is a surface with a low A number and a low work function just another way of looking at a patchy surface with a moderately high A number and a moderately high work function?

Future work will seek the answer to this question.

The answer to the whole problem is in the determination of the function F . The first approach was to use a rather simple and much overworked two-patch model. This gave an expression which indicated the following: When the temperature of the surface was low, the low work function patches did most of the emitting; but as the temperature was raised, the higher work function patches emitted more and more until, in the limit as $T \rightarrow \infty$, the average work function was just halfway between the lower and upper work functions. In fact, the agreement with physical observation was correct in every limiting case, but the model gave no new information about the makeup of the surfaces.

The next approach to the problem of determining F was to assume N different work functions on the surface of the emitter. The value of each work function is exactly β volts higher than the next lower work function and β volts lower than the next higher work function; that is, every $\Delta\phi_i$ is exactly β volts. This is not really an assumption, but a way of numbering each patch, since the distribution of areas of each patch is the important factor. For example, if a patch having a work function of $\phi_{\text{minimum}} + 10\beta$ does not exist, its fractional area is given a value of 0. Now, only a value of f_i is needed in order to determine F .

As, perhaps, the simplest value of f_i for theoretical purposes, $f_i = \frac{1}{N}$ (for $i = 1, 2, \dots, N$) was chosen, with the assumption that all patches between ϕ_{minimum} and ϕ_{maximum} have equal area. This is a rather naive assumption, but it is an appropriate step in that it provides for the possibility of emission from all patches. More realistic distribution functions are also under study at this time. Even with the simple distribution used, however, some rather interesting and real effects have fallen

out of the equations. These include increased temperature dependence of J , an expression for the explicit dependence of A upon the patchy structure, and a relation between ϕ and apparent A number.

At this stage of the work, it is not certain what can be predicted from the theory. Certainly, however, if it can be verified from experiment that the surface is, as it appears, made up of many little surfaces, each obeying Richardson's equation, design methods for more efficient energy converters will soon be found. Also, it is now fairly certain that any method of determining work function in which an A of 120 is assumed to hold for all surfaces at all times will fail, as the following example will demonstrate.

A surface has a true work function of 1.8 volts at a temperature of 1200° K. With an A value of 120 amp/cm²-°K², the current density J is 2.94 amp/cm². If the current density were to decrease to 0.63 amp/cm², the A value remaining at 120, the work function ϕ would rise to 2.0 volts. The drop in current density, however, could possibly be explained by a decrease in A to 26 amp/cm²-°K² with the work function remaining constant at 1.8 volts. Probably, neither of these two views is exactly correct, and some compromise between them would be better. One thing is certain: If there is any rearrangement of patch structure, there will be a change in apparent A number. This study will be continued during the following quarter.

4. Conclusions

Low, reasonably stable collector work functions can be obtained if an oxidized collector is used. The use of oxidized collectors required investigation of a procedure for assembling the emitter and collector together without causing contamination of the emitter.

It is well known that oxygen will readily contaminate a barium-impregnated emitter; an oxidized collector presents a permanent oxygen source. Such contamination is suspected to have occurred in the diodes prepared for Unit 2B. Obviously, a proper procedure for assembling has not been prepared. This was not an oversight, but a matter of time, inasmuch as the oxidized collectors were tested only recently (early in March). It might have been well to make Unit 2B as planned during January when a clean collector was to have been used. The change in plans occurred because (as is substantiated by now) a clean collector could never have yielded a stable work function below about 1.95 volts and (since Unit 2B was expected to have a high performance) the encouraging results of the oxidized collector tests were compelling.

C. SUBTASK 5.4--CURIUM FUEL CAPSULE

Work on this subtask was initiated in January 1961. The Curium-242 fuel capsule is to be employed as the heat source for the vacuum diode generator being developed under Subtask 5.3. The work is to be closely integrated with Task 6, Fuel Technology Program.

1. Objectives

The objective of this subtask is to design, develop, fabricate and test an encapsulated Curium-242 heat source suitable for fueling the vacuum diode generator presently being developed under Subtask 5.3.

The heat source will:

- (1) Produce approximately 120 thermal watts at an effective power density of 30 watts/cc.
- (2) Be capable of operation at 1650° K with a vapor pressure less than 10^{-7} mm Hg.
- (3) Be compatible with the vacuum diode generator and fueling procedure.
- (4) Be safe in operation.

2. Scope of Work

Subtask 5.4 consists of six sub-subtasks, which are:

- (1) Fuel capsule design and analysis (5.4.1).
- (2) Fuel form development (5.4.2).
- (3) Testing of capsule design (5.4.3).
- (4) Preparation of curium fuel pellets (5.4.4).
- (5) Encapsulation and testing of final fuel capsule (5.4.5).
- (6) Decontamination and waste disposal (5.4.6).

During the quarter, work was performed only on Sub-subtask 5.4.1, which consisted of effort in the following areas:

- (1) Survey of suitable container materials for the curium fuel capsule.
- (2) Temperature-time and pressure-time analyses of the fuel capsule under the following conditions:
 - (a) In a normal air environment.
 - (b) In a vacuum environment.
 - (c) In a water environment.
 - (d) Contained within the vacuum diode generator under normal operating conditions.
- (3) Calculation of the temperature distribution throughout the fuel capsule as a function of the capsule surface temperature. This would make the results of Item 2 applicable to this analysis.
- (4) Determination of the initial surface temperature of the capsule, given that the surface is at 1650° K after three and six months of operation.
- (5) Radiation levels as a function of distance from an unshielded capsule.
- (6) Evaluation of the most feasible container concepts generated during Fiscal Year 1961 from the standpoint of safety.
- (7) Generation of several fuel container designs which will be compatible with available materials, present design of the vacuum diode generator and the fueling procedure of the generator.
- (8) Stress analysis of the more promising container designs which are generated.
- (9) Establishment of experimental procedure for testing the container under simulated operational conditions.

