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SUMMARY REPORT
FOR
ALUMINUM BONDED LEAD TELLURIDE THERMOELECTRIC MODULE STUDY FOR NON-MAGNETIC AND VACUUM APPLICATIONS

(5 March 1968 – 5 November 1968)

Contract No. NAS 5-11514

Prepared by
Atomics International
A Division of North American Rockwell Corporation
Post Office Box 309
Canoga Park, California 91304

for
Goddard Space Flight Center
Greenbelt, Maryland

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SUMMARY

The 8-month program described herein, covering the period March 5, 1968 to November 5, 1968, has continued the research and development begun on a prior contract, NAS5-10184. That work, performed by Atomics International (AI), a Division of North American Rockwell Corporation, for the Goddard Space Flight Center, had given early but promising results concerning the power density, efficiency, and stability of aluminum-contacted PbTe thermoelectric test modules made wholly of non-magnetic materials and operated in vacuum.

The principal tasks in this contract have been to continue the life tests already in operation, to replace damaged or seriously degraded test modules, to build and operate a large-module efficiency demonstration, to perform a mechanical (vibration) test on one small module, and to evaluate all information obtained, including the results of destructive analyses.

Evaluation of all test results to date indicates that PbTe thermoelectric converters using the bonded aluminum contact technology can be engineered to meet the following specifications:

1) Neutrally magnetic materials throughout
2) Operable in high vacuum without containment
3) Capable of withstanding launch conditions
4) Moderately high specific power (about 6 watts/lb of converter)

5) Moderately high overall conversion efficiency (5 to 6%)

6) Good stability.

Stability in very long-term operation is only partially demonstrated, and requires extended testing. The longest single module test, about 1-3/4 years at about 720°F average hot-junction temperature, if extrapolated conservatively to 5 years, yields a predicted total power degradation of less than 17%. This of course assumes that no abrupt, catastrophic change will occur.

It is concluded that this program substantiates the original optimism concerning the aluminum contact technology, and merits continued life testing.

I. INTRODUCTION

A prior contract, NAS5-10184, was begun in May 1966 and completed approximately 14 months later, in July 1967. The purpose had been to investigate the basic characteristics of the AI proprietary bonded aluminum contact used with 3M Co. PbTe types TEGS-2N, -3N, and -2P; and to place on operational test several 3-couple modules in order to make practical measurements of electrical characteristics and obtain information on operational stability. All aspects were so favorable that it was proposed to continue the small-module tests that were in operation, and to add certain technical tasks of special interest. Reports of this initial program are identified as AI-66-158, AI-66-260, AI-67-99 and AI-67-132.

During the period August 1967 to March 1968, Atomics International continued the operational testing of six 3-couple modules (SMT-1 to -6), in order that the accumulation of operational time would not be interrupted. In March 1968, the program reported herein was initiated. This was to be an 8-month program, continuing the operational tests and adding new tasks as described in II-B below.

II. DISCUSSION

A. STATUS AT BEGINNING OF CONTRACT

1. Tests in Operation

On March 5, 1968, four 3-couple tests were operating and two had been shutdown for repair. Some of the pertinent characteristics at that time are listed:

TABLE I

TESTS OPERATING MARCH 5, 1968

MODULE NO.	(T_H) MAX	TOTAL HOURS	TOTAL DEGRADATION*
SMT-1	750°F	9165	4.65%
SMT-2	700°F	8170	2.55%
SMT-3	800°F	6545	19%
SMT-4	800°F		
SMT-5	750°F	3735	3.7%
SMT-6	750°F		

*Exclusive of damage directly related to operational accidents; see below.

SMT-1 and -2 had suffered very minor damage early in life, from a slight air leak and a mechanical mishap, respectively. SMT-5 and -6, which were in a common test chamber, had experienced a series of more severe air leaks through a cracked metal-glass seal. The total damage was above $5\frac{1}{8}\%$, and it was feared that this impaired the validity of subsequent test data. Therefore, this test had been shutdown on January 23, 1968, for repair.

2. Test Under Construction

Under the prior contract, NAS5-10184, construction of a 10-watt module had been initiated. On March 5, 1968, the vacuum test chamber for this test was essentially complete, but the Vac-Ion pump, although on hand, required modification to adapt to a new throat size. The module components were nearly complete. During the interim period between contracts, discussions indicated that the technical emphasis should be directed toward efficiency demonstration, rather than power and stability. This change would call for several modifications in the 24 test couples, so as to enlarge fin area, to improve the thermal insulation, and to reduce heat shunting. Altogether, there was a substantial amount of work remaining to be done on both the module and the test station.

B. SCOPE OF CONTRACT, TASKS

In accordance with the contract Statement of Work, and discussions with the technical monitor, the following tasks were assumed:

1) Continue testing SMT-1, -2, -3, -4. This task was straightforward, requiring periodic maintenance, data recording, and reduction.

2) Replace SMT-3, -4, -5, and -6. The damaged modules SMT-5 and -6 would be replaced as soon as possible. SMT-3 and -4, which were showing excessive degradation, were to be replaced at sometime after 1 year of operation.

3) Complete the 10-watt Module Test. This test, LMT-1, was to have moderate priority.

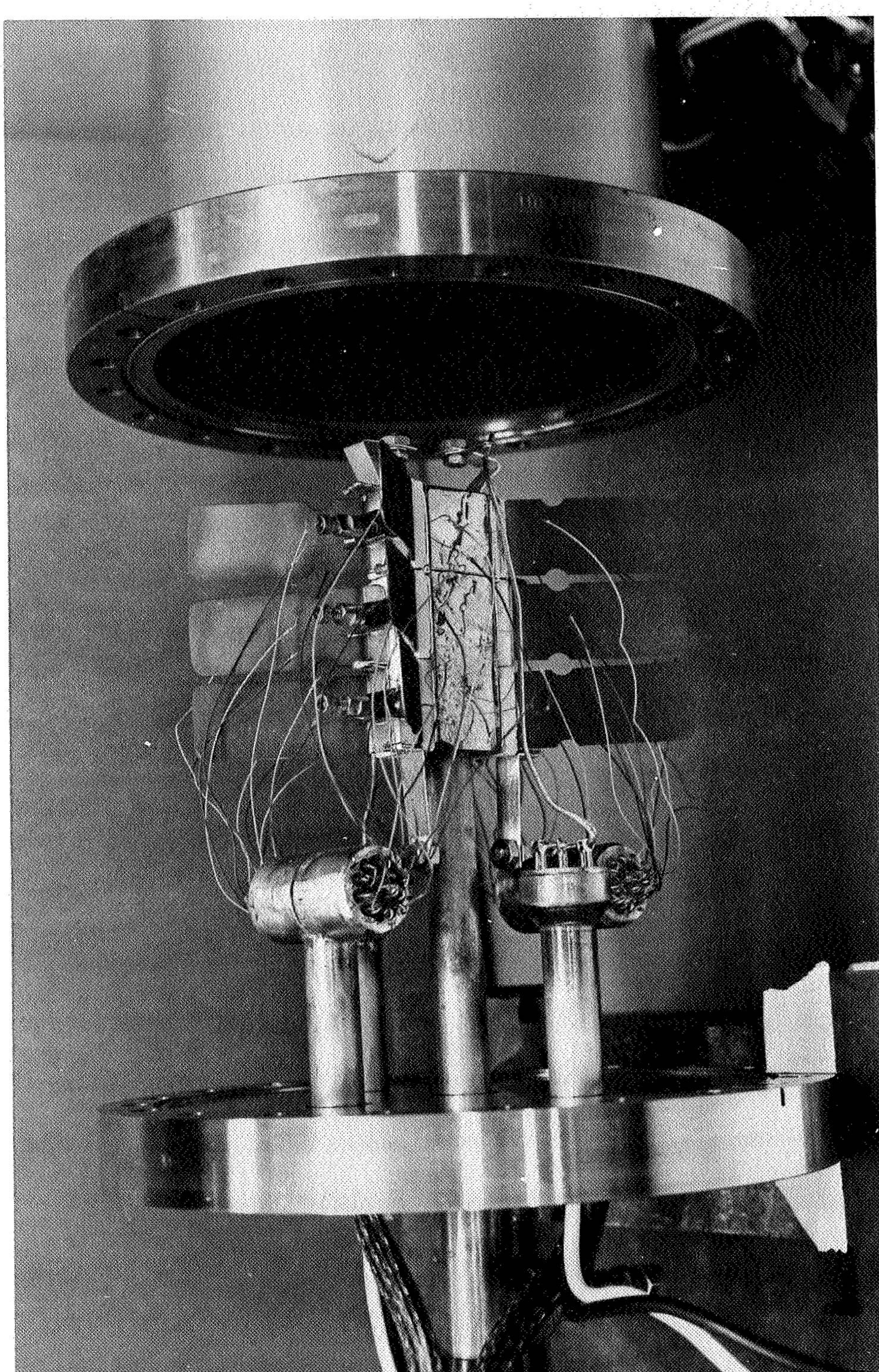

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Figure 1. Two 3-Couple Modules and Test Station

4) Vibration Test. This task required construction of an SMT-type 3- couple module, and vibration testing under both room temperature and elevated temperature conditions.

