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SPACE ISOTOPE POWER

A COMPENDIUM IN SUPPORT OF THE RATIONALE FOR SELECTING SiGe AS THE THERMOELECTRIC TECHNOLOGY FOR A LONG-LIFE, MULTIHUNDRED-WATT RADIOISOTOPE THERMOELECTRIC GENERATOR (U)

Edited by: R. D. Kinney, 9522

Contributors:

J. R. Holland, 5321
W. A. Stark, 5321
D. Spatz, 9521
H. J. Gerwin, 9522
D. R. Johnson, 9522
D. J. Mottern, 9522
P. D. O'Brien, 9522
I. G. Waddoups, 9522
W. F. Windle, 9522

SPACE ISOTOPE POWER DEPARTMENT 9520

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Sandia Laboratories, Albuquerque

October 1969

ABSTRACT (U)

This report presents the rationale for choosing SiGe thermoelectric technology for a long-life, multihundred-watt radioisotope thermoelectric generator. Typical mission requirements, generator performance goals, and safety criteria are discussed. Heat source technology and current thermoelectric and converter technology relative to a long-life, multihundred-watt RTG are described. The reasons for selecting SiGe technology are summarized.

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The cutoff date for information in this report is September 1969.

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SUMMARY

Section 1 of this report contains a discussion of typical mission requirements, generator performance goals, and safety criteria which represent the current AEC interpretation of future needs for space isotopic power in the multihundred-watt range.

Section 2 presents conclusions regarding the selection of SiGe as the thermoelectric technology for a converter meeting the performance goals outlined in Section 1.

Section 3 presents a brief discussion of current thermelectric and converter technology that has applicability to a long-life, multihundred-watt generator. Life test data are presented as the primary basis for assessing the capability of each technology relative to the requirements stated in Section 1.

Section 4 contains a discussion of heat source technology. The present status of capsule development, fuel/material compatibility, and helium generation are discussed in an effort to reach some conclusions about the present and future capability of heat sources for high-temperature, long-life operation.

FOREWORD

In anticipation of the electrical power needs of projected National Aeronautics and Space Administration (NASA) and Department of Defense (DOD) satellites and planetary probes, the United States Atomic Energy Commission (USAEC) proposes to develop a long-life, multihundred-watt, radioisotope thermoelectric generator (RTG) having sufficient versatility to satisfy most of these needs. This report documents the rationale and presents a summary of supporting information for selecting SiGe as the thermoelectric material for this generator. The selection of SiGe technology represents the concensus of Sandia Laboratories, the USAEC, and all but one of the contractors who submitted proposals for the design and development of the above multihundred-watt generator.

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A COMPENDIUM IN SUPPORT OF THE RATIONALE FOR SELECTING SiGe AS THE THERMOELECTRIC TECHNOLOGY FOR A LONG-LIFE, MULTIHUNDRED-WATT RADIOISOTOPE THERMOELECTRIC GENERATOR

1.0 Basic Requirements for the Multihundred-Watt Power System

The objective of the multihundred-watt power system development program is to develop a modular, long-life, multihundred-watt, plutonium-fueled radioisotope thermoelectric power system which will be available for a number of potential space missions. Emphasis will be placed on a modular design in order to provide flexibility in using the power system for the various missions which may occur in the 1973 to 1978 time frame.

Typical missions in this time frame are listed below.

a. Earth orbital spacecraft using an RTG power system of approximately 200 to 300 watts electrical and having at least a 5-year life, with an initial launch sometime during 1973-1974.

b. Lunar surface roving vehicle using an RTG power system of approximately 200 to 500 watts electrical having at least a 1-year life, to be launched during 1973-1974.

c. Deep space probe mission requiring approximately 550 watts electrical for at least 3 to 5 years, with an initial launch sometime during 1973-1974; and a later space probe mission requiring approximately 500 watts electrical for at least 12 years, to be launched during 1976-1978.

In order to develop a power system which can be adapted to the different missions listed above, the following generator characteristics were established as design guidelines.

a. Power output (end of generator design life): 100 to 200 watts.

b. Minimum generator output voltage: 6 volts (matched load).

c. Design life: 3 to 12 years.

d. Weight: minimum.

e. Power/weight ratio: 2 watts/lb minimum for 5 years.

f. Envelope: minimum consistent with other mission requirements.

g. Fuel: $^{238}PuO_2$ (solid solution cermet).

The safety philosophy will be to maximize the probability of plutonium containment under all normal and accident environments by virture of redundancies of both containment and plutonium immobilization mechanisms inherent in the fuel form. Containment and/or plutonium immobilization is to be assured at impact.

2.0 Conclusions

The decision to use SiGe technology for the multihundred-watt, long-life generator is based on evaluation of the current and projected status of thermoelectric materials, converters, and heat sources, as discussed in Sections 3.0 and 4.0, and on the following conclusions.

a. The technology for SiGe material production, couple fabrication, and reproducibility has been convincingly established.

b. Available life test data on thermoelectric generators, modules, and couples show that SiGe devices have the least performance degradation per unit time of operation.

c. Life test data for SiGe modules are for substantially longer time periods than are available for other thermoelectric materials. Hence, this technology gives the highest confidence in extrapolating performance.

d. SiGe material is capable of operation in either vacuum or air without protection.

e. The present status of fuel capsule development indicates the establishment of high-temperature fuel capsule technology within the time frame proposed for generator development. This would ensure realization of the full potential of an SiGe converter.

f. SiGe converters are competitive at lower temperatures of operation. This provides flexibility for lowering heat source temperature if mission length dictates a less severe thermal environment for the heat source.

g. Additional study in the time available before the selection of the basic design approach would not produce new information.

3.0 Converter Technology

3.1 Sealed Converter Technology

The technology for the SNAP-19 and SNAP-27 converters is well established, and designs using this technology are potential candidates for the multihundred-watt RTG described in Section 1.0. However, design life for both converters is considerably less than the 3 to 12 years desired for the multihundred-watt generator, and long-life capability must be demonstrated before one can gain confidence in these approaches.

Life testing is an important source of information for evaluating converter capability, and converters of the SNAP-19 and SNAP-27 designs have been placed on long-term vacuum tests by different organizations. The results of several tests are summarized in Figures 1 through 5 where data from three SNAP-19 and three SNAP-27 units are presented. These tests are not exhaustive, but they represent a fair cross section of SNAP-19 and SNAP-27 generators and include long term data which are of primary importance for a multihundred-watt generator.

Three SNAP-27 generators, MOD 5, MOD 8, and a prime flight qualification generator, MOD 10, were life tested in the environmental test chambers at GE-MSD. The tests were terminated as planned on July 10, 11, and 12, 1968, for MOD 8, MOD 5, and MOD 10, respectively. Thermal sink conditions for all generators were cycled to simulate maximum temperatures for lunar day and lunar night operation. Since test conditions were cycled between a lunar day and night environment, power output was normalized to beginning of life (BOL) power for Figures 1, 2, and 3. Absolute power output is given in Table I.

Data for the SNAP-19 units are presented as time history of total converter power output.

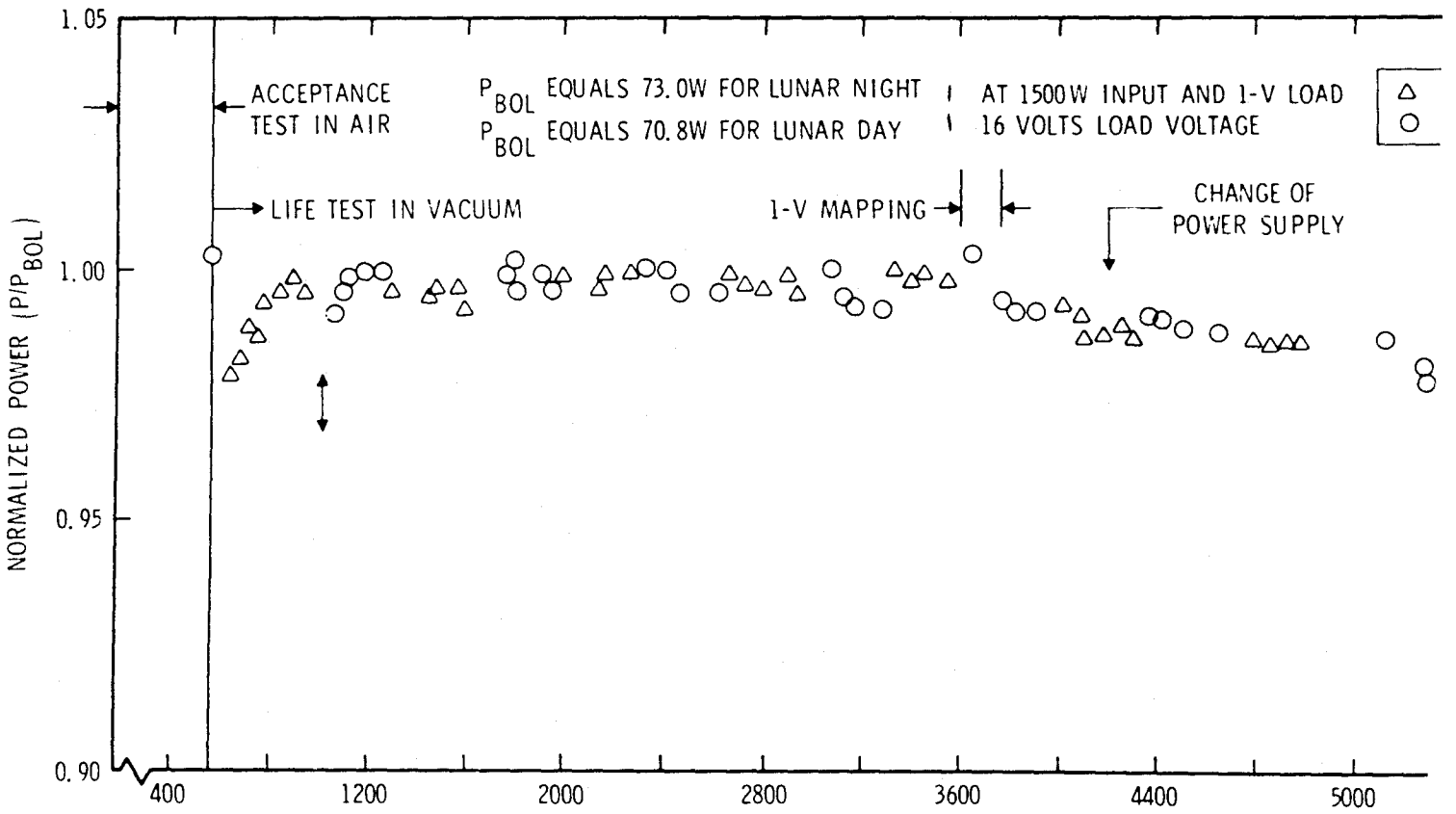
TABLE I

SNAP-27 Generator Life Test Summary

Generator	Total Accumulated Test Time (hr)	Power Output (BOL) (W)		Heat Input (W)	Power Output (EOL) (W)	
		Lunar Day	Lunar Night		Lunar Day	Lunar Night
MOD 5	12,165	70.8	73.0	1500; after 10,000 hours, reduced to 1485	66.6	68.7
MOD 8	8,341	65.0	66.0	1415	62.0	62.9
MOD 10	7,697	71.2	73.3	1505	71.5	73.1

The long-life capability of the SNAP-19 and SNAP-27 is dependent, in part, on suppressing the sublimation of the PbTe thermoelectric material. Pressurization with inert gases is the primary means for suppressing sublimation in the SNAP-19 and SNAP-27 generators. However, other factors can also cause suppression, and, therefore, the consequences of losing gas pressure in a sealed generator have not been fully established.