The scope of effort this quarter consisted of defining the problem involved in the design of the fuel pellet container and obtaining several design concepts.

3. Technical Discussion

a. Materials survey

The survey of fuel container materials has been completed. The materials were evaluated on the bases of vapor pressure, strength fabricability and oxidation resistance. The containment of gas pressure was considered of paramount importance. The complete materials survey can be found in Appendix A of this report.

The materials selected to be evaluated are:

- (1) Molybdenum with 0.5% titanium (Mo-0.5 Ti).
- (2) Molybdenum with 0.5% titanium and 0.07% zirconium (Mo-0.5 Ti-0.07 Zr).
- (3) Tungsten with 2% thorium dioxide (W-2 ThO₂).
- (4) Tantalum with 10% tungsten (Ta-10 W).
- (5) Tantalum with 7.5% tungsten (Ta-7.5 W).

Procurement of these materials has been initiated.

Experiments to determine the effects of gas pressure and temperature on Mo-0.5 Ti fuel containers have begun. Figure 18 shows the configuration tested. Although this configuration is not to be used in the final design, the experiments will give an indication of the amount of pressure the materials can withstand and will aid in development of a test.

The pressure at the test temperature representing a particular isotope life was obtained by pressurizing at room temperature with the amount of gas which would give the desired pressure at elevated temperature, as determined by the gas law. The first test was designed to give 23 atmospheres (the pressure expected after 21 days) at 1650° K. Capsules as shown in Fig. 18 were dimensionally checked before testing and after 1, 3 and 64 hours at temperature (1650° K). The capsules were tested in a hydrogen-atmosphere furnace. No dimensional changes were found. The seal plugs in these capsules were then drilled open under water to make sure the gas was still contained. They were then pressurized to obtain 57 atmospheres at 1640° K and were dimensionally checked as above. No changes were evident. No further testing has been conducted because of the pressure limitation of the existing welding equipment. An autoclave will be adapted to weld the capsules at higher pressure.

Fig. 18. Capsule Configurations

b. Analytical studies

The results of the analytical work have, for the most part, been recorded graphically. These graphs are included in this report with a short explanation of how and why they were generated. The result of this mode of presentation will be a permanent record of the effort to date which will prove useful in the continuing effort next quarter.

(1) Figure 19. Pressure per Gram of Cm-242 as a Function of Time

This figure represents the initial effort in defining the extent of the pressure buildup problem which would be encountered in the design of the fuel container. Because of the well insulated structure of the vacuum diode generator, it was surmised that the temperature decay of the fuel capsule surface with time would be very gradual. To give a general idea of the pressure problem, Fig. 19 ignores the temperature decay with time. The graph was plotted through the use of the following relations.

$$p = \frac{n(t) RT}{v} \quad (15)$$

where

p = pressure

R = universal gas constant

T = absolute temperature

v = void volume

$n(t)$ = moles of helium present at any time t .

It can easily be shown that $n(t)$ is given by the expression

$$n(t) = \frac{M_0 [1 - \exp(-\lambda t)]}{242} \quad (16)$$

where

M_0 = grams of Curium-242 present at time zero

λ = isotope decay constant

t = time.

Fig. 19. Pressure per Gram of Cm-242 Versus Time

Initial estimates based on the current design of the vacuum diode were that the fuel container would be loaded with one gram of Curium-242, have approximately one cubic centimeter of void volume and operate with an initial surface temperature of 1650° K. Figure 19 shows that, under these conditions, the pressure in the fuel container is approximately 100 to 450 atmospheres. This indicates that a serious problem exists in the design of the fuel container.

(2) Figure 20. Capsule Surface Temperature as a Function of Normalized Power

This figure is an interim plot required in obtaining the temperature-time relation for the capsule in an air and in a vacuum environment. In this study it is considered that the capsule is a cylinder 1.0 inch in diameter and 19/32 inch in length and is sitting upright on an insulated base. The relation shown in Fig. 20 is

$$P_{\text{rad}} = \sigma \epsilon A (T^4 - \theta^4) \quad (17)$$

where

P_{rad} = radiated energy

σ = Stefan-Boltzmann constant

ϵ = capsule surface emissivity

A = lateral area plus top area of the capsule

T = capsule surface temperature

θ = infinite sink temperature.

It is realized that the final design shape of the capsule will not be cylindrical, but, at this point, the final concept is not known. The nature of the thermal analysis (dominance of the T^4 term) tends to nullify, to a large extent, changes in the fuel container geometry. Large changes in the capsule area cause only small changes in the surface temperature with all other parameters equal.

(3) Figure 21. Normalized Power as a Function of Time

This figure is another interim plot which is convenient for almost any of the studies on the fuel container. It is simply the decay curve of the Curium-242 isotope. The relation is

Fig. 20. Capsule Surface Temperature Versus Normalized Power

Fig. 21. Normalized Power Versus Time

$$P_0/P = \exp(\lambda t) \quad (18)$$

where

P_0 = initial isotope power

P = isotope power at any time t

λ = Curium-242 decay constant

t = time.

(4) Figure 22. Capsule Surface Temperature as a Function of Time

This figure is the final result of the temperature-time relation of the fuel capsule in vacuum. It is assumed that the capsule is sitting on an insulating base and radiating to an infinite sink at 25° C. The graph is parametric in initial isotope power level divided by capsule emissivity. The initial fuel loading P_0 should be approximately 120 watts, and the capsule emissivity will probably be in the neighborhood of 0.6. Therefore, the resulting value of the ratio P_0/ϵ is 200 watts, and Curve 2 will give the desired result.