5) Data Analysis. This was to include the effort necessary to correlate data and prepare graphic or other presentations to illustrate behavior and to disclose trends such as power degradation.

6) Destructive Analysis. Selected elements, to be taken from SMT-3, -4, -5 and -6, were to be subjected to metallography and electron beam microprobe analysis.

7) Reporting and Travel. This required 7 monthly letter reports, and one trip to the Space Flight Center.

C. DESCRIPTION OF NEW MODULE TESTS

1. SMT-7 and -8

These are two 3-couple modules of the type shown in Figure 1, operated in a common vacuum chamber. They were made to replace the damaged SMT-5 and -6. During the replacement period, the test chamber was modified in the following ways:

(a) A longer heater cartridge was installed, to give more uniform temperature distribution (this was achieved).

(b) The heater leads and insulation were changed so as to reduce the shunt-heat path, also aiding temperature distribution.

(c) The metal-glass lead-throughs were moved from the former recessed position in the tank, to an external location. This placed the seals in a cooler zone, and also would permit easy replacement should seal failure occur again.

The two modules, as installed, were of good quality except for high resistance in one P element of SMT-8, as indicated by initial readings. It was felt that shutting down the test to replace one couple was not justified. The defective element has improved slightly during operation. Its resistance was initially 4.36 mΩ, but has improved to 4.10 mΩ; normal is about 3.2 mΩ.

2. SMT-9 and -10

These 3-couple modules also have the appearance of Figure 1. They were made to replace SMT-3 and -4, which had been operated at 800°F (max.) hot-junction temperature for 11,653 hours and experienced nearly 28% power degradation. Analysis of the test data showed that the degradation was almost totally in the P elements. It was speculated further that only the hot junctions were affected. This was confirmed by subsequent destructive analysis.

Atomics International had been investigating concurrently, on an in-house program, the behavior of "2P" elements contacted with tungsten at the hot junctions. About 2 years' test data were available which indicated a stability at 800°F that was 3 to 5 times greater than obtainable with the plain aluminum contact, and with only a slightly higher contact resistance. Permission was therefore sought from and given by the SFC technical monitor, to install tungsten-faced barriers at the "2P" hot junctions of the new modules SMT-9 and -10, between the thermoelectric material and the normal aluminum strap. SMT-9 and -10 are, then, identical with SMT-3 and -4, except that the nominally 0.20-inch long "P" elements are actually shortened to 0.18 inch by inclusion of barriers approximately 0.020 inch thick. This construction is considered to be proprietary.

The test chamber for SMT-9 and -10 was modified slightly, to incorporate the longer heater cartridge and to reduce heat shunts, so that a better temperature distribution would be achieved.

3. <u>LMT-1, 10-Watt Module Test</u>

This module uses the standard aluminum contact technology of SMT-1 through -8. The 24 couples are, however, superficially different. Referring to Figure 2, a closeup photo of part of the module, it is seen that the elements are linearly arranged. The current path is straight, rather than "zig-zagged" as in the SMT construction (see Figure 1). This allows two fins per element, rather than one; they are of the same length, but narrower. Altogether, the fin area per element is larger by 55%. It had been calculated that this increase was necessary in order to compensate for the larger number of couples in relation to the size of the test chamber. Also, a water jacket was installed on the test chamber. In spite of these efforts, the cold-junction temperature of the module could not be held quite as low as desired.

Figures 3 and 4 show the instrumented module ready for insertion in the test chamber. The module is formed of two rows of 12 couples each, mounted back-to-back with a heater between. (The heater eventually used was specially made of 16 folded lengths of commercial, clad heater wire fitted into a 1/2-inch-diameter stainless-steel tube.)

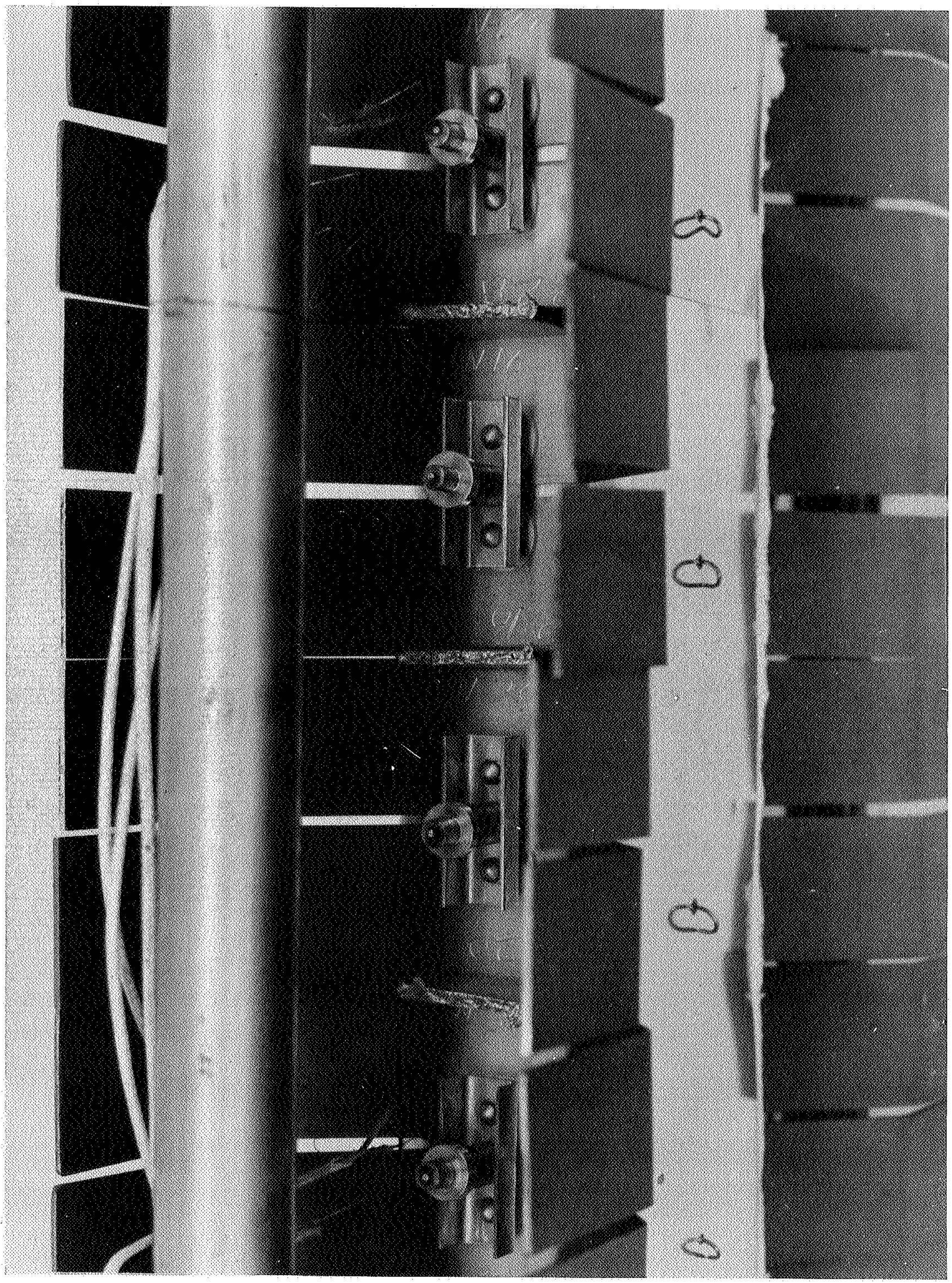

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Figure 2. LMT-1, 10-Watt Module (closeup)