Both power output and internal pressure data are available as a function of time for several SNAP-19 units (Figures 4 and 5), but no immediate conclusions about generator performance and gas pressure can be drawn from a comparison of these data. The internal pressure was not monitored on the SNAP-27 units shown in Figures 1, 2, and 3.


1.05
1.00
0.95
0.90
NORMALIZED POWER (P/P_{BOL})
400
1200
2000
2800
3600
4400
5000
ACCEPTANCE TEST IN AIR
LIFE TEST IN VACUUM
P_{BOL} EQUALS 73.0W FOR LUNAR NIGHT
P_{BOL} EQUALS 70.8W FOR LUNAR DAY
AT 1500W INPUT AND 1-V LOAD
16 VOLTS LOAD VOLTAGE
1-V MAPPING
CHANGE OF POWER SUPPLY

Figu

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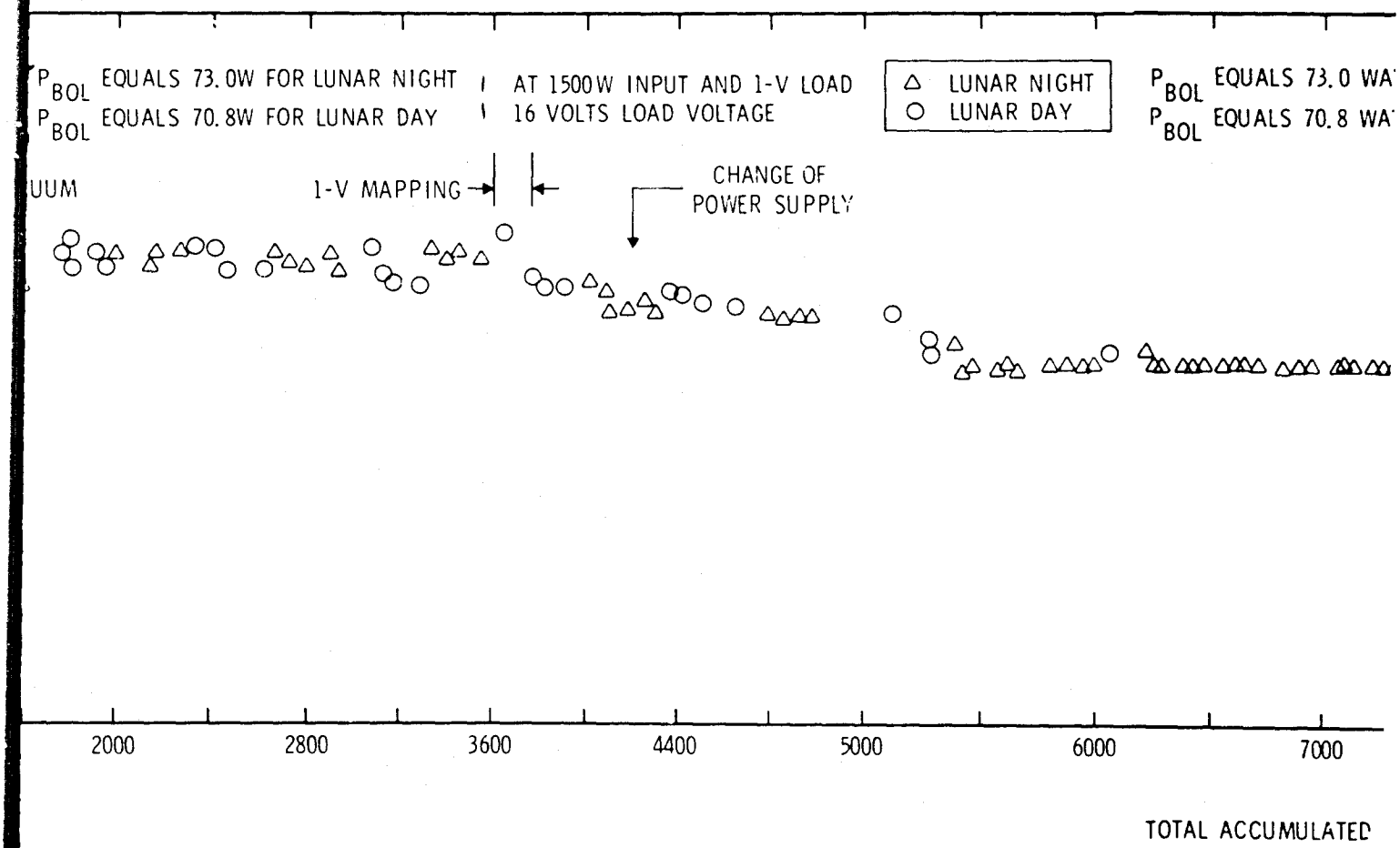

Figure 1. Life test data for

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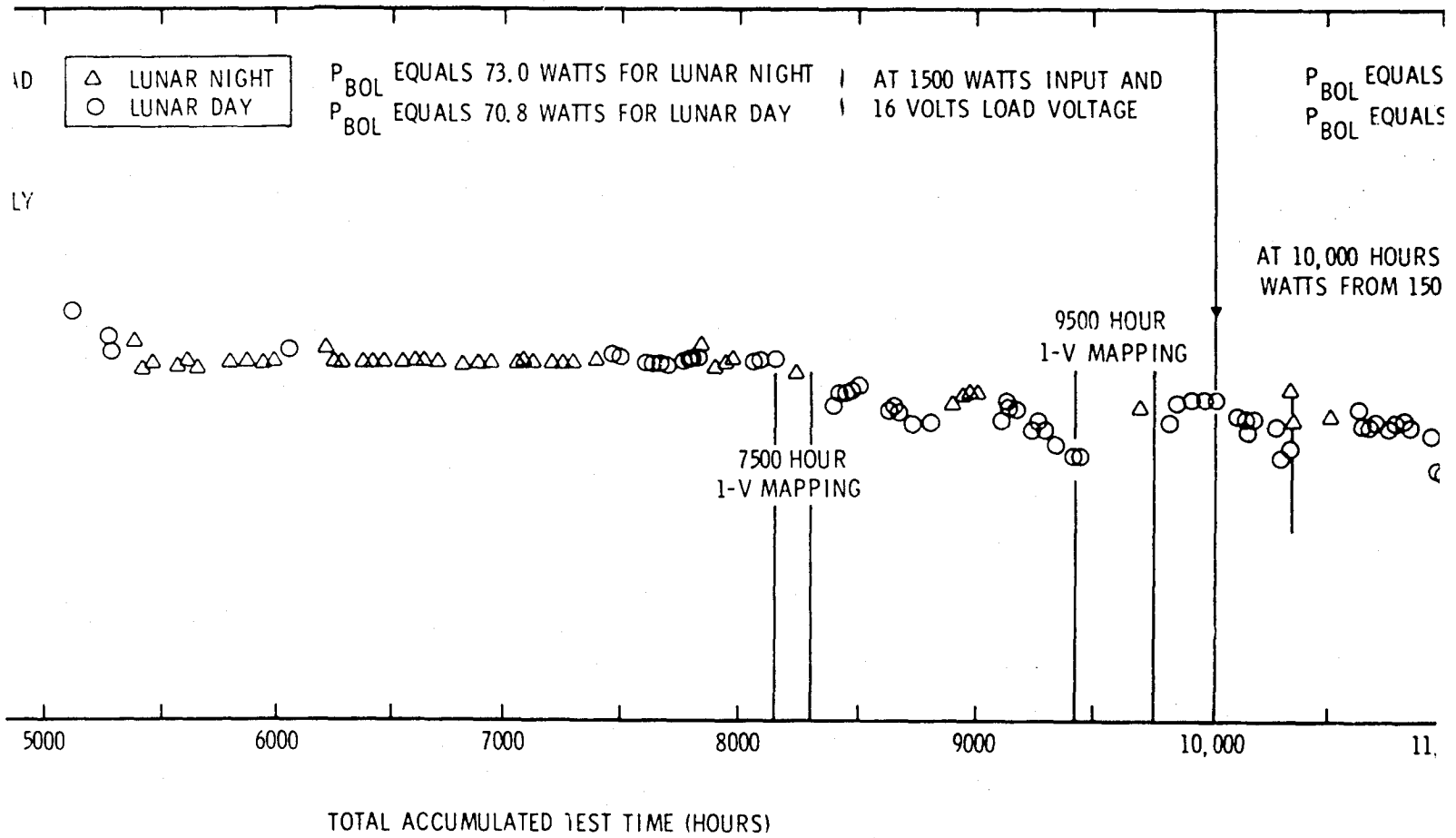