(5) Figure 23. Capsule Surface Temperature Versus Convected Heat

Figure 23 is an interim plot required in obtaining the temperature-time relation for the fuel capsule while in a normal air ambient. It gives the amount of heat convected from the capsule surface as a function of the capsule surface temperature. Again it is assumed that the capsule is sitting on an insulated base and rejecting heat to air at 25° C. The relation for this graph is given by the expression

$$P_{\text{conv}} = hA (T - \theta) \quad (19)$$

where

P_{conv} = convected heat

h = heat transfer coefficient

A = lateral and top surface area of the capsule

T = capsule surface temperature

θ = ambient air temperature.

Radiation to an infinite sink at 25°C while on an insulated base

Vacuum environment

Fig. 22. Capsule Surface Temperature Versus Time

Fig. 23. Capsule Surface Temperature Versus Convected Heat

The heat transfer coefficient h is given by

$$h = 0.35 (T - \theta)^{0.25} \quad (\text{top}) \quad (20)$$

$$h = 0.25 (T - \theta)^{0.25} \quad (\text{side}). \quad (21)$$

Equations (20) and (21) are empirical relations applicable to a turbulent situation. In these equations $(T - \theta)$ must be expressed in $^{\circ}\text{R}$ and h will be in $\text{Btu/ft}^2\text{-hr-}^{\circ}\text{F}$. This is important for the sake of consistency.

(6) Figure 24. Capsule Surface Temperature Versus Total Heat Dumped

This is again an interim plot which is obtained simply by adding the abscissas of Figs. 20 and 23. It gives the total heat (radiated and convected) which may be dumped to an infinite sink at 25°C while the capsule is sitting on an insulated base.

(7) Figure 25. Capsule Surface Temperature Versus Time

This figure represents the final result for the temperature-time determination of the capsule surface temperature while on an insulating base in air environment at 25°C . The graph is parametric in emissivity. This figure is a result of combining Figs. 21 and 24 for an initial fuel loading of 120 watts.

(8) Figure 26. Pressure per Gram of Cm-242 per Cubic Centimeter of Void Volume Versus Time

This figure represents the pressure-time relation for the capsule under the conditions given by Fig. 25. If the initial fuel loading is 120 watts (1 gram of Cm-242) and the void volume is 1 cc, the pressure in the capsule will reach a maximum of 170 atmospheres after 350 days.

This high pressure emphasizes the problem imposed in designing this capsule. A worse situation will be incurred for the capsule contained within the vacuum diode generator.

(9) Figure 27. Heat Dumped Versus Generator Temperature

This figure is an interim graph of the radiant heat dump capabilities of the vacuum diode generator as a function of the generator surface temperature. The surface emissivity of the generator is included as a parameter. It is assumed that the generator is radiating from all surfaces to an infinite sink at 25°C .

The capsule is on an insulated base and dumping heat into an infinite sink at 25°C.

Fig. 24. Capsule Surface Temperature Versus Total Heat Dumped

Fig. 25. Capsule Surface Temperature Versus Time

Fig. 26. Pressure per Gram of Cm-242 per Cubic Centimeter of Void Volume Versus Time

Fig. 27. Heat Dumped Versus Generator Temperature

(10) Figure 28. Generator Surface Temperature Versus Time

This is a plot of the vacuum diode generator surface temperature versus time considering that the generator is in a vacuum environment consisting of an infinite sink of 25° C. The initial loading of the generator is taken to be 120 watts.

(11) Figure 29. Capsule Surface Temperature Versus Time

This figure is the temperature-time relation for the fuel capsule contained within the vacuum diode generator, assuming a temperature of 1650° K at the end of three and six months. An initial isotope loading of 120 watts is assumed.

It is assumed that the isotope fuel container temperature is independent of the temperature of the surroundings. This will be a fair approximation of the actual case for the capsule at high temperature. The approximation will become worse for decreasing surface temperature. Therefore, there is absolutely no justification for extrapolating the given relation below 1650° K. The means of generating these curves is given by the expression

$$q = CT^4 = q_0 \exp(-\lambda t) \quad (22)$$

where

q = radiated heat

q_0 = initial isotope loading

T = capsule surface temperature

λ = Curium-242 decay constant

t = time.

The constant C in Eq 22 is solved for times of three months and six months with the surface temperature at 1650° K. The time is then varied back to the zero point and the resulting surface temperatures calculated.

(12) Figure 30. Heat Loss Through Radiation Shields Versus Generator Surface Temperature

This figure gives the heat loss through the radiation shields around the fuel capsule as a function of the generator surface temperature. It is estimated from Fig. 28 that the maximum temperature the generator

Fig. 28. Generator Surface Temperature Versus Time

Fig. 29. Capsule Surface Temperature Versus Time

Fig. 30. Heat Loss Through Radiation Shields Versus Generator Surface Temperature

surface can attain is 480° C. Therefore, the heat loss through the shields is relatively invariant with respect to the generator surface temperature.

The equation which describes this system is*

$$Q'_{\text{net}} = \frac{\sigma}{n-1} \langle A\mathfrak{F} \rangle (T_1^4 - T_n^4) \quad (23)$$

where

Q'_{net} = heat flowing through the shields

σ = Stefan-Boltzmann constant

n = number of radiation shields (includes supports, capsule surface and generator surface)

$\langle A\mathfrak{F} \rangle$ = average area of shield times view factor for concentric cylinders

T_1 = temperature of capsule surface

T_n = temperature of generator surface.