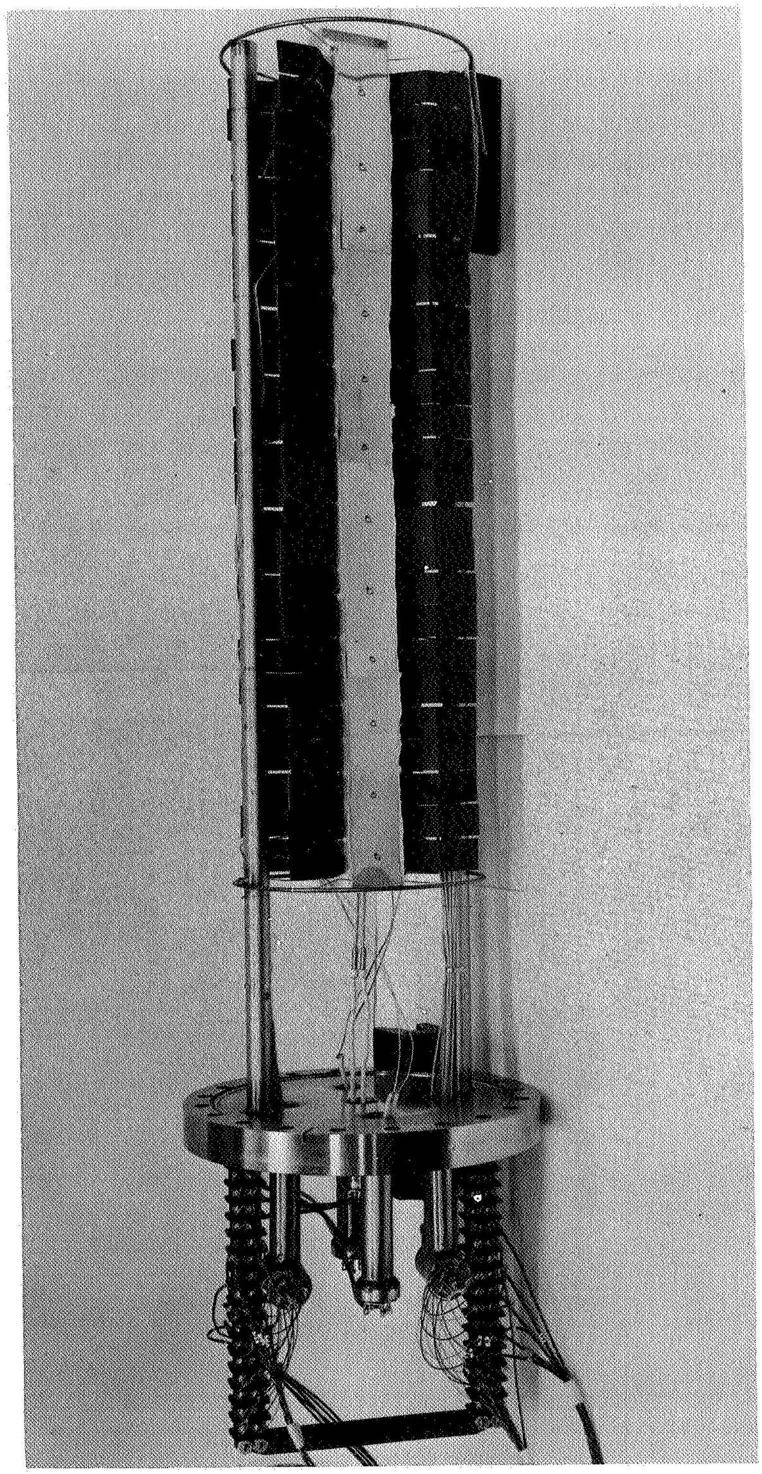

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Figure 3. LMT-1, 10-Watt Module (side view)

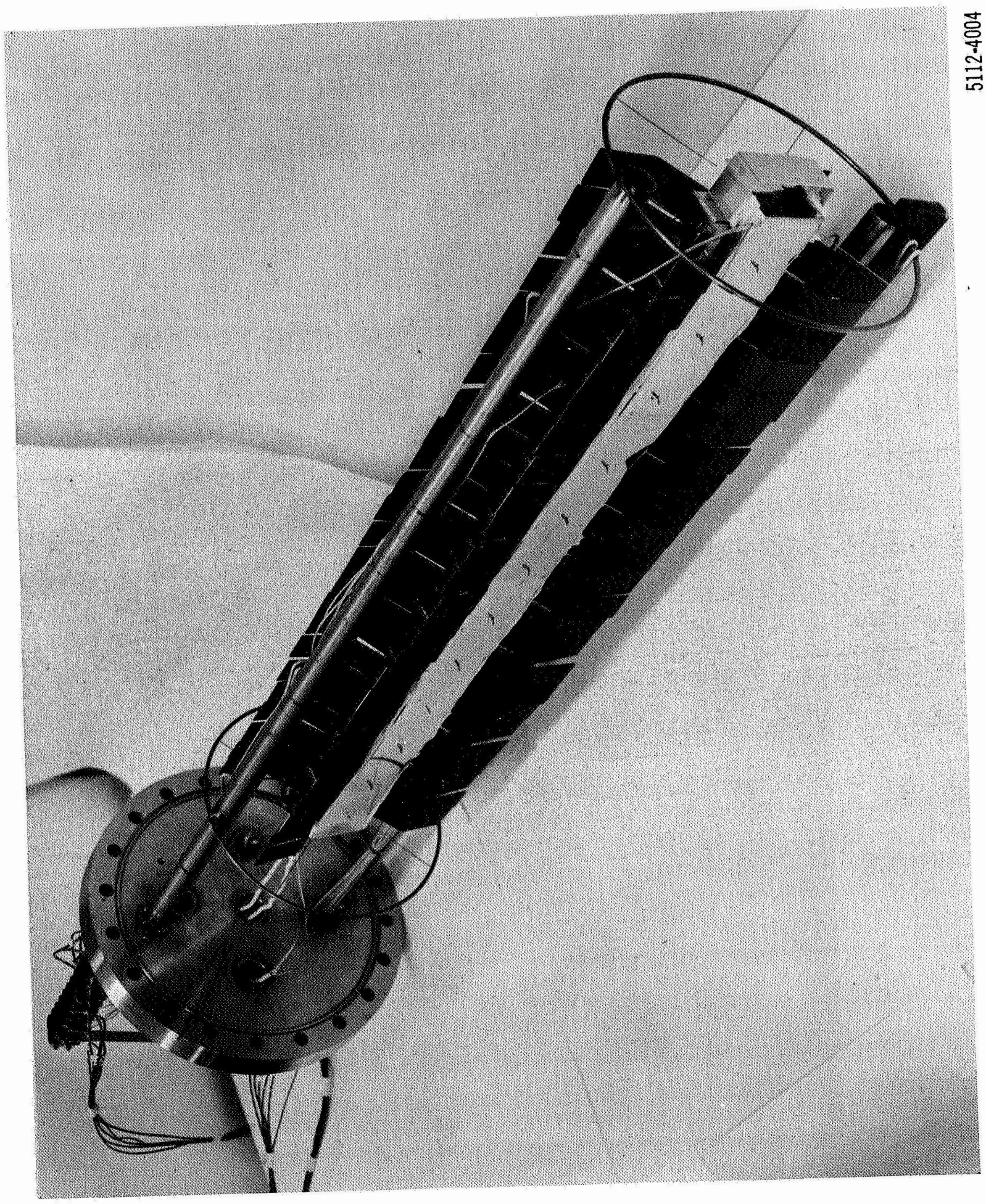

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Figure 4. LMT-1, 10-Watt Module (end view)

The heater and couples are attached to a structural "box" which hangs, in turn, from the rings seen at either end, on 8 fine wires, so that heat shunting is minimized. The whole assembly hangs vertically in the tank, seen in Figure 5, and all instrumentation leads come out at the upper end. The lower end of the tank rests on the throat of a 50 ℓ/sec Vac-Ion pump. Also seen in the photo are the ion pump power supply (below); heater input wattmeter (left); a box (center) containing a coarse-reading ammeter, a precision shunt for accurate reading of current, a Variac heater control, a timer, and a current switch; a coiled-wire resistive load (right side); and a potentiometer to read current (far right).