Figure 1. Life test data for SNAP-27, MOD 5 converter

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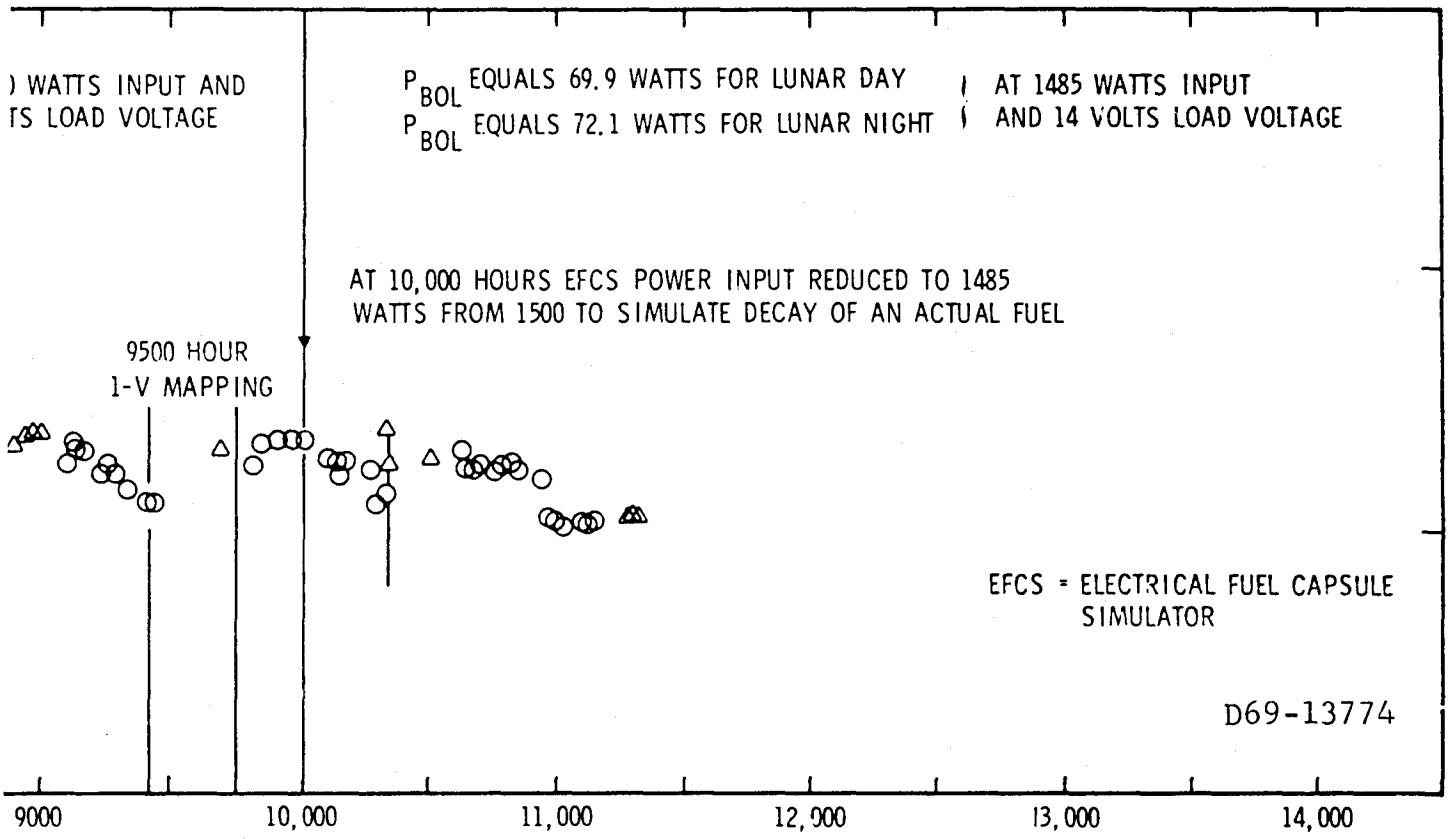
) WATTS INPUT AND
TS LOAD VOLTAGE
P_{BOL} EQUALS 69.9 WATTS FOR LUNAR DAY
P_{BOL} EQUALS 72.1 WATTS FOR LUNAR NIGHT
AT 1485 WATTS INPUT
AND 14 VOLTS LOAD VOLTAGE
AT 10,000 HOURS EFCS POWER INPUT REDUCED TO 1485
WATTS FROM 1500 TO SIMULATE DECAY OF AN ACTUAL FUEL
9500 HOUR
1-V MAPPING
EFCS = ELECTRICAL FUEL CAPSULE
SIMULATOR
D69-13774
9000
10,000
11,000
12,000
13,000
14,000

ter

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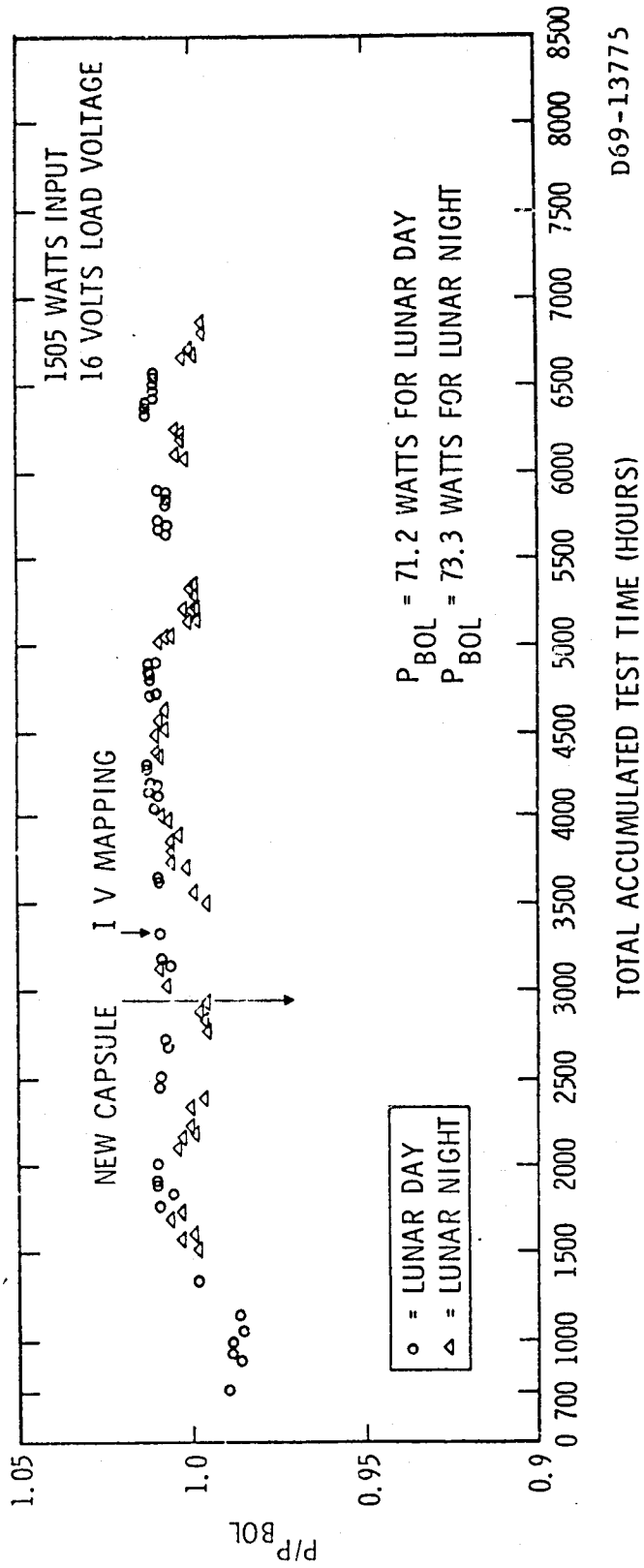

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Figure 2. Life test data for SNAP-27, MOD 10 converter

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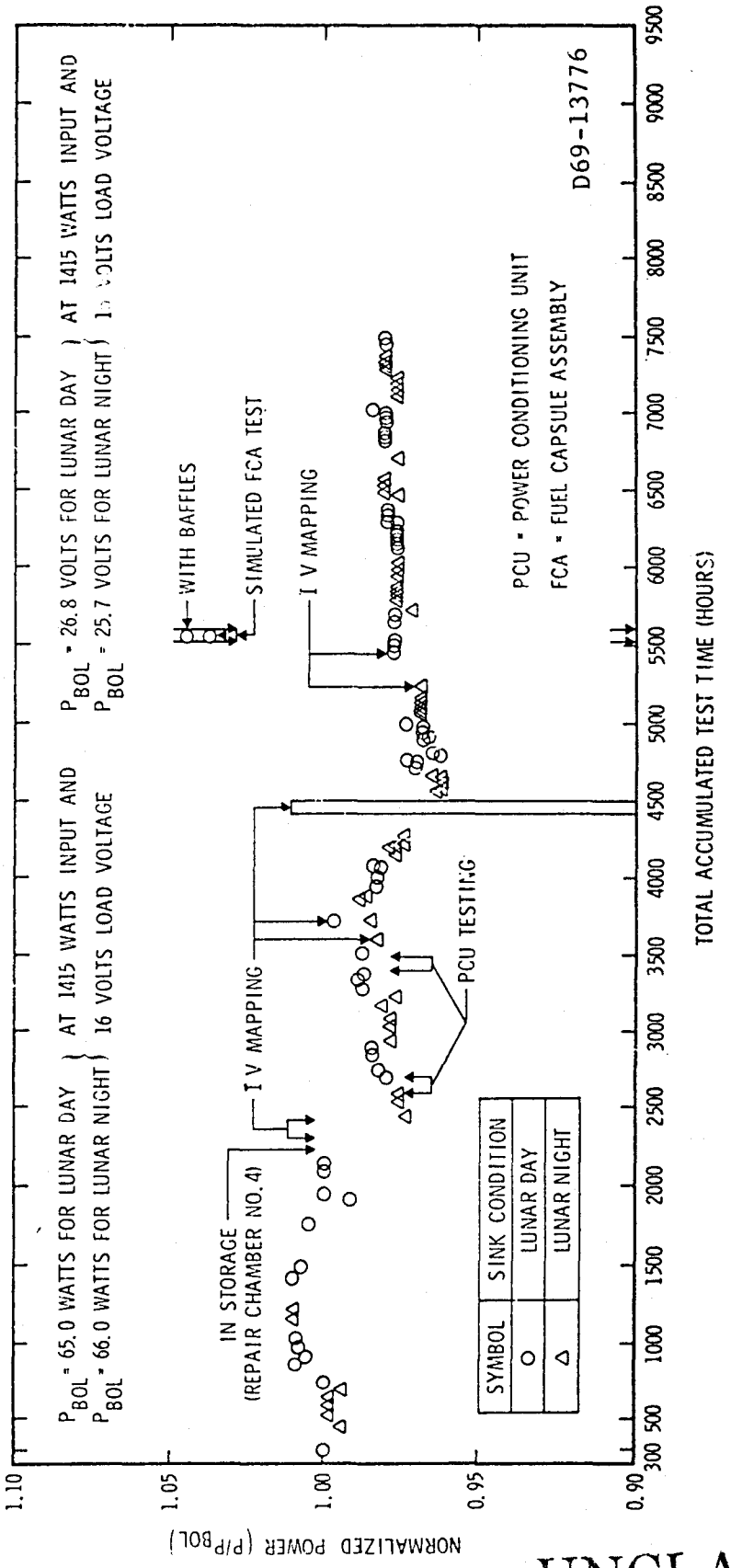