(13) Figure 31. Heat Loss Through Radiation Shields Versus Fuel Capsule Surface Temperature

This figure is simply a cross plot of Fig. 30 and again stresses the invariance of the heat loss through the radiation shields with respect to the generator surface temperature.

The foregoing provides a description of the analytical effort expended in the third quarter of Fiscal Year 1961 toward providing the operational temperature and pressure data for the curium fuel capsule under various conditions. This effort is not complete at this time as the temperature-time and pressure-time relations for the fuel capsule contained within the generator have not been obtained. Figures 27, 28, 29 and 30 represent the preliminary steps in obtaining this information.

In order to provide an analytical basis for initiating the design of the fuel capsule, temperature-time and pressure-time relations for an operational fuel capsule were estimated. These relations were obtained by utilizing the equation

*This derivation can be found in Appendix B.

Fig. 31. Heat Loss Through Radiation Shields Versus Fuel Capsule Surface Temperature

$$q_0 \exp(-\lambda t) = CT^4 \quad (24)$$

where

q_0 = initial loading of the isotope

λ = decay constant of Curium-242

t = time

C = constant

T = temperature of fuel capsule.

It was assumed that the surface temperature of the capsule would be 1650° K after 22 days (when the isotope power has decayed from 120 to 110 watts) and the constant C was calculated for this point. The temperature-time relation was then easily generated. To compensate for the roughness of the approximation of the temperature-time determination, a safety or uncertainty factor has been utilized. This factor is given by Fig. 32. The uncertainty factor has been tapered to be higher at lower fuel capsule temperatures for reasons explained previously.

The temperature-time relation is given by Fig. 33, and the pressure-time by Fig. 34. The time scale has been carried out to only 90 days, the length of time required for the initial design effort on the fuel capsule. Figures 33 and 34 represent only a rough estimate of the temperature and pressure which will be incurred by the fuel capsule while it is contained within the vacuum diode generator. The continuing analysis will attempt to improve upon these estimates.

c. Preliminary design of the fuel capsule

The primary objective in the mechanical design of the fuel container is the minimization of stress in the container wall. For any wall thickness of the fuel container, there will be a resulting stress from the helium pressure buildup. Naturally, a thick-walled container would be stronger and able to contain higher pressures than a thin-walled container. However, the space available for the capsule within the vacuum diode generator is fixed, and increasing the container wall thickness decreases the void volume available. This means that even though a thick-walled container would be stronger, it would also be subjected to higher pressures. There is only one wall thickness that will be subjected to minimum stress.

Fig. 32. Safety Factor Versus Time

Fig. 33. Capsule Surface Temperature Versus Time

Fig. 34. Pressure Per Cubic Centimeter of Void Volume Versus Time

The exact final design configuration of the fuel capsule is as yet unknown. However, initial investigations indicate that the container will probably consist of a cylindrical lateral surface with spherical heads. Therefore, the stress minimization analysis was worked out for a cylinder and a sphere. The final result will probably consist of a superposition of the two analyses.

(1) Cylindrical capsule analysis

The stress in a cylindrical vessel containing a pressure is

$$S = \frac{PR_i}{t} \quad (25)$$

where

S = stress

P = internal pressure

R_i = inner radius of cylinder

t = wall thickness.

The void volume of the container is given by

$$v = \left[\pi (R_i - t)^2 L \right] - C \quad (26)$$

where

v = void volume

L = internal length of cylinder

C = volume of fuel.

Substituting the perfect gas law into Eq (25) and utilizing Eq (26) results in

$$S = \frac{(R_i - t) nRT}{t \left\{ \left[\pi (R - t)^2 L \right] - C \right\}} \quad (27)$$

where

R = universal gas constant

T = capsule temperature

n = moles of helium present.

Setting $dS/dt = 0$ and solving for C/L in terms of t , with $R_i = 0.527$ inch, give an expression for the wall thickness which will result in minimum stress:

$$\frac{C}{L} = -11.905t^3 + 15.690t^2 - 6.07t + 0.869 \quad (28)$$

(2) Spherical capsule analysis

The stress in a spherical vessel containing a pressure is

$$S = \frac{PR_i}{2t} \quad (29)$$

The steps of the cylinder analysis are repeated, and the final result is

$$C = 47.687t^4 - 83.739t^3 + 53.028t^2 - 13.972t + 1.228 \quad (30)$$

where the void volume is given by

$$v = \frac{4}{3} \pi R_i^3 - C. \quad (31)$$

(3) Interpretation of results

The results of these two analyses, Eqs (28) and (30), are presented graphically in Figs. 35 and 36. In applying the results of the spherical analysis to an ellipsoidal container, C must be adjusted to provide an equivalent void volume. In other words, the $(4/3 \pi R_i^3)$ term in Eq (31) will not be correct for an ellipsoidal container.

Fig. 35. Ratio of Fuel Capsule Volume (C) to Cylinder Length (L) Versus Wall Thickness

Fig. 36. Volume of Fuel Capsule Versus Wall Thickness

Two conceptual design sketches (Figs. 37 and 38) have been generated according to the foregoing analyses. Based on a fuel pellet volume of 0.146 in.³ (power density, 50 watts/gram of Cm_2O_3), the resulting pressures are extremely high. Tables on Figs. 37 and 38 show the decrease in pressure that is obtained by increasing the allotted envelope size within the generator. It may be seen that increasing the envelope size by 3/16 inch in the axial dimension decreases the resulting pressure buildup by a factor varying between 2 and 3. These pressure determinations are based on Fig. 34 at the 90-day time.