Referring again to Figures 3 and 4, the side surfaces of the heater box are covered with 1/4-inch-thick quartz-fiber mat, which is covered in turn by gold-coated reflective foils. The latter are anchored by thin wires that are welded to the heater box; the turned-over ends are visible in the photos.

Originally, this module was equipped with 10 each, cold and hot-junction thermocouples. Three of these were lost during assembly operations. Thermocouples were also installed on the hot-wall plate and on the reflective foil.

D. LIFE TEST RESULTS

1. SMT-1

This 3-couple module, described in prior reports (see Section I above), was placed in operation in early January, 1967. Except for several shutdowns connected with operational problems such as ion pump

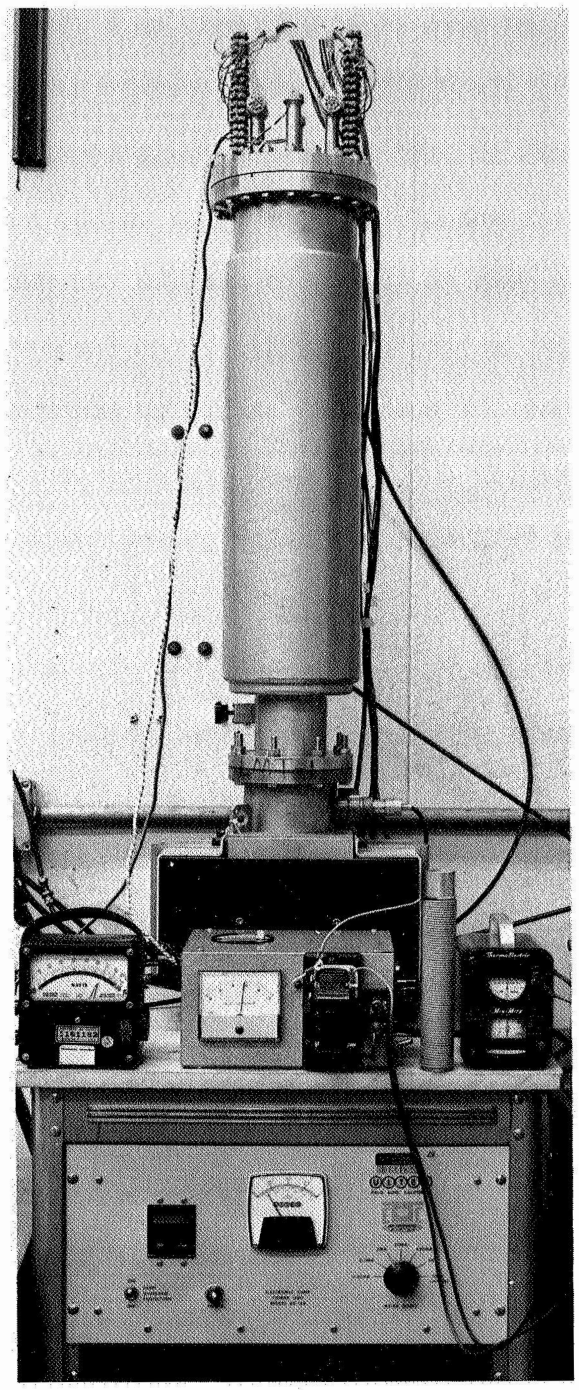

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Figure 5. 10-Watt Module Test Station

overloading, it has been operated continuously since that time. The heat input has been adjusted, as required, to hold the hottest hot junction among the 6 elements in the range 750°F ± 10°F. Typically, the average T_H has been 720°F ± 10°F, and the coldest hot junction has been 700°F ± 20°F. The graph of normalized power output $(P_{max}/(\Delta T^2)$ vs the square root of time is shown in Figure 6. It is observed that the total power degradation at 15,000 hours (except for accidental damage at 2900 hours) is 6.90%, and the current rate is about 0.45%/1000 hrs, or less than 4%/yr. The degradation rate is constantly decreasing according to the expression $dP/dt \propto -\frac{1}{\sqrt{t}}$, so that a straight-line extrapolation can be applied when the square-root time scale is used. At five years, deducting the accidental damage, the predicted power will be about 83% of the initial power. The upper curve of Figure 6 shows that the degradation consists of module resistance increase.

2. SMT-2

This 3-couple module, also described in prior reports, was placed in operation in late March, 1967. It suffered very slight damage during mechanical repair, at 3600 hours. It has been operated almost continuously at (T_H)max = 700°F ± 10°F, (T_H) average = 670°F ± 10°F, and (T_H) min = 625°F ± 10°F. The operational graph is seen in Figure 7. The total power degradation at 14,000 hours is 4.78%, and the current rate is about 0.2%/1000 hrs, or less than 1.75%/year.

3. SMT-3 and -4

These two 3-couple modules, shown in Figure 1, were placed in operation in a common vacuum chamber in late May, 1967. They were

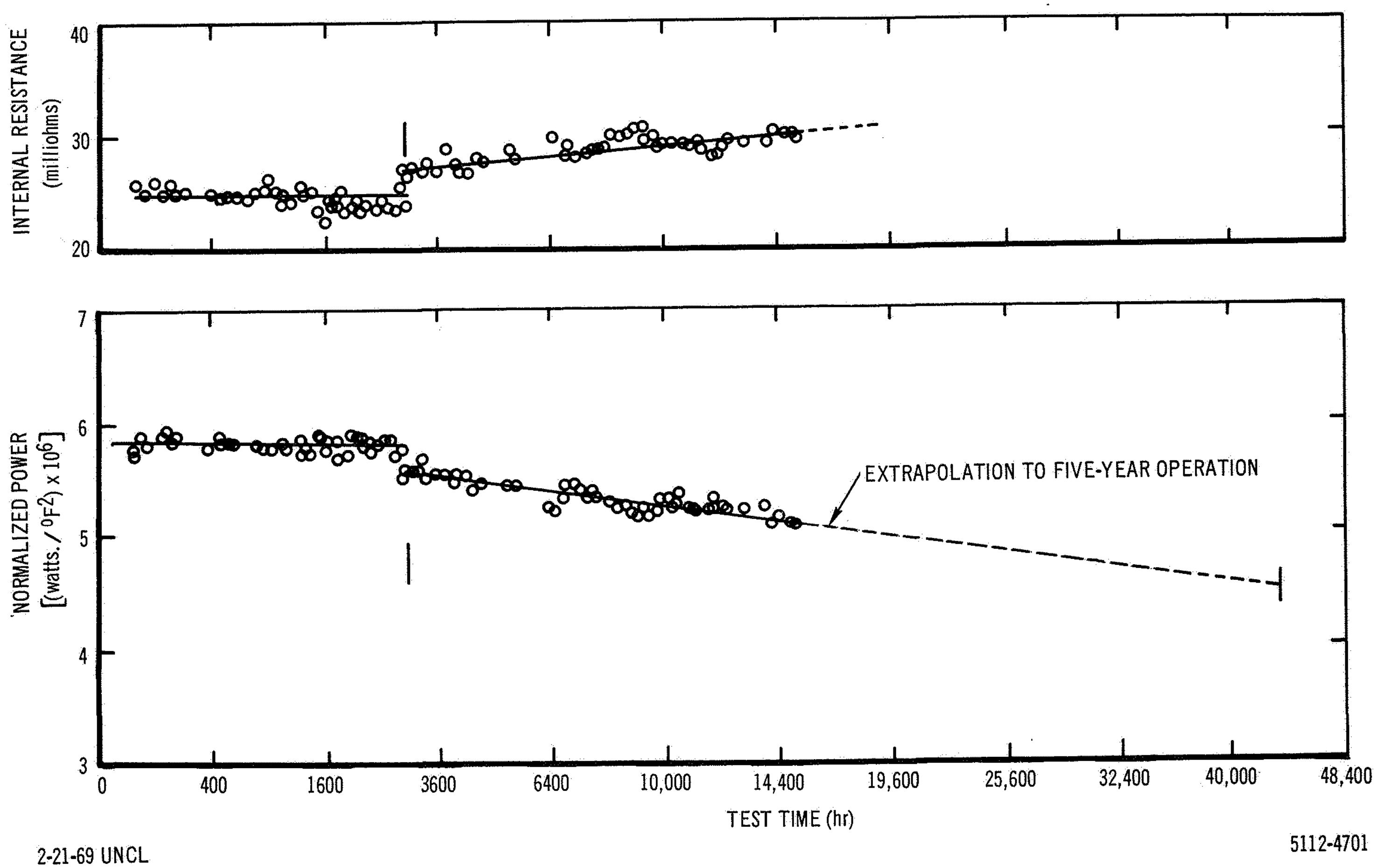

Figure 6. Internal Resistance and Normalized Power of SMT-1 as a Function of (Test Time) $^{\frac{1}{2}}$