Figure 3. Life test data for SNAP-27, MOD 8 converter

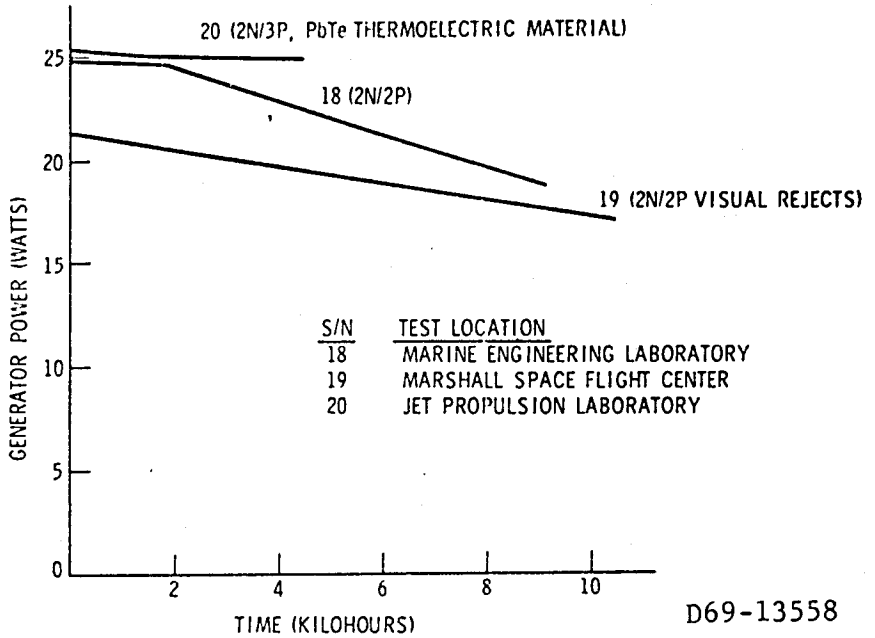

Figure 4. Power output history for three SNAP-19 generators in vacuum

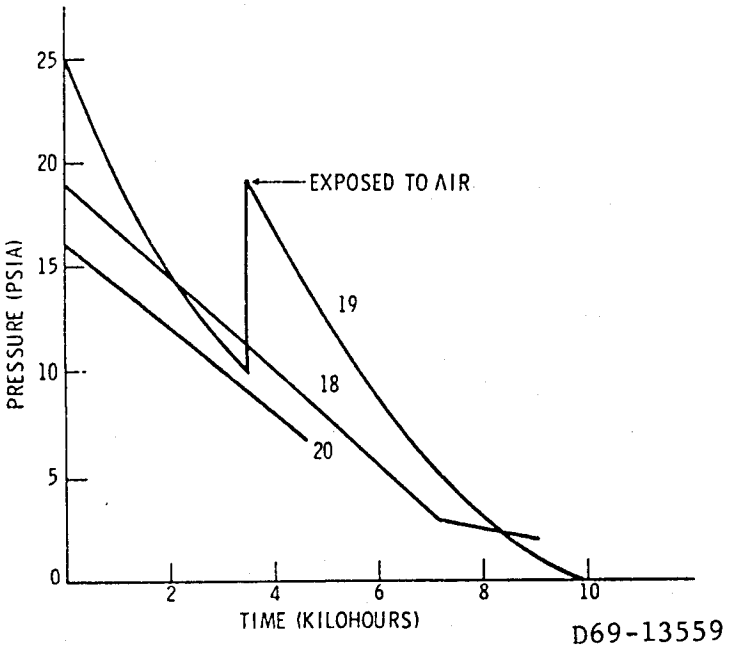

Figure 5. History of internal pressure for three SNAP-19 generators

3.2 SNAP-27 Couple Tests

General Electric and the 3M Company in conjunction with the SNAP-27 program have placed modules of the SNAP-27 type on life test. The results of these tests provide additional data for determining long-term behavior.

Eight 10-couple and six 104-couple modules with 3N/3P thermoelements have had preliminary analyses and were compared for power degradation as a function of hot junction temperatures. The results are listed in Table II. Test times for the 10-couple modules range from 22,000 to 25,000 hours. Data for the 104-couple modules extend from 15,000 to 19,000 hours. The data accumulated since the cutoff time for these analyses show no obvious change in degradation rates. A definite correlation exists between degradation rate and hot junction temperature; however, the spread in the data is appreciable.

TABLE II

10/104-Couple Module Summary

Number of Couples	Serial Number	Test Duration (kilohours)	T_h (°F)	Degradation (% per kilohour)
10	C7	22	1050	0.10
10	C8	22	1050	0.08
104	B13	18	1050	0.33
104	B14	18	1050	0.39
10	B2	25	1100	0.45
10	C4	24	1100	0.48
104	B2	15	1070	0.62
104	B10	19	1100	1.48
104	B12	17	1100	0.82
10	C5	23	1130	2.05
10	C6	23	1130	0.88
104	B11	19	1130	1.88
10	C9	22	1200	2.09
10	C10	21	1200	3.11

Several more modules are on test under a variety of designs and test conditions to investigate the effects of cover gas pressure, spring pressure, and temperature cycling, but only preliminary analyses have been made of these results.

3.3 Isotec Converter System

The Gulf General Atomic Isotec thermoelectric panel demonstrated the feasibility of operating unsealed PbTe elements at low temperature (400°C) for long periods without severe loss of material by sublimation. This approach removes the need for a gas environment around the thermoelectric elements.

P-elements of the thermocouples have demonstrated integrity up to 24,000 hours of operation at normal operating temperature. On the other hand, first designs for the N-element showed bond separation relatively early (~10,000 hours) in the life tests. Figure 6 shows test data for some early modules. The time to failure for different modules is plotted against hot junction temperature.

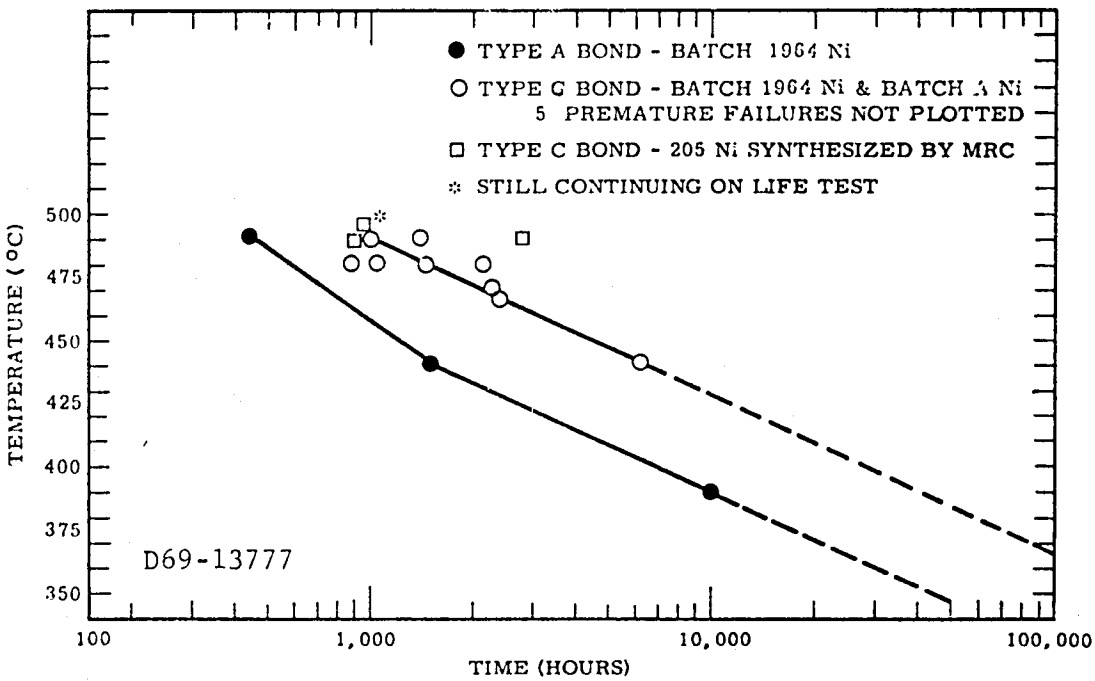

Figure 6. Lifetimes of Isotec N-element modules using type-A and type-C bonds

Gulf General Atomic now uses a mechanical and metallurgical (M&M) bonding technique which has lessened the concern of bond failure of the N-element. The N-element and P-element are schematically described in Figure 7. Life test data for N-element modules using the M&M bond show no module failures by open circuits after 10,800 hours at 500°C hot junction temperature, whereas earlier N-elements failed in less than 2000 hours at this temperature. Table III gives a summary of the latest testing for modules using the M&M bond technology.