Several assembly concepts have been generated and are illustrated in Figs. 39a through 39e. Since the structural strength of a seal weld is questionable, it is hoped that some type of mechanical fastening can be employed. This will mean that the seal weld will only have to provide a gastight closure.

4. Conclusions

The conclusions concerning the materials survey are:

- (1) Based on the established criteria for the Subtask 5.3 vacuum diode generator, the materials which may be considered for the fuel container are the refractory metals tungsten, molybdenum and tantalum and their alloys. Specifically, tungsten, the titanium- and zirconium-containing molybdenum alloys, and the tantalum alloy containing 10% tungsten appear to be the more promising in satisfying the prominent strength requirement.
- (2) The lack of sufficient data on elevated-temperature properties prevents establishing design allowables for the refractory metals and alloys.
- (3) Based on a capsule design with a weld closure and the attendant metallurgical changes, data on powder metallurgy products, which exhibit higher strength values, are not applicable. If a design which does not depend on a weld closure is used, the powder products may be considered.
- (4) Tungsten is the only material which satisfies the requirement of oxidation resistance at 1310° F; however, the normal variability in oxidation performance of different melts of material makes tungsten a potential borderline case. The acceptability of tungsten is predicated on:
 - (a) Immediate drop in temperature to 1310° F.

A dimension	Void volume (cm ³)	Pressure, 90 days (psi)
0.552 (as shown)	1.065	2277
0.615 (+ $\frac{1}{16}$)	1.703	1426
0.677 (+ $\frac{1}{8}$)	2.344	1035
0.740 (+ $\frac{3}{16}$)	3.016	803

Note:

1. All dimensions in inches.
2. Operating temperature = 2500°F

Fig. 37. Curium-242 Capsule with Concave Heads

A dimension	Void volume (cm ³)	Pressure, 90 days (psi)
0.552	1.655	1466
0.615 (+ $\frac{1}{16}$)	2.229	1088
0.677 (+ $\frac{1}{8}$)	2.835	856
0.740 (as shown) (+ $\frac{3}{16}$)	3.426	708

Note: 1. All dimensions in inches
2. Operating temperature = 2500°F

Fig. 38. Curium-242 Capsule with Inverted Heads

Fig. 39a. Capsule Assembly Concept

Fig. 39b. Capsule Assembly Concept

Fig. 39c. Capsule Assembly Concept

Fig. 39d. Capsule Assembly Concept

Fig. 39e. Capsule Assembly Concept

- (b) No loss in strength due to a thinning of the wall while oxidation occurs.
- (c) Uniform attack--no preferential or intergranular attack.
- (5) The application of coatings would probably involve a prohibitively long investigation, even if a satisfactory coating could be found.
- (6) Insufficient experiments have been conducted on the Mo-0.5 Ti capsules to draw definite conclusions. Further testing will be performed.
- (7) From the initial analytical and design effort, it has been found that the pressure buildup within the fuel container will pose a formidable problem. Means are available for minimizing this effect: cutting away the cathode backup plate or increasing the axial dimension of the generator to provide more room within the generator for the fuel capsule. Both of these methods will be investigated, with the emphasis being placed on the first method.

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III. REFERENCES

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APPENDIX A

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MATERIAL SURVEY FOR THE PURPOSE
OF SELECTING CONTAINER MATERIALS FOR
SUBTASK 5.4,
CURIUM FUEL CAPSULE FOR
VACUUM DIODE GENERATOR*

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*Survey by John Kane

I. INTRODUCTION

The following criteria were assumed in the survey of materials for the purpose of selecting a container for the Subtask 5.4 vacuum diode:

- (1) Operating temperature--2507° F (1640° K).
- (2) Vapor pressure--less than 10^{-7} mm Hg at temperature.
- (3) Containment of helium pressure buildup.
- (4) Oxidation resistance--satisfactory to resist oxidation for 240 hours, maintaining integrity.

The materials considered most suitable, based on the above criteria, were determined by a process of elimination. Initially, only those materials with vapor pressures lower than 10^{-7} mm Hg were considered. These metals were screened to eliminate all that did not have melting points greater than 3400° F, 900° F above the operating temperature of the container, since they would not have sufficient strength to contain the anticipated helium gas pressure. Next, the fabricability of the remaining materials was considered, and those metals which have never been fabricated successfully, or for which fabrication procedures are extremely limited, were rejected. The remaining metals and their alloys were then evaluated for strength and oxidation resistance.

The materials to be used for the container of the curium fuel capsule are first restricted to those which have vapor pressures less than 10^{-7} mm Hg at the operating temperature. Since the constituents of an alloy exhibit their characteristic vapor pressures, the alloys to be surveyed must not contain large amounts of elements which have vapor pressures above 10^{-7} mm Hg. If minor additions of these elements are made, tests will be required before acceptance.

The only metals which have vapor pressures lower than 10^{-7} mm Hg are listed in Table A-1 with their respective vapor pressures at 2507° F (1640° K).

TABLE A-1
Metals Considered for Vacuum Diode Container

<u>Metal</u>	<u>Vapor Pressure (mm Hg)</u>	<u>Metal</u>	<u>Vapor Pressure (mm Hg)</u>
Tungsten	1×10^{-15}	Ruthenium	$1 \times 10^{-10*}$
Tantalum	4×10^{-15}	Zirconium	1×10^{-9}
Rhenium	8×10^{-15}	Hafnium	2×10^{-9}
Columbium	$1 \times 10^{-12*}$	Iridium (1)	4×10^{-9}
Osmium	$2 \times 10^{-11*}$	Thorium	4×10^{-8}
Iridium (2)	$2 \times 10^{-11*}$	Rhodium	4×10^{-8}
Molybdenum	3×10^{-10}	Platinum	1×10^{-7}

* Extrapolated from higher temperature.