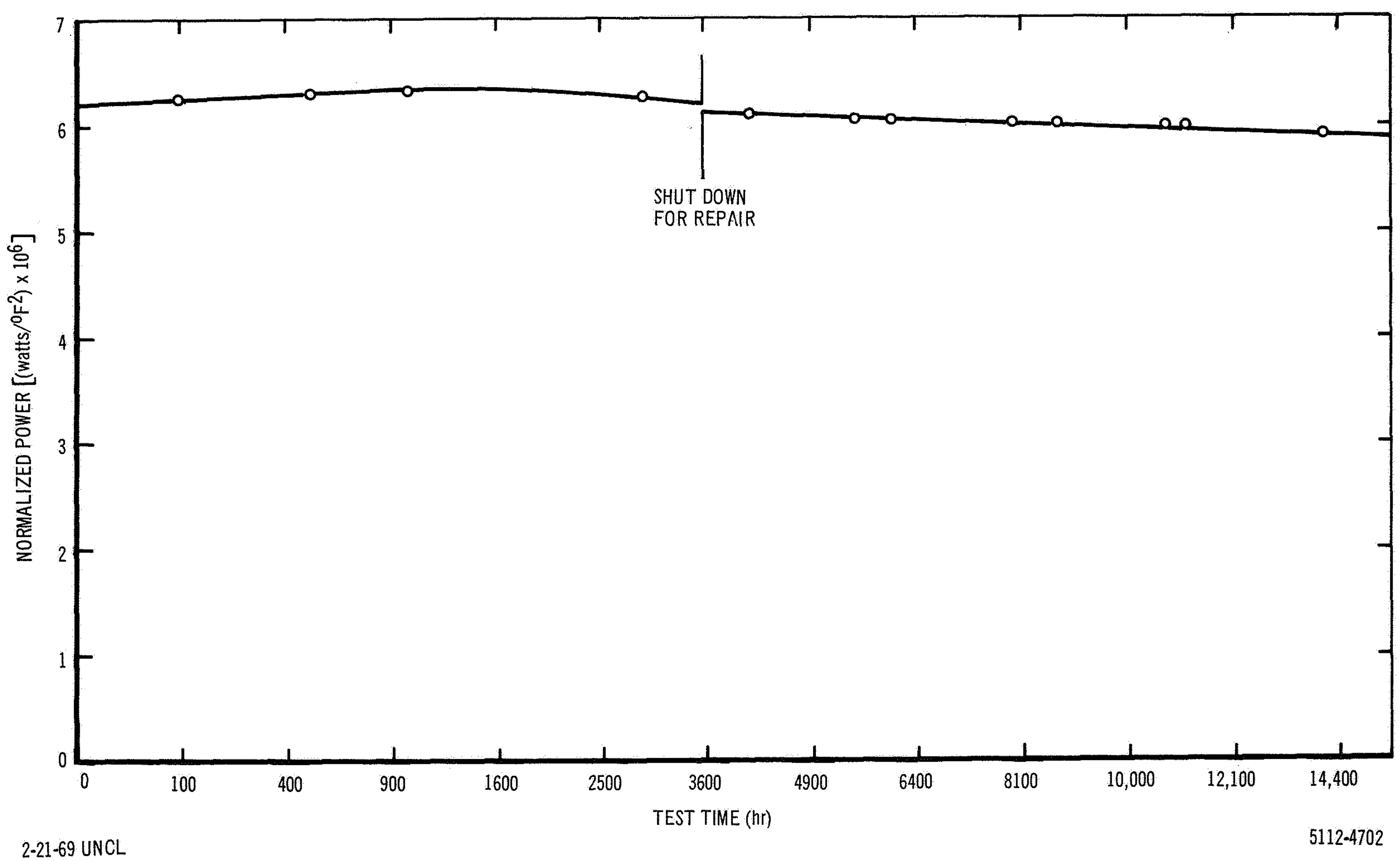

Figure 7. Normalized Power of SMT-2 as a Function of (Test Time)$^{\frac{1}{2}}$

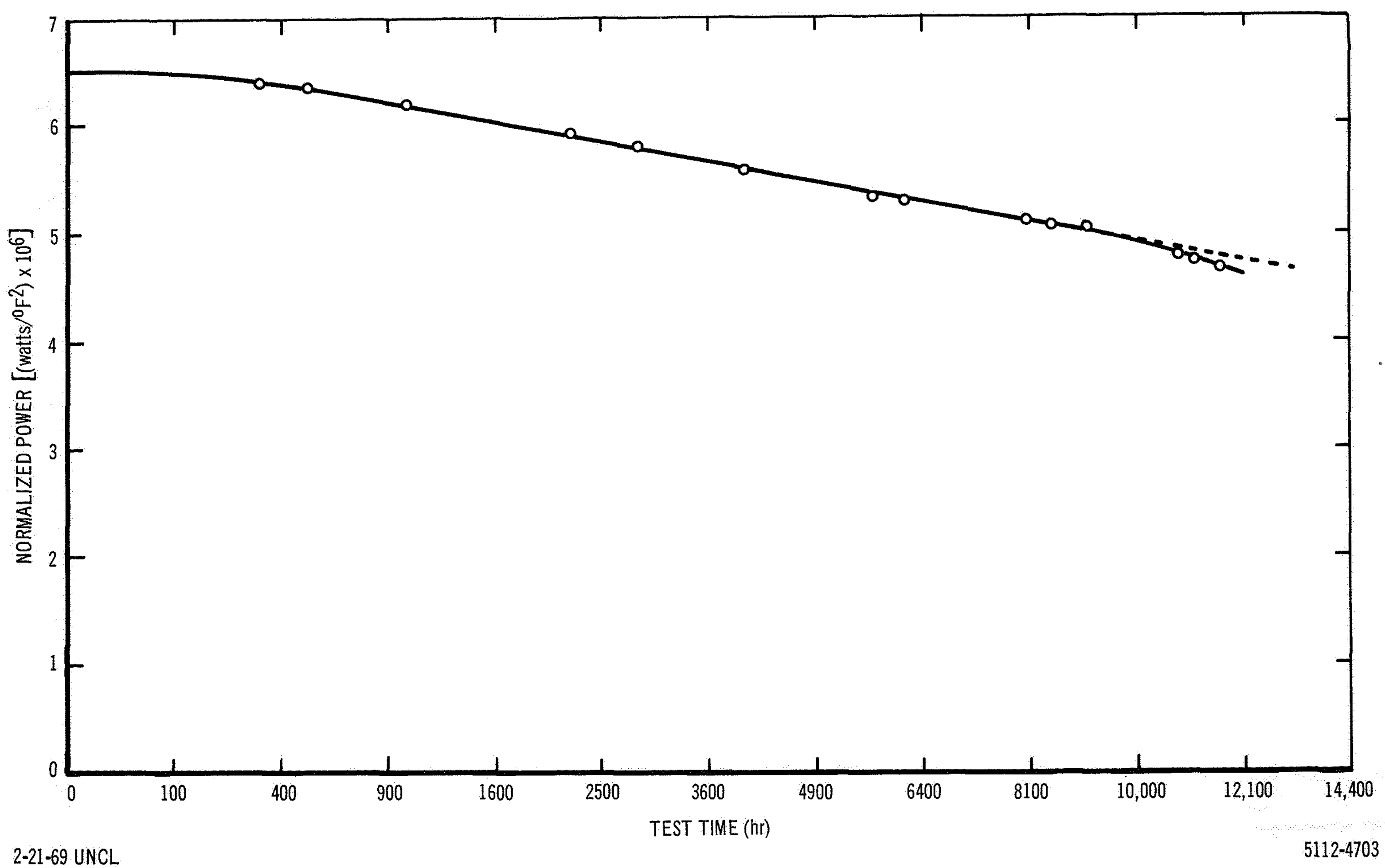

Figure 8. Normalized Power vs (Test Time) $\frac{1}{2}$
Average of SMT-3 and SMT-4

operated continuously, without incident, until early October, 1968. The conditions held have been (T_H) max = 800°F ± 10°F, (T_H) average = 755°F ± 10°F, and (T_H) min = 725%F ± 10°F. The operational graph is shown in Figure 8. It is noted that the power output degraded at a declining rate until 10,000 hours, at which point the total degradation was almost 23%. After 10,000 hours, the rate increased sharply, indicating the onset of serious failure. It has been concluded, of course, that the standard aluminum contact is not suited for long-term operation at 800°F.