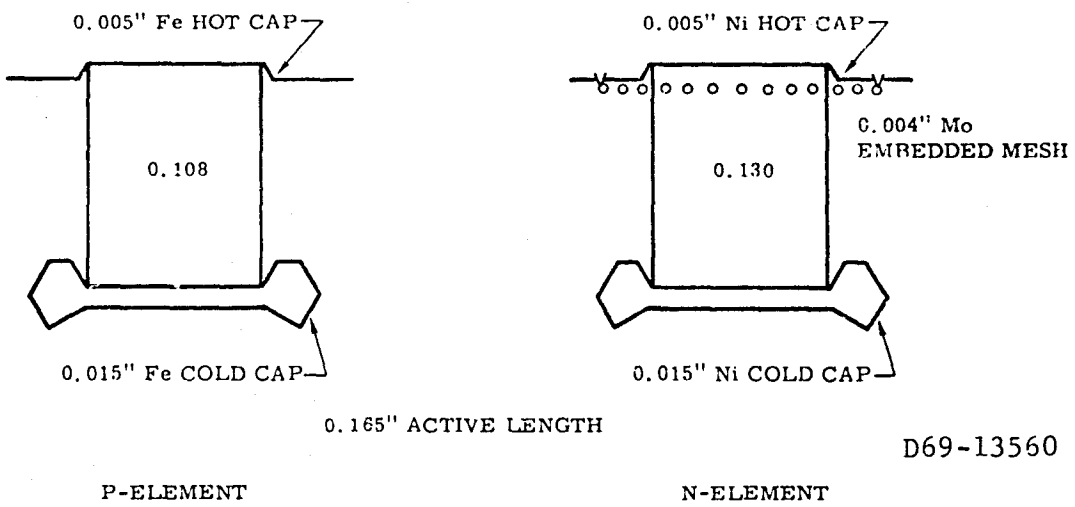

Figure 7. Schematic of Isotec converter thermoelectric elements

TABLE III

Testing Summary for Modules and Converters with M&M Bond (Data as of June 1969)

N-element modules with M&M bonds	
X-113	10,800 hours at 500°C (W & SS mesh material) (No open circuit failures)
4N-117	8,900 hours at 500°C (Mo mesh material) (No open circuit failures)
Converter using M&M bond	
GC04	30.6 watts at 4.82% efficiency (BOL) (Placed on life test in June 1969)

Table IV shows life test data for Isotec generator GC02. This generator uses Isotec panels with the type-C bond on the N-element. Figure 8 is a plot of the output power of the generator normalized to $\Delta T = 244°C$.

TABLE IV

Testing Summary for Isotec Generator GC02

Test Time (hr)	T_h (°C)	T_c (°C)	ΔT (°C)	P_o [1] (W)	P_i [2] (W)	P_n [3] (ΔT = 244)	V [4] (V)
1	392	169	223	23.58	607	28.2	3.917
1027	389	170	219	22.66	612	28.1	3.840
2031	388	169	221	22.61	601	27.5	3.835
2981	396	187	209	20.65	579	28.1	3.672
4042	398	187	211	20.41	580	27.3	3.644
4635*	401	186	215	20.07	580	25.8	3.608

*Test suspended.

[1] P_o = generator power output.

[2] P_i = source power input.

[3] P_n = generator power output normalized to ΔT = 244°C with constant hot junction.

[4] V = generator terminal voltage.

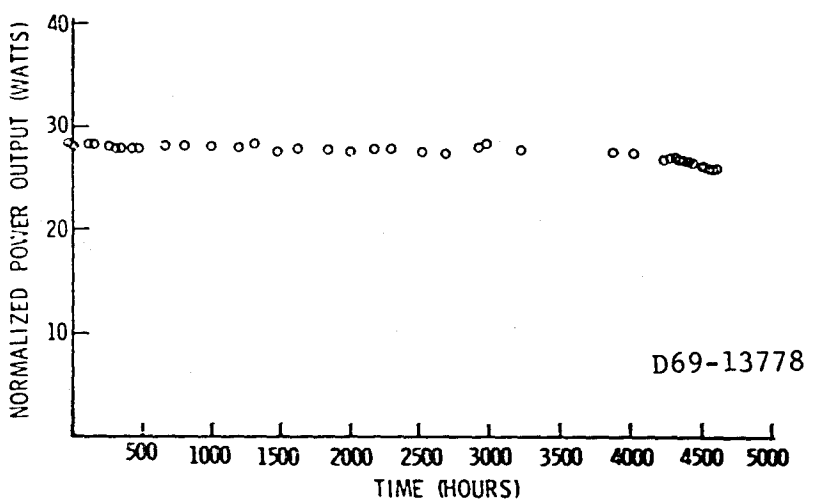

Figure 8. Life test history of GC02 Isotec generator

3.4 Tubular Module

The tubular module developed by Westinghouse Astronuclear Laboratory is of a rugged, void-free construction; it requires no cover gas to preserve the PbTe material and apparently has long-life capability. Figures 9 and 10 show test results for three tubular modules with greatest accumulated time on test. These modules represent a cross section of tubular module technology. Table V presents a summary of the measured performance data for several modules as of September 1969. Most life test data have been on first generation modules (TEM-1 S/N-1) which have now accumulated over 30,000 hours of test time. Initially, the degradation of these early modules was quite severe; however, performance has remained constant since about the first 5000 hours. Modules of improved design (TEM 9U-13, 9U-14, 9U-19, 9U-22, and 9Q-1) show less initial degradation, but test times are for considerably shorter periods.

TABLE V

Summary of Performance Data
for Tubular Module as of September 1969

TEM Module	Present Test Conditions	$\bar{T}_H$ (1) (°F)	$\bar{T}_C$ (1) (°F)	Circuit Resistance (milliohms)	V_{oc} (V)	Q_H (4) (W)	P_e (W)	Circuit Efficiency (%)	Elapsed Time (hr)
1-1	CT,ML	992	415	171.6	5.98	120/1485/120	52.0	3.78	33,317.0
9E-2	CP,OC	1048	398	188.8	6.88	140/1280/140	60.1	4.25	24,377.6
9U-13	CT,ML	1198	389	85.7	7.35	250/2130/200	157.8	8.01	1,342.1
9U-14	CT,ML	1101	430	83.7	6.35	250/1765/200	120.5	7.37	933.8
9U-19	CT,ML	1101	386	89.6	6.37	250/1766/200	113.1	6.95	4,677.0
9U-22	CT,ML	1100	388	96.1	6.48	175/1790/175	109.3	6.61	4,977.9
9Q-1	CP,ML	1013	392	8[illegible].3	4.99	140/1520/140	75.4	5.38	17,436.1
9A-3 (2)	CT,OC	1190	394	71.6	--	250/1540/000	--	--	5,059.1
9M-2 (2)	CT,OC	1153	389	102.9	--	230/1520/230	--	--	5,198.1
10-3 (3)	CP,ML	1032	401	90.4	5.46	2200	82.5	3.75 (3)	8,333.3
10-4	CT,ML	998	392	66.5	5.47	150/2072/150	112.6	5.43	503.3
10-7	CT,ML	1001	391	70.9	5.70	150/2015/150	114.4	5.68	142.5

NOTES:

(1) $\bar{T}_H$ average hot side temperature; $\bar{T}_C$ average cold side temperature.

(2) Nonpower producing materials study module.

(3) TEM-10 S/N-3 on test in heat pipe system. System heat input and system efficiency are listed for this module.

(4) Heat input rates are listed as three separate quantities. Lower guard/main heater upper guard heater power are listed.

CT - tested with $\bar{T}_H$ and $\bar{T}_C$ held constant.

CP - tested with P_i and $\bar{T}_C$ held constant.