The philosophy of thermionic fuel form containment requires that decay product gases be completely contained in the fuel capsule. The necessity for containment of gas pressure buildup automatically precludes the use of any materials which have melting points near (within 900° F) the operating temperature of the generator. The materials in this category are zirconium, hafnium, thorium, rhodium and platinum; these were, therefore, not considered further.

The materials remaining for consideration were the refractory metals and the platinum group metals osmium, ruthenium and iridium. Since osmium and ruthenium had never been successfully worked, they were excluded. Iridium, very difficult to work, was excluded because of its poor oxidation resistance, complete lack of mechanical property data and the requirement of high reliability.

The two strength properties which are of interest in evaluating materials for generator use are creep and creep rupture. There are available only a limited amount of data concerning creep at the operating temperature of the generator. Some data at lower temperatures, although not applicable to design, will be included to give an idea of the rapidly diminishing strengths at these elevated temperatures. Conventional

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tensile strength data, which are often included in such material surveys, are not considered in this report as they are not applicable in the design of a capsule. However, a curve giving the relative strengths of the alloys of the various refractory metals is included to indicate the relative usefulness of these materials.

Refractory metal alloys may be classified into two types with respect to their mechanical properties:

- (1) High strength alloys.
- (2) Oxidation-resistant alloys.

The high strength alloys are those of molybdenum and tungsten which contain strong carbide formers, and those of tantalum and columbium which contain solid solution strengtheners.

The oxidation-resistant alloys are generally those of columbium or tantalum with additions of up to 10% titanium. The oxidation-resistant alloys, however, have significantly lower strength.

It should be noted that the majority of the oxidation-resistant refractory alloys contain metal additions which have vapor pressures greater than 10^{-7} mm Hg; therefore, these alloys cannot be used in a vacuum thermionic diode.

II. STRENGTH

Since containment of gas pressure without deformation or rupture is the most important criterion to be met, in the proof-of-principle unit, strength was given preference over resistance to oxidation. This approach was based on a knowledge of the generally poor strength of available materials at 2507° F (1640° K), the design temperature. Table A-2 gives the data available for molybdenum and its alloys, the materials for which the most data were available. The majority of data are for temperatures below the operating temperature, while one reference gives data for the operating temperature. These particular data are for powder-product molybdenum, which is not applicable for a design based on weld closure; the weld would be cast metal and would exhibit entirely different, and lower, strength values.

The data given for molybdenum alloys in the recrystallized condition at 2400° F (that material condition for which the data would be applicable) indicate that the Mo-0.46 Ti-0.08 Zr-0.08 C alloy would be the strongest molybdenum alloy.

TABLE A-2

Available Stress Rupture and Creep Data of Molybdenum Alloys

Alloy	Temperature (° F)	Stress x 10 ⁻³ (psi x 10 ³)	Time to Rupture (hr)	Creep Rate (%/hr)
Unalloyed Mo (r)	2000	17.5	0.15	--
Unalloyed Mo (r)		12.0	5.3	3.0
Mo-0.5 Ti (r)	2000	25.0	4.4	6.0
Mo-0.5 Ti (r)		20.0	221.1	0.11
Mo-0.5 Ti (r)		18.0	280.0 (d)	0.023
Mo-0.5 Ti (r)	2400	5.0	258.9	0.013
Mo-0.5 Ti (r)		3.0	834.4 (d)	0.0023
Mo-0.5 Ti (r)		2.0	361.0 (d)	0.0003
Mo-0.5 Ti (r)		1.0	700.3 (d)	0.00011
Mo-0.5 Ti 0.2C (r)		9.9	100	--
Mo-0.46 Ti-0.08 Zr- 0.08 C (r)	2400	20.0	15.0	0.90
		16.0	32.9	0.46
		14.4	100	--
Mo-1.27 Ti-0.29 Zr- 0.30 C (r)	2400	11.8	100	--
Mo-25 W-0.1 Zr-0.05 C (r)		5.5	100	--
Mo-(pm) (r)	2507	8.0	5	--
Mo-(pm) (r)		5.0	80	--
Mo-(pm) (r)		3.5	2500	--
Mo-(pm) (r)		2.5	40,000	--

All material arc cast except where noted

(r)--Recrystallized

(d)--Test discontinued

(pm)--Powder metallurgy.

The data on the other refractory metals are more limited than for molybdenum. All data on tungsten (as shown in Table A-3) are for the powder product and, again, are not applicable to a design based on weld closure. There are no creep rupture data available on arc-melted tungsten; however, short-time tensile data show it to be significantly stronger than other alloys considered.

Tantalum is weaker than the molybdenum alloys; however, data on Ta-10 W indicate that it is considerably stronger than tantalum at 3000° F. Based on the higher strength at this temperature, it can be assumed that the alloy may be significantly stronger at the operating temperature of the generator. Tantalum-10 W or any other commercially available tantalum alloy should be considered for evaluation as a container material.

The strength values for the two columbium alloys in Table A-2 indicate a prohibitive creep rate at 2500° F. No other data are available for temperatures at or near the operating temperature.

All of the pertinent data given in Tables A-2 and A-3 for the various metals and alloys give only time to rupture. It should be emphasized at this point that rupture data should not be interpreted as meaning that gases will be contained until rupture, but rather that leakage will probably occur long before rupture. Also, a considerable amount of deformation (e.g., approximately 20 to 30% for the molybdenum alloy) occurs before the indicated rupture time is reached, and, therefore, legitimate creep data are required. Designs should be predicated on a known long-term rate of growth. The appropriate creep data are not available in the literature.