It had been continually observed that the degradation was occurring almost exclusively in the "P" elements. An analysis was made of the long-term behavior of the P elements of SMT-2, -3 and -4. It was observed that the normalized power $P_{max}/(\Delta T)^2$, plotted against the square root of time, yielded straight lines. That is, the curves could be fitted to the formula $P = Po\ (1 - \alpha t^{\frac{1}{2}})$, where α was a function of hot-junction temperature of the particular element. It was further predicted, and confirmed, that the degradation constant α could be related to the temperature by the expression $\alpha = \alpha_0 \exp \left(\frac{-E}{RT}\right)$. Figure 9 shows α plotted on a log scale vs reciprocal temperature. The points shown are for the six P elements of SMT-3 and -4, and the two hottest P elements of SMT-2.

The same analysis was attempted for the N elements of these test modules, but was abandoned when the degradation rate was found to be zero within the measurement accuracy, at all temperatures up to and including 805°F.

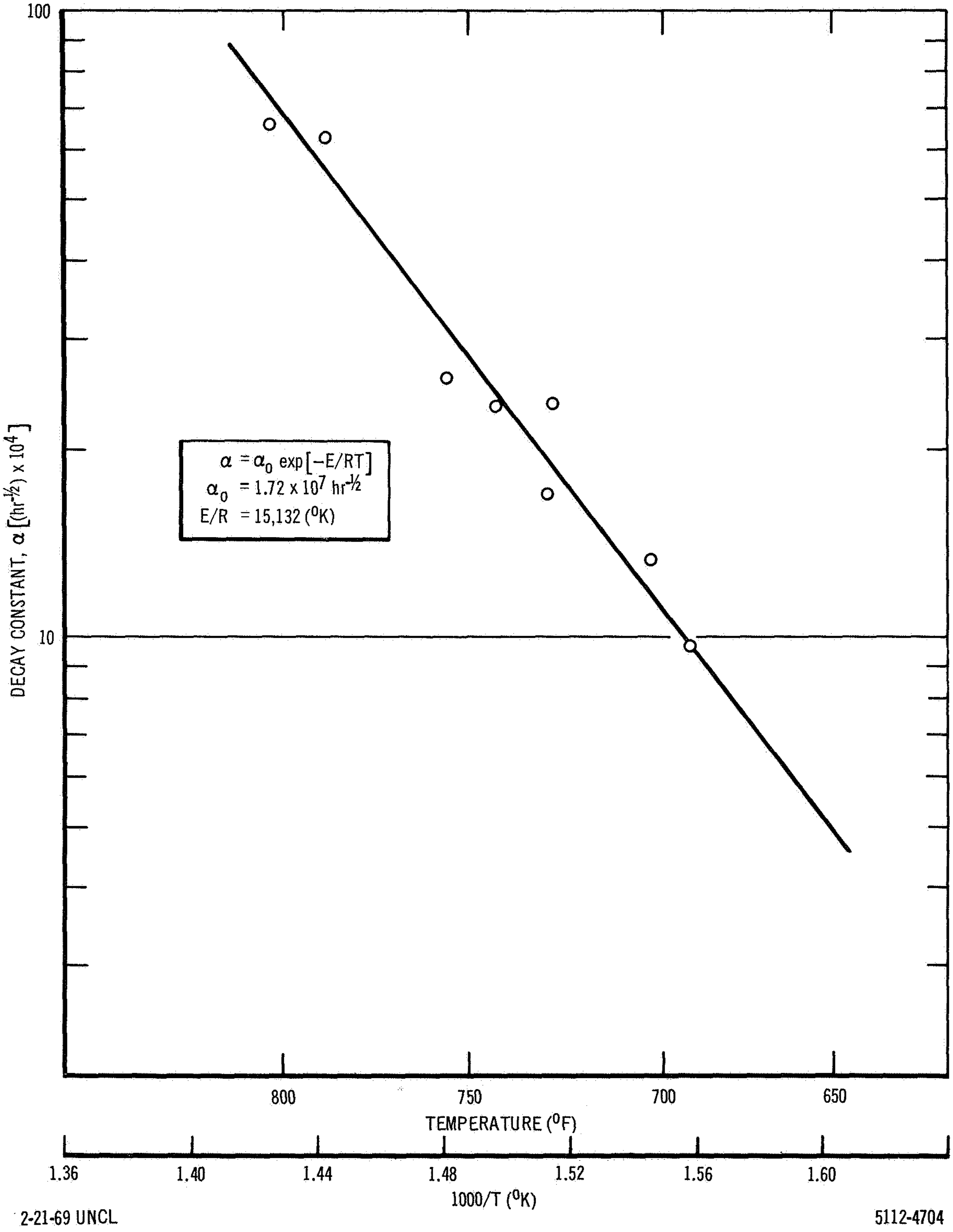

Figure 9. Plot of Decay Constant α as a Function of 1/(Absolute Temperature)

4. SMT-5 and -6

These two 3-couple modules, similar to SMT-3 and -4, were operated from July, 1967 to January, 1968. At that time, during the interim period between funded programs, a series of air leaks caused damage resulting in the decision to terminate the test. The operational graph, Figure 10, is included as a matter of record. No significant operational degradation was observed at (T_H) max = 750°F ± 10°F, in the 3735 hours of operation.

5. SMT-7 and -8

These two 3-couple modules, identical to SMT-5 and -6, were placed in the same (slightly modified) test chamber and put into operation in early July, 1968. The conditions held have been (T_H) max = 750°F ± 10°F, (T_H) average = 740°F ± 10°F, and (T_H) min = 700°F ± 10°F. The operational graph is shown in Figure 11. The total degradation at 3500 hours is about 2.5%, and the current rate is about 0.8%/1000 hrs.

6. SMT-9 and -10

These two 3-couple modules were made as replacements for SMT-3 and -4, the 800°F modules that had degraded rapidly (see II.C.2 and II.D.3 above). The operational graph is shown in Figure 12. Note that the initial normalized power output was approximately 5% higher than in the predecessors, SMT-3 and -4 Figure 8. This can be explained by the 10% shorter length of the "P" legs of the couples; however, the expected loss from tungsten hot-junction contact resistance was not experienced. No explanation for this has been developed.