ML - matched load test

OC - open circuit test

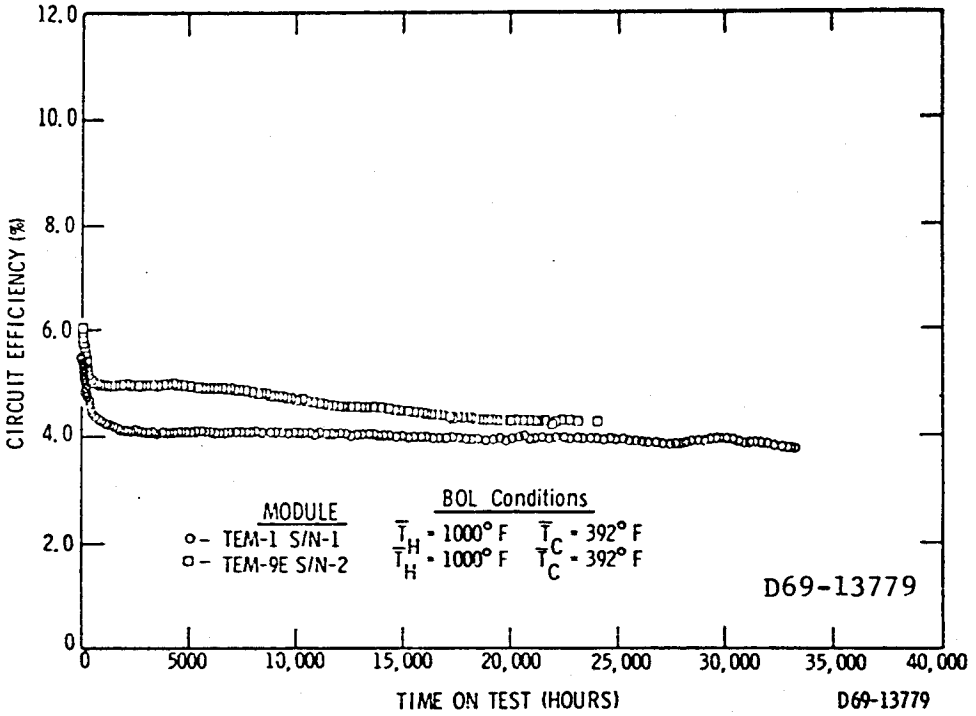

Figure 9. Life test history for TEM-1 and TEM-9E tubular modules

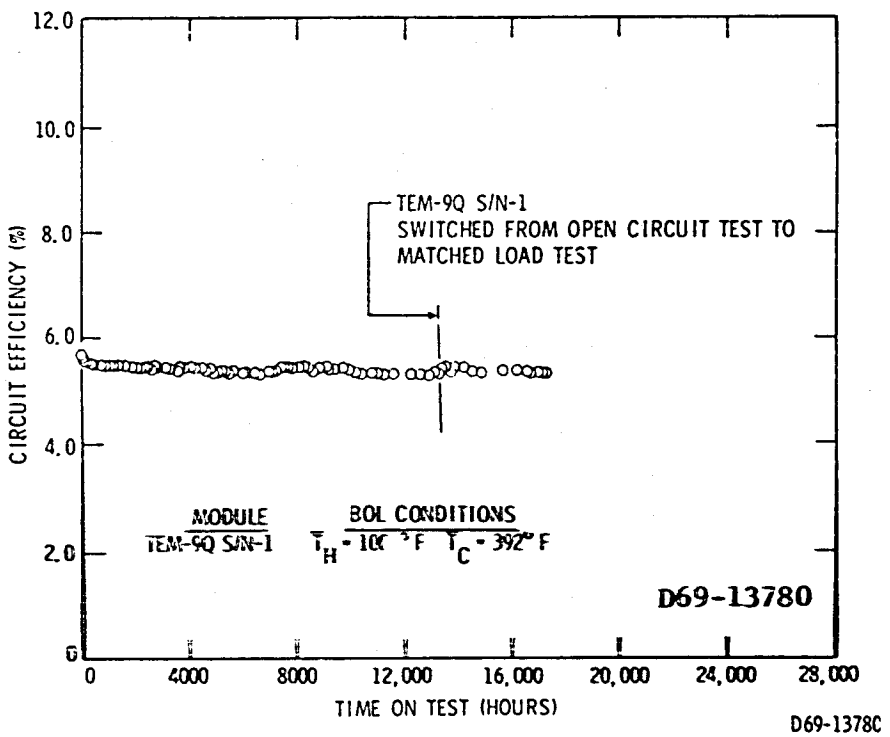

Figure 10. Life test history for TEM-9Q tubular module

Most test modules have been of the 0.50-inch (ID), 1.625-inch (OD) design. Modules of a size practical for fueling with PuO_2 have not been fabricated. Test data from the smaller diameter modules may be applicable to larger sizes in cases where materials are the same; however, one cannot have confidence in a larger ID design until fabrication procedures have been established and long term life tests have been performed.

3.5 Silicon-Germanium Air-Vac Technology

The SiGe Air-Vac thermocouple and converter technology is presently being developed by RCA. Figure 11 is a drawing of a couple using the fleximod construction, typical of the currently available technology. In past programs, RCA has made a large number of Air-Vac couples and modules of various configurations for NASA, DOD, and AEC. A number of these remain on test at various locations. Table VI summarizes the current status of five modules on test at RCA which represent the longest term tests on Air-Vac couples. In addition, Figure 12 shows the performance of a 60-couple "Reference Converter" that was designed and fabricated recently to be used as a reference for future improvements in materials and fabrication procedures.

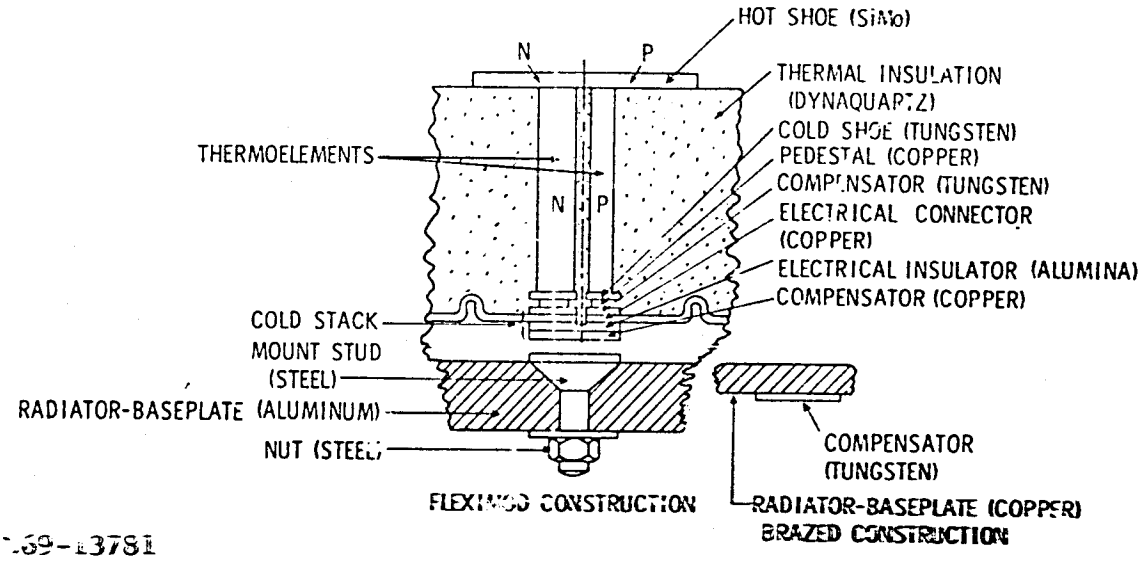

Figure 11. Schematic of SiGe couple

TABLE VI

RCA SiGe Thermoelectric Module
Life Tests - SNAP-17A Modules
as of June 24, 1969

Module Number	Module Hours Total	Number of Couples	Leg Diameter (in.)	Vacuum (torr)	Nominal T_h (°C)	Measured T_h (°C)	Measured T_c (°C)	Measured ΔT (°C)	Normalizing ΔT (°C)	R mΩ Per Couple	E_{oc} mv Per Couple Normalized	Po mw Per Couple Normalized	Percent of 500 Hour Po
MTA4-5	41781	6	0.230	7.10^{-7}	850	847	258	589	606	88.0	304	262	97.0
MTA7-10	40323	6	0.275	3.10^{-6}	850	848	281	567	606	74.0	315	335	89.8
MTA8A-9	39827	4	0.275	3.10^{-6}	1050	1062	359	703	606	72.0	358	451	94.9
MTA8A-18	37868	6	0.275	7.10^{-6}	825	822	281	541	612	74.0	355	426	105.1
MTA9-1	40452	14	0.275	Air	825	795	196	599	612	73.1	290	296	88.1

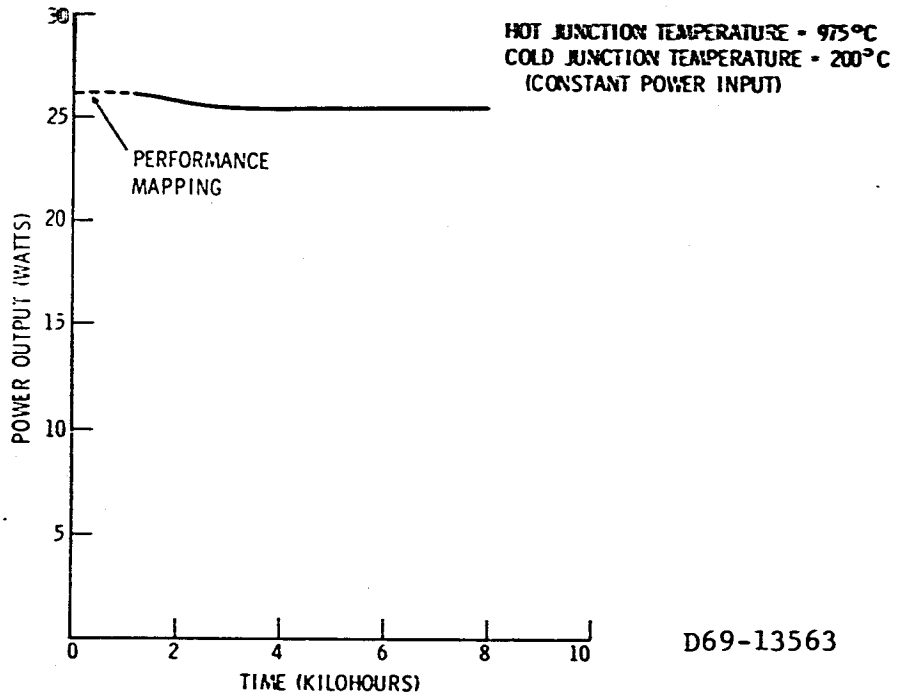

Figure 12. Power output history of Air-Vac reference converter

An important advantage of SiGe thermoelectric devices is the ability to operate in vacuum for long periods without a protective cover gas. Only the tubular module and Isotec panels offer this capability among the PbTe converters.

3.6 TAGS Thermoelectric Material

The Isotopes Incorporated TAGS material has been used with PbTe in SNAP-29 modules and SNAP-19 generators.

Work was recently initiated to test an electrically heated model of the SNAP-29 RTG along with the life-performance tests of the T/E module-radiator assemblies. Two of the T/E module-radiator assemblies (SN001 and SN002) have been under test for approximately 4200 hours (180 days). Figure 13 shows the corrected output power history of these two modules. Two other T/E module-radiator assemblies (SN007 and SN008) began life-performance testing early in this quarter but failed after approximately 100 hours of operation. In both cases, the NaK diaphragm developed a leak, and the NaK shorted out the T/E element circuit within the module.

The RTG model has been operated approximately 1800 hours; however, only one of the four T/E modules has been producing significant electrical power. The other three modules exhibited symptoms of NaK diaphragm failure similar to T/E module-radiator assemblies SN007 and SN008. This testing was discontinued because the power history data are not considered reliable due to wide temperature variations.

Six SNAP-19 generators (Nos. 26, 27, 28, 29, 30, and 31) were fabricated with TAGS elements. Of these generators only one, No. 26, has had any appreciable testing. After approximately 3900 hours on test, there has been no discernible change in power output. Beginning of life power was 43.7 watts for hot and cold junction temperatures of 1046° and 660°F, respectively.

3.7 TPM 217 Thermoelectric Material

The 3M Company's TPM 217 thermoelectric material appears to offer higher efficiency than PbTe. However, at the present state of development only the p-type material has been made. Isothermal and thermal gradient compatibility tests have been started with assorted bulk insulation and candidate metallic contact materials, and exploratory development of n-type compounds is planned for FY70. No couples using TPM 217 p-type material have been fabricated in this program, and, therefore, no life test data are available for this material.