Figure A-1 is included in this report to give an indication of the relative short-time strengths of the refractory metal alloys being considered.

III. OXIDATION

Although the refractory metals are noted for strength at elevated temperatures, they possess extremely poor oxidation resistance and there is a considerable volume change when oxides of refractory metals are formed. Therefore, these oxides will not adhere to the surface of the metal and protect the metal. Also, the oxides of the refractory metals have very low melting points and are vaporized at relatively low temperatures. Attempts have been made to increase the oxidation resistance of these materials by alloying them with metals that possess a higher oxidation resistance. None of these attempts has proved successful, and, as mentioned before, many of the best metallic additions

TABLE A-3
Available Stress Rupture and Creep Data of Tungsten,
Tantalum and Columbium Alloys

<u>Alloy</u>	<u>Temperature (° F)</u>	<u>Stress x 10³ (psi x 10³)</u>	<u>Time to Rupture (hr)</u>	<u>Creep Rate (%/hr)</u>	
W (pm) (r)		4.5	100	--	--
W-2 ThO ₂ (pm) (w)	2500	9.1	100	--	
W-2 ThO ₂ (pm) (w)		20	100 another source	--	--
W (pm) (r)		15.0	9	--	
W (pm) (r)		12.0	140	--	--
W (pm) (r)	2507	10.0	1000	--	
W (pm) (r)		8.0	6400	--	
W (pm) (r)		5.0	112,000	--	
Ta (ac) (w)		2.2	8	--	
Ta (ac) (w)	2552	1.0	100	--	
Ta (ac) (w)		4.0	0.05	--	
Ta (ac) (w)	3000	2.5	0.31	--	
Ta-10 W (ac) (w)		15.0	0.076	--	
Ta-10 W (ac) (w)	3000	12.5	0.44	--	
Ta-10 W (ac) (w)		10.0	0.60	--	
Ta-130 ppm N ₂ -5.6 ppm O ₂		1.5	100	--	--
Cb-10 Mo-10 Ti (D-31)	2500	2.5	--	2.0	--
Cb-20 W-10 Ti-6 Mo (D-41) (heat treated)		9.0	--	2.0	--

(pm)--powder metallurgy
(r)--recrystallized
(ac)--arc cast
(w)--worked.

Fig. A-1. Estimated Tensile Strengths Attainable with the Best Tungsten, Molybdenum, Tantalum and Columbium Base Alloys

TABLE A-4
Comparison of Selected Columbium-Base Alloys

<u>Alloy</u>	<u>Composition</u>	<u>Investigator</u>	<u>Tensile Strength at 2200° F (psi x 10³)</u>	<u>10-Hour Rupture Stress at 2200° F (1000 psi)</u>	<u>Strength: Density Ratio</u>		<u>Fabricability Rating</u>	<u>Depth of Oxidation in 10 Hours at 2200° F, Scale + Contamination (mils)</u>
Cb-74	Cb-10 W-5 Zr	Union Carbide	45	19	--	136	11	>74, <145
F-48	Cb-15 W-5 Mo-1 Zr-0.05 C	General Electric	42	24	180	130	7	75
D-41	Undisclosed	duPont	42	12	178	128	9	12
Cb-7	Cb-28 W-7 Ti	Union Carbide	41	19	147	120	10	17
F-50	Cb-15 W-5 Mo-5 Ti-1 Zr-0.05 C	General Electric	35	15	152	106	6	32
Cb-16	Cb-20 W-10 Ti-3V	Union Carbide	26	11	174	104	8	7
D-31	Cb-10 Mo-10 Ti	duPont	25	9	121	86	5	28
80	Cb-0.75 Zr	Fansteel	--	--	--	--	3	> 145
82	Cb-33 Ta-0.75 Zr	Fansteel	21	--	--	--	4	145
Cb-65	Cb-7 Ti-0.8 Zr	Union Carbide	15	--	55	--	2	56
Cb		--	9	--	--	--	1	45

for improving oxidation resistance are not usable in vacuum diode applications because of their high vapor pressures. Figure A-2 gives oxidation data for a number of unalloyed refractory metals and other metals at various temperatures. The refractory metals show considerable oxidation attack. There are very few data available on alloying of refractory metals for oxidation resistance in any material except columbium. Tables A-4 and A-5 give oxidation rates for a number of columbium alloys. It can be seen that these alloys, although the best developed, still leave much to be desired.

TABLE A-5
Mils Lost Per Side at 2000° F

	<u>8 hr</u>	<u>16 hr</u>	<u>24 hr</u>	<u>32 hr</u>	<u>Composition</u>
Mo	200	400	650	1000	--
Cb-1 Zr	22	41	60	--	0.75-1.0 Zr, commercial
F-48	6.1	9	13	16	15 W-13 Mo-1 Zr, pilot
Cb-16	2.3	4.1	6.0	6.5	20 W-10 Ti-3 V, lab
F-50	2.3	3	3.5	3.9	15 W-5 Mo-1, Zr, standard pilot
10 Ti-10 Mo (D-41)	3.5	5	7.5	9.3	

Table A-6 shows the effect of the most promising additions of metals to tantalum to improve oxidation resistance.

Fig. A-2. Oxidation of the Refractory Metals

Table A-6 shows the effect of the most promising additions of metals to tantalum to improve oxidation resistance.

TABLE A-6
Air Oxidation Rates of Tantalum Alloys at 1400° C

Nominal Composition (wt %)	Weight Gain After Indicated Time (mg/cm ²)			
	1 hr	2 hr	8 hr	4 hr
100 Ta*	--	--	--	--
Ta-2 Ti	63.5	96.4	111.6	--
Ta-40 Ti	47.0	78.0	100.0	--
Ta-10 Hf	116.9	--	--	--
Ta-20 Hf	70.1	94.5	106.1	--
Ta-40 Zr	57.4	68.3	73.6	77.7
Ta-10 V-20 Ti	67.4	99.0	--	--

* Oxidized too fast to record.