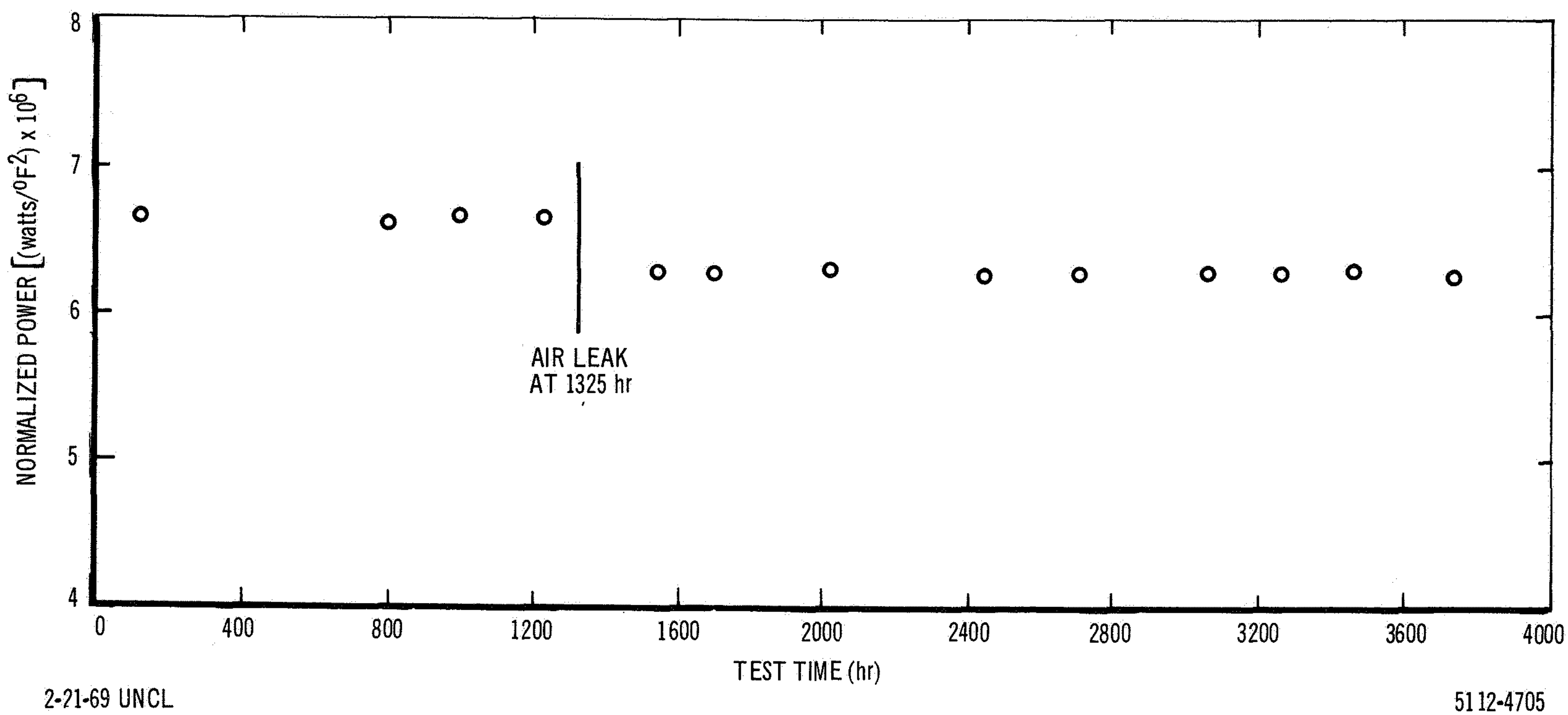

Figure 10. Average Normalized Power vs Test Time for SMT-5 and SMT-6

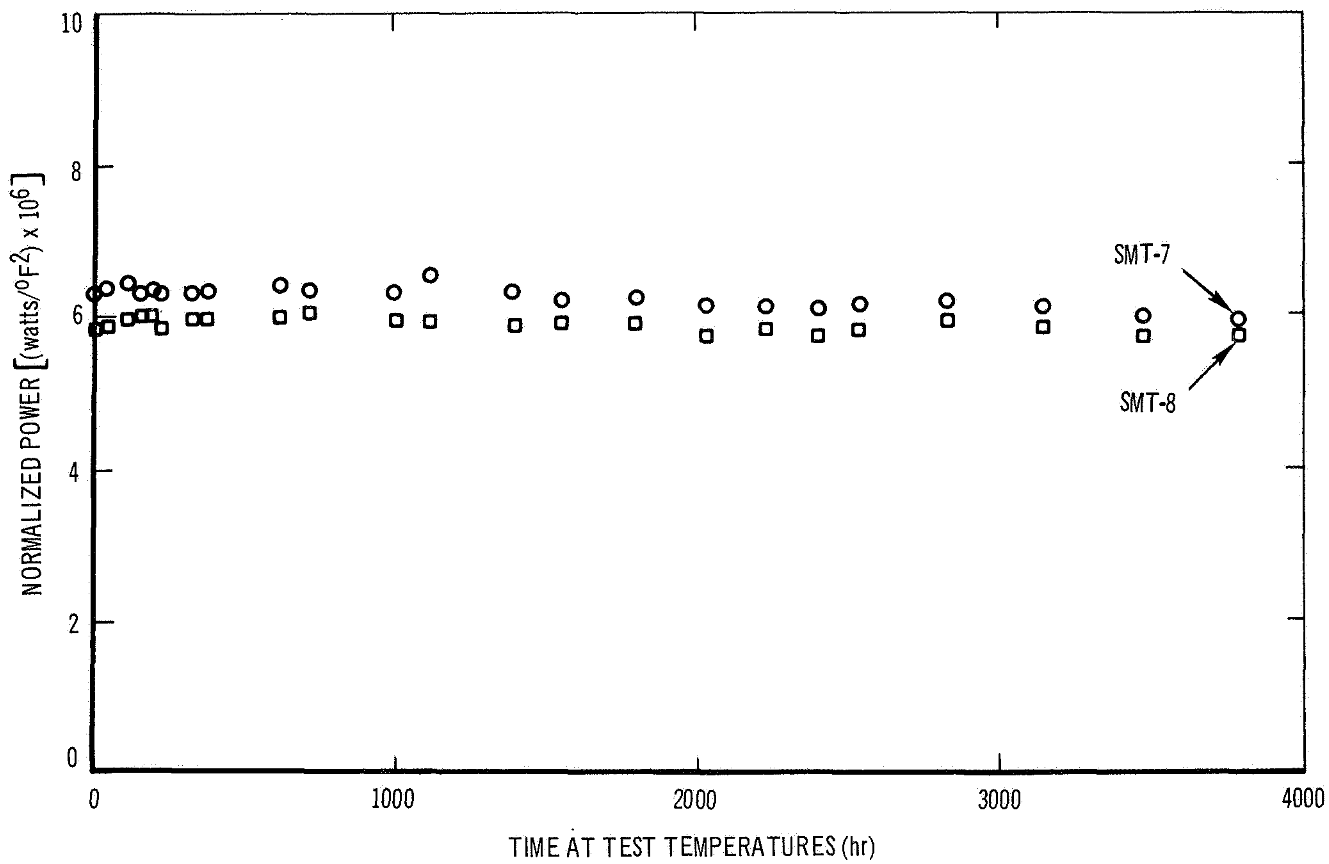

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Figure 11. Normalized Power vs Test Time for Modules SMT-7, -8