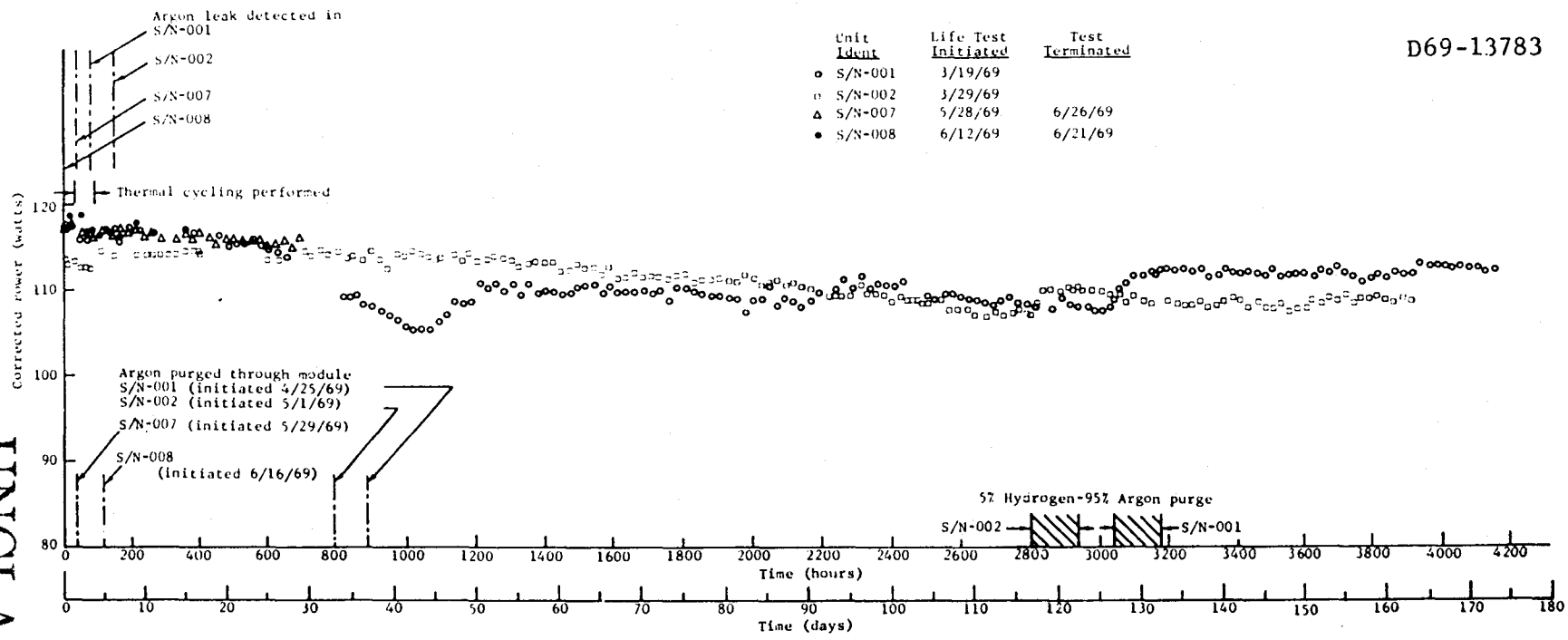

Figure 13. Corrected power history of SNAP-29 TAGS diaphragm modules; (T_{HJ} = 1050°F, T_{CJ} = 375°F)

3.8 Generator Efficiencies

Efficiency is another factor in overall generator performance. For a comparison of the potential capability of generators, Figure 14 presents a plot of theoretical material efficiency as a function of hot and cold junction temperatures for SiGe and PbTe materials. Included in the graph of Figure 14 are points indicating material efficiency for several practical generators. The number in parentheses is the actual efficiency achieved by the generator at the temperature shown.

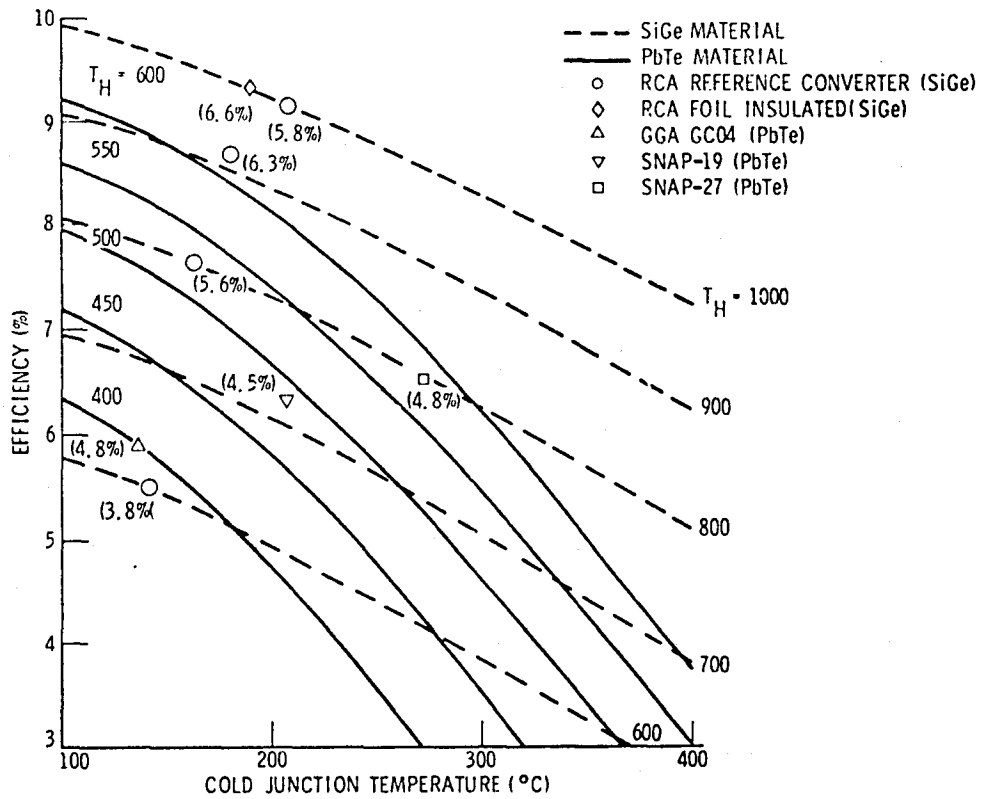

Figure 14. Graph of theoretical efficiency for SiGe and PbTe materials as a function of temperature

To realize the full potential of SiGe thermoelectrics, the hot junction should operate above 1000°C, probably over 1100°C. However, SiGe generators are competitive at much lower temperatures. As indicated in Figure 14, an SiGe generator with a cold junction of approximately 200°C and a hot junction of 800°C has achieved an efficiency of 5 percent. This is competitive with PbTe generators.

3.0 Summary of Thermoelectric Material Test Data

Test data for the different modules and converters under life test reveal that the SiGe Air-Vac modules give the longest and most stable results. The next oldest units are the early Westinghouse tubular modules at over 30,000 hours and the SNAP-27, 1100°F, 10-couple modules at 25,000 hours. Degradation in these latter devices is greater than in the SiGe modules.

The SNAP-27 10-couple modules have shown fairly stable performance at a 1050°F hot junction temperature and therefore give some credibility to the long-life capability of a SNAP-27 type converter at this temperature. However, the need to maintain integrity of the sealed system during operation tempers expectations that may result from these findings.

Test results for the tubular module show data out to approximately 30,000 hours for some modules; however, data for the improved modules are for considerably shorter periods. No modules of the size practical for fueling with ^{238}Pu have been fabricated.

The mechanical and metallurgical bonding technique for the Isotec converter has apparently improved the life potential of this low temperature, unsealed system. However, more testing will be needed before confidence in a long-life capability can be established.

New thermoelectric material, TPM 217, has not been sufficiently characterized or tested to allow any conclusions to be drawn about its ultimate capability.

TAGS thermoelectric material has demonstrated capability for improving generator performance, but only short term data are available.

4.0 Heat Source Technology

4.1 Fuel and Materials Compatibility

In considering the compatibility of fuels and capsule liner materials for use in the multihundred-watt generator, it is assumed that SiGe thermoelectric conversion will be used and that the mission

life may be as short as 3 years or as long as 12 years. These are very important constraints because these combinations of temperature and operating lifetime strongly influence fuel and materials selections. One should realize that the bulk of the available data on fuels compatibility is based on relatively impure (up to 2 percent by weight nonactinide impurities) plasma torched $^{238}PuO_2$ in microsphere form. At this writing, there is a scarcity of compatibility data on the more recent cermet, solid solution, or cermet-solid solution $^{238}PuO_2$ fuel forms. Most of the existing data are for 1200°C or less; however, there are current experiments which will extend the temperature and time base of the data and will include $^{238}PuO_2$ microspheres with markedly reduced impurity levels.

For fuel liner interface temperatures in the 1000° to 1200°C range, and for the 12-year service life, there is considerable reservation about using tantalum base materials as a liner. Also, in this temperature range platinum and platinum/rhodium alloys appear to react with substoichiometric PuO_2, and this presents a potential problem for platinum or platinum/rhodium liners in vented capsules. Therefore, one can eliminate from serious consideration at this time two of the materials that have proven useful as capsule liners. There are, however, at least three materials that appear to be potentially useful for these service conditions: (1) molybdenum and molybdenum base alloys, (2) tungsten and tungsten base alloys, and (3) iridium. Molybdenum has been chosen as the matrix material for the cermet fuel form. Based on BMI data, there appears to be no reaction between PuO_2 microspheres and molybdenum for 96 hours at 1500°C or for 24 hours up to 2000°C. Tungsten appears to be more reactive than molybdenum above 1500°C. There are very little data for either molybdenum/rhenium, tungsten/rhenium, or tungsten/molybdenum/rhenium, but more compatibility data on these materials should be forthcoming from the Mound Laboratory compatibility experiments. Data for iridium are almost completely lacking; however, it has been reported by some experimenters that iridium does not react noticeably with PuO_2 in a reducing atmosphere of H_2. Based on this rather meager evidence, one might anticipate that iridium may prove useful as a capsule liner, and there are plans to obtain Ir/PuO_2 compatibility data at Mound Laboratory.