These data give an example of the types of additives needed to improve the oxidation resistance of tantalum. Unfortunately, the effects of these additives on the strength of tantalum are not known; therefore, their use would be questionable.

Table A-7 gives the results of work done at The Martin Company on oxidation of refractory metals at 1310° F.

TABLE A-7
Results of Oxidation of Refractory Metals at 1310° F

<u>Material</u>	<u>Weight Loss (mg/cm²-hr)</u>	<u>Mils Lost/Side/Hour</u>
Molybdenum	38.21	1.5
Tantalum	38.74	0.9
Tungsten	3.83	0.1
Columbium	17.94	0.8

Based on the criterion of acceptance (240 hr at 1310° F), tungsten appears to be the only satisfactory unalloyed material.

Only a cursory survey has been made of coatings for the capsules; most of the coating work, however, has been for protection at 2192° F and below, and the effectiveness of these coatings at 2792° F is very questionable. It should be noted that these coatings are generally useful for only a short time at operating temperature. The requirement of operation at 2507° F in a vacuum of 10^{-7} mm Hg also restricts the use of many of the more promising coatings as many of the coating components have high vapor pressures. The elevated temperature of the curium capsule would pose a problem in coating, and new methods of application would probably have to be devised if a suitable coating existed.

IV. CONCLUSIONS AND RECOMMENDATIONS

The conclusions given below are based on satisfying a requirement for a reliable proof-of-principle generator.

- (1) Based on the established criteria for the Subtask 5.3 vacuum diode generator, the materials which may be considered for the fuel container are the refractory metals tungsten, molybdenum and tantalum and their alloys. Specifically, tungsten, the titanium- and zirconium-containing molybdenum alloys, and the tantalum-10% tungsten alloy appear to be the more promising in satisfying the strength requirement.
- (2) There are insufficient elevated-temperature property data to allow establishing design allowables for the refractory metals and alloys.
- (3) Based on a capsule design with a weld closure and the attendant metallurgical changes, data on powder metallurgy products, which exhibit higher strength values, are not applicable. If a design which does not depend on a weld closure is used, the powder products might be considered.
- (4) Tungsten is the only material which satisfies the requirement of oxidation resistance at 1310° F; however, the normal variability in oxidation performance of different melts of material makes tungsten a potential borderline case. The acceptability of tungsten is predicated on:

- (a) Immediate drop in temperature to 1310° F.
 - (b) No loss in strength due to a thinning of the wall while oxidation occurs.
 - (c) Uniform attack--no preferential or intergranular attack.
- (5) The application of coatings would probably involve a prohibitively long investigation, even if a satisfactory coating could be found.

The material studies have led to the following recommendations:

- (1) Pressure containment tests of capsules made from the recommended container materials should be performed to determine their suitability and relative performance in long-term service.
- (2) The cermet should be made from an oxidation-resistant metal which would contain a major portion of the isotope in a solid shape, even if the outer container is oxidized away. The isotope compound to be contained by the oxidation-resistant metal matrix should also be oxidation-resistant to prevent rupture of the cermet.

V. REFERENCES

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APPENDIX B

DERIVATION OF THE HEAT TRANSFER EQUATION FOR PURE
RADIATION SHIELDING OF CYLINDRICAL GEOMETRY. *

*Derivation by Thomas S. Bustard.

The following is the derivation of Eq (23), given in Chapter II, Section C of this report.

Fig. B-1. Generator in Cross Section

The surfaces shown in Fig. B-1 are the thermal shields. This derivation makes the following assumptions:

- (1) There is no temperature drop in the radial thickness of a shield.
- (2) The shield is at uniform temperature.

(3) There is no axial heat loss from the shields.

Then

$$\begin{aligned}
 Q_{\text{net}} &= A_1 \widetilde{f}_{12} \sigma T_1^4 - A_2 \widetilde{f}_{21} \sigma T_2^4 \\
 &= \dots = A_{n-1} \widetilde{f}_{n-1,n} \sigma T_{n-1}^4 - A_n \widetilde{f}_{n,n-1} \sigma T_n^4
 \end{aligned} \tag{B-1}$$

where

Q_{net} = heat flow through shields

σ = Stefan-Boltzmann constant

T = shield temperature

A = shield area

$\widetilde{f}$ = Hottel reflection-view factor.

$$\begin{aligned}
 Q_{\text{net}} &= \frac{\sigma}{n-1} \left[A_1 \widetilde{f}_{12} T_1^4 - A_2 \widetilde{f}_{21} T_2^4 + A_2 \widetilde{f}_{23} T_2^4 \right. \\
 &\quad - A_3 \widetilde{f}_{32} T_3^4 \dots \dots \dots + A_{n-1} \widetilde{f}_{n-1,n} T_{n-1}^4 \\
 &\quad \left. - A_n \widetilde{f}_{n,n-1} T_n^4 \right]
 \end{aligned} \tag{B-2}$$

Assume

$$A_i \widetilde{f}_{i, i+1} = A_{i+1} \widetilde{f}_{i+1, i} \tag{B-3}$$

which is a close approximation for concentric cylinders.

Then

$$Q_{\text{net}} = \frac{\sigma}{n-1} < A \widetilde{f} > (T_1^4 - T_n^4) \tag{B-4}$$

where

$< A \widetilde{f} >$ = an average area times view factor to dampen out Assumption (B-3).