4.2 Helium Release from $^{238}PuO_2$ Microspheres

Safety considerations will most likely require that a heat source for the multihundred-watt generator be capable of venting the helium generated during decay of the fuel. This is the safety philosophy currently used on the Transit capsule. Knowledge of the helium release mechanism from the fuel is important in designing a vented capsule and in understanding the safety problems associated with the helium buildup; however, little information is available. While research on the helium release from $^{238}PuO_2$ plasma torch fused (PTF) microspheres has continued for over a year at such laboratories as LASL, ORNL, and MRC, the exact mechanism for the gas release has not been determined. Several very different types of behavior, including classical gas release, small quick spike release, and broader burst release, have been observed. None of these are unequivocally explained by impurity and porosity effects, radiation damage effects, or simple surface effects on the "classical" interstitial-interstitial diffusion which might be expected from a size analysis based on the dimensions of the PuO_2 lattice interstitial site and of the helium atom. It is, in short, a baffling phenomenon, complicated by many factors.

An approximation to the release behavior has been formulated using a simple diffusion model which does reproduce in a very general manner the results seen experimentally. It must be remembered, however, that this model does not predict bursts and/or "spikes" of gas release, and that it is really appropriate only to those situations in which the major part of the gas release occurs in a time short compared to the total generation time of the gas.

Initial calculations on early data indicated that the gas release was approximately that predicted by Fick's second law for diffusion with no source term:

$$\frac{\partial C(r,t)}{\partial t} = D\nabla^2 C(r,t) \qquad (1)$$

where $C(r,t)$ is the concentration of gas at radius r and time t, and D is the diffusion coefficient.

Assuming that the diffusion coefficient is temperature dependent, with the form

$$D = D_o \exp - \frac{E}{RT} , \qquad (2)$$

the diffusion coefficient data were plotted as ln D versus 1/T(K). The plot yielded, at temperatures greater than approximately 800°C, a mean activation energy of

$$E = 49.2 \text{ Kcal/mole} \pm 10\%$$

and a mean pre-exponential term at

$$D_o = 3 \times 10^{-3} \text{ cm}^2/\text{sec} \pm 600\%.$$

The error limits noted reflect the large scatter of data, and represent one standard deviation. Eighty percent of the experimental data points lie within the maximum and minimum boundaries set by these limits.

In those cases where sufficient data were available at lower temperatures (less than ~600°C), an examination of the ln D versus 1/T plots indicated that there are probably two branches to the temperature dependence of D. The form is

$$D = D_{LO} \exp\left(- \frac{E_L}{RT}\right) + D_{HO} \exp\left(- \frac{E_H}{RT}\right). \qquad (3)$$

The phenomenological effect of this two-term diffusion coefficient is to allow a small, but not necessarily negligible, diffusion coefficient at temperatures less than about 600°C and yet yield the higher diffusion coefficient to explain the accelerated release seen at temperatures of 1000°C and above.

Table VII shows calculated values for the two-term diffusion coefficient at several fuel temperatures and the approximate time for 90 percent of the total helium inventory. It must be pointed out that this is a simple diffusion approximation which neglects the helium generation. Thus, the times given in column four are underestimates, particularly for the lower temperatures where the steady-state condition

might be reached before 90 percent release. (One experiment at ORNL indicated that at 800°C = 1472°F approximately 48 days were required to reach a steady state.)

TABLE VII

Calculated Values for Two-Term Diffusion Coefficient at Various Fuel Temperatures

T (°F)	D (cm^2/sec)	Approximate Time for 90 Percent Release (days)
800	7×10^{-16}	(2×10^4)
1200	8×10^{-15}	(1×10^4)
1600	1×10^{-12}	100
2000	4×10^{-11}	3
2400	5×10^{-10}	0.2

4.3 LRHS Capsule Program

The LRHS capsule program was established to develop the technology for an isotope fuel capsule capable of operating in a space environment at a surface temperature of 2000°F for 5 years or more. The major objectives have been the design, fabrication, and evaluation of a test capsule whose design takes into account the operating requirements of a typical heat source. Accomplishment of these objectives has included process, fabrication, and test activities.

The principal tasks which have been pursued include program management, analytical evaluation, capsule design, process development, fabrication and assembly, biaxial creep testing, coupon compatibility, coating development, impact testing, fire testing, and posttest evaluation.

The materials investigated for the basic test capsule design were the tantalum alloys Ta-10W for the fuel liner and T-111 for the structural member, and the noble metal alloy Pt-10Rh for an oxidation clad. Both chemical vapor deposition (CVD) and plasma arc sprayed

(PAS) Al_2O_3 coatings were evaluated for use as a diffusion barrier between the c' le metal clad and the T-111. Iron titanate (Fe_2TiO_5) was the reference emittance coating.

Technology development has been successful in these areas, and the pertinent results are described below.

4.3.1 Process Development, Fabrication, and Assembly -- Significant advances were made in the fabrication of the tantalum alloys. Processes for deep drawing and coining or stamping the alloys were developed and used in the capsule program to provide reproducible, high-quality production parts. Similar processes were developed for the noble metal alloy parts. Further, the process and assembly steps for production of the capsules were accomplished; this included qualification of tooling, weld procedures, application of coatings, nondestructive testing, and materials and process specifications.

4.3.2 Compatibility -- A three-temperature by three times exposure matrix was used to study the interfaces of interest. Results for as much as 1000 hours and 2500°F indicate that there are no likely problems at the operational temperature of 2000°F and for the environment as studied. Higher operational temperatures and/or the presence of additional components such as carbon are likely to promote reactions between the coatings and their substrates.

4.3.3 Coating -- An acceptable procedure was developed for CVD Al_2O_3 on a T-111 substrate without degradation or significant contamination. Development was done on subscale specimens. The process for application of PAS Al_2O_3 coatings to T-111 was also developed, and this process has been used extensively in the test program.

4.3.4 Creep -- The original test program was initiated to establish the basic biaxial creep properties of the T-111 structure and the interplay of the composite capsule under the influence of a continuously increasing stress (such as would be the case for a real fuel capsule containing an alpha emitting fuel). The biaxial testing was conducted on three capsule configurations at 2000°F in a vacuum of 10^{-8} torr or less. A number of capsules were tested, and a preliminary creep correlation was evolved.

4.3.5 Impact -- Prototype capsules were impacted on granite at velocities between 250 and 350 feet per second. The structural wall thickness was varied from 90 to 127 mils, the impacts were principally end-on, and the impact temperature was approximately 1200°F. In all tests the fuel annulus remained intact and the structural members experienced bends of less than one wall thickness without cracking.

The technology development in this program is continuing. Studies of alternate materials fabrication and size fluctuation are to be investigated. Additional impact tests will include aged and pressurized capsules, capsules with thinner walls, and capsules impacted at higher temperatures. Biaxial creep results will help identify possible interplay between coatings and structures. Additional compatibility tests and emittance coating work will be conducted.

4.4 Coating Development

Coatings technology exists for both diffusion barriers and emittance coatings at approximately 2000°F. However, the emittance coating area is presently clouded by degradation of the coatings during total mission exposure. Also, presently characterized diffusion barriers such as Al_2O_3 may exhibit significant reactions at slightly higher operational temperatures or during reentry.

4.5 Transit Capsule Program

The Transit program at TRW Systems, Inc., is scheduled to develop a 30-watt 5-year generator by April 1971. The first fueled capsule for the converter is scheduled for delivery by Mound Laboratories in October 1970. To meet these dates, TRW has chosen to use current technology as much as possible for both the converter and the heat source.

The heat source technology to be used will be that developed by Atomics International on the Large Heat Source Program (AEC Contract AT(29-2)-2338). Only minor modifications will be made to the capsule configuration to accommodate the increased fuel loading. Except for the outer emissive coating, the capsule material will remain the same.

The major perturbation to the capsule design is the incorporation of a vent/burst disc into the capsule wall. Although vents are not yet considered state of the art for long-term operation, the present safety philosophy is that some vent system, even if less than desired, is safer than a completely sealed capsule.

Current data indicate that the Ta-10W capsule liner material is compatible with PuO_2 at the 1600°F operating temperature of the capsule. These data and those generated on other capsule characteristics in the Large Heat Source Program give confidence that a capsule life of at least 7 years can be obtained with current technology. The major uncertainty is the long-term operation of a vent.

4.6 SIREN Capsule Program

The SIREN capsule concept embodies a protective layer of carbon or graphite yarn, wound in the manner of a golf ball, surrounding a central fuel body. Following the winding operation, graphite is deposited on the individual carbon fibers, forming a rigid, heat-resistant coating of any desired thickness. At present, the technique has been applied only to spherical and near-spherical capsule geometries.

A recently completed 4-month Phase I program was directed principally toward the development of a hollow capsule into which PuO_2 microspheres could be poured through a loading port. The proposed Phase II program will concentrate on "hot" winding capsules around solid fuel bodies made of either cermet or solid solution PuO_2.

4.7 Summary of Heat Source Technology

The continuing capsule development program at Atomics International has demonstrated the potential for long-life (3 to 12 years) capability for current capsule designs, at least for operational temperatures near 1600°F. However, there is a need for much more fuel liner compatibility data, especially long-term data in the 1800° to 2200°F range. Preliminary data indicate that molybdenum, tungsten, and iridium liner materials would be useful for a long-life, 2000°F capsule.

The area of high-temperature coatings, whether diffusion or emittance, definitely requires investigation in the areas of compatibility, stability, adherence, and process development.

Control of the helium in the fuel capsule is a problem caused primarily by lack of long-term data on vents and good understanding of the release of helium from the fuel. Studies have been made to determine the quantity and history of helium release from PuO_2 microspheres. Using a simple diffusion model to analyze the results of helium release experiments on PTF PuO_2 microspheres, an approximate two-term diffusion coefficient can be obtained. This coefficient can be put into standard diffusion equations for those cases where the helium release takes place in a time which is short compared to the total time required to generate the helium. The two-term diffusion coefficient reproduces, to some extent, many experimental results. However, it does not explain all the observed phenomena, such as gas "bursts," nor is it very appropriate for use at long times with continual helium generation. The analysis for longer periods of gas release is more difficult and at present is not understood.

The present safety philosophy for the Transit program dictates the use of some venting system in lieu of a sealed capsule.

The availability of a flight-certified SIREN capsule for an early 1970 mission can only be regarded as speculative. On the basis of information developed during a 4-month Phase I program ending in July 1969, the concept appears very promising, but little work has been directed toward a specific application.

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