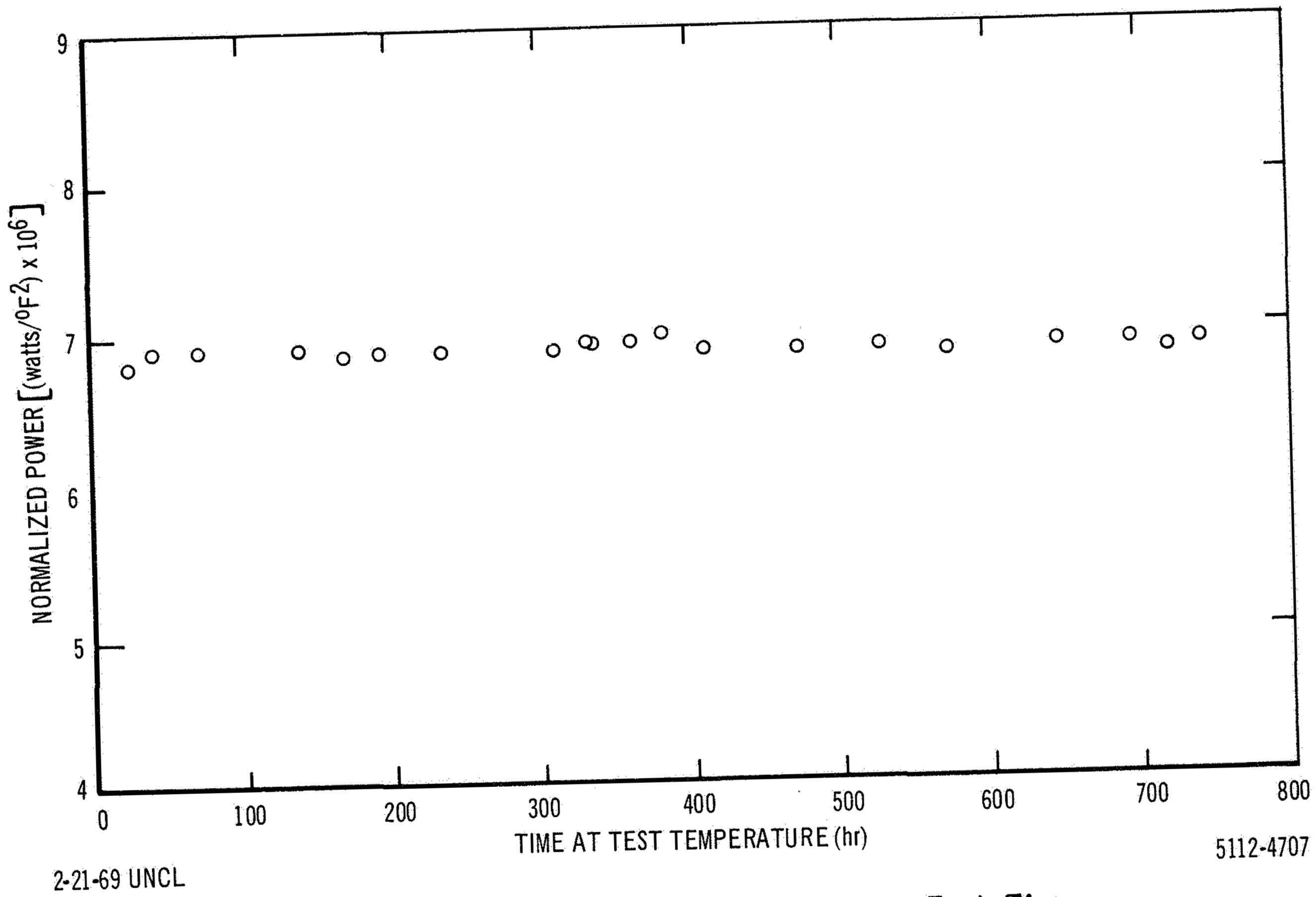


Figure 12. Average Normalized Power vs Test Time for SMT-9 and SMT-10

The significantly reduced initial degradation rate, at (T_H) max = 800°F, is easily evident by comparison of Figures 8 and 12. It is too early to evaluate the degree of improvement, but it must be at least equal to the 3X to 5X predicted.

7. LMT-1, 10-Watt Module

This 24-couple module, designed to be an efficiency demonstration, was submitted initially to a short series of parameter studies by varying heater input and, hence, the hot-junction temperature. Table 2 gives the most pertinent data from these excursions.

TABLE 2

LMT-1 INITIAL TEST DATA

Watts In	T_H, °F	T_C, °F	ΔT, °F	Watts Out	Efficiency,%
125	629	339	290	3.47	2.77
150	707	376	331	4.78	3.19
150*	640	312	328	4.76	3.17
160	674	321	353	5.58	3.49
170	703	331	372	6.31	3.71
175	719	334	385	6.83	3.905
186	747	343	404	7.44	4.00
190	776	353	423	8.06	4.24

*Cooling water in jacket turned on here

The efficiency points, when graphed as in Figure 13, show that this experiment if operated at an average ΔT of 450°F, would yield an overall efficiency of about 4.35%. It is emphasized that the quoted

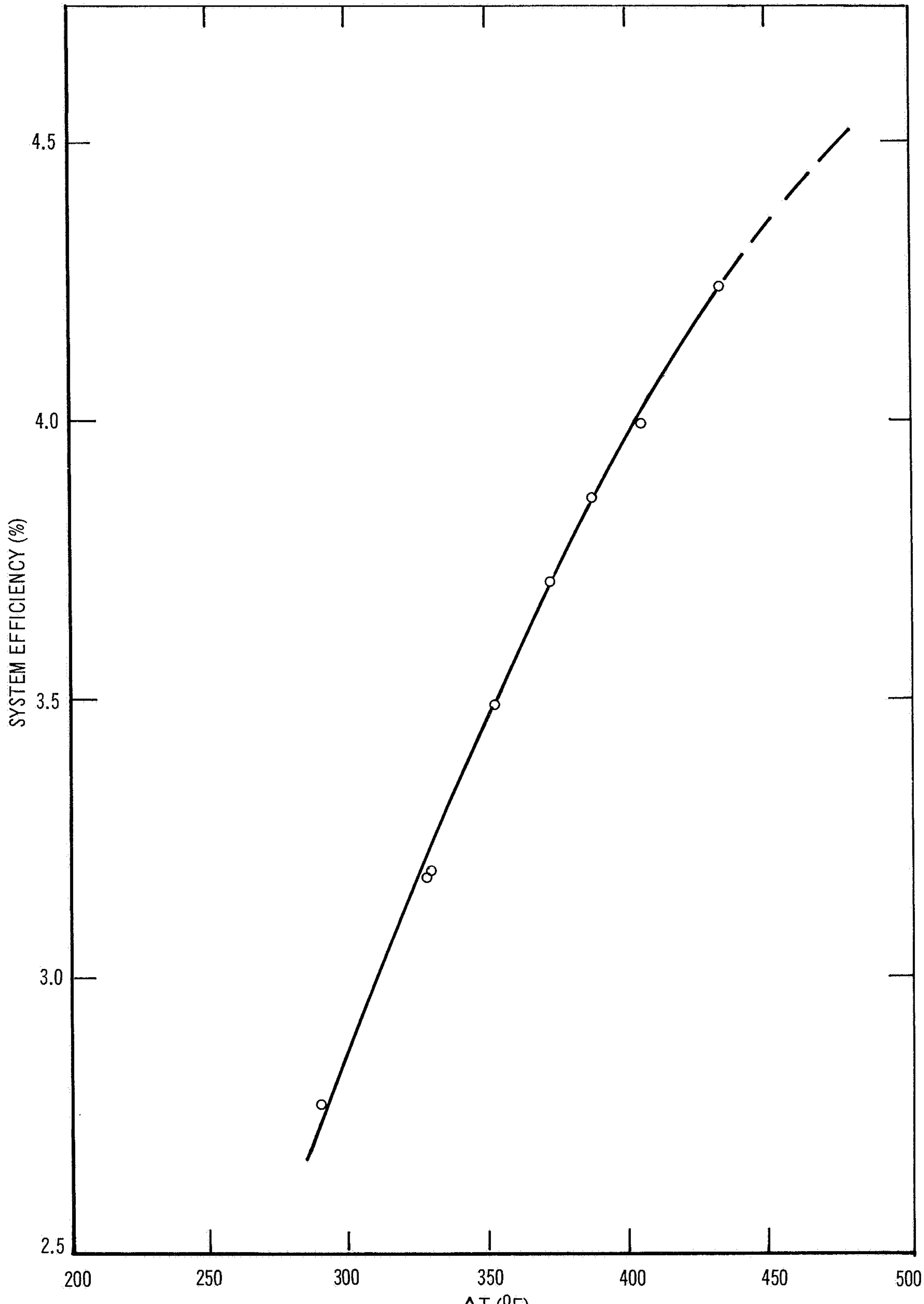


Figure 13. System Efficiency vs ΔT for LMT-1

efficiency is a "raw" efficiency number, i.e., obtained from the electrical output in watts divided by the heater input in watts, with no adjustment made for losses associated with the experimental setup. These numbers perhaps represent what could be achieved in a real system with an isotope heat source and its associated losses. If one allows that the heat source suspension and thermal insulation losses are in the range 10 to 20%, which is a typical figure, then the true module efficiency is in the range 4.8% to 5.2%. Some reasonable improvements in the couple thermal insulation, together with electrical optimization, should raise this number to the range 5.5% to 6.0%. The latter range is, then, a predicted engineering design quantity that can be achieved for this type of converter module in future systems.

The power output can be obtained from the normalized power, approximately 48.0×10^{-6} watts/°F^2, multiplied by the $(\Delta T)^2$. At 450°F ΔT, it would be 9.72 watts, or about 0.405 watt per couple. From an in-house program (STAR Panel), the measured weight for a technologically similar couple is 34.4 grams. including all support structure. From these quantities, a module specific power of 5.32 watts/lb is calculated. Again, optimization should increase this to 6 watts/lb or greater.

The module test was adjusted to a steady 170 watts input, after the initial excursions; this has produced an average hot junction temperature of about 700°F, and a maximum hot-junction temperature of about 725°F. This condition was selected to give long-term stability. The operational graph is shown in Figure 14 . The "raw" efficiency has remained approximately constant at 3.65% and the measured power output is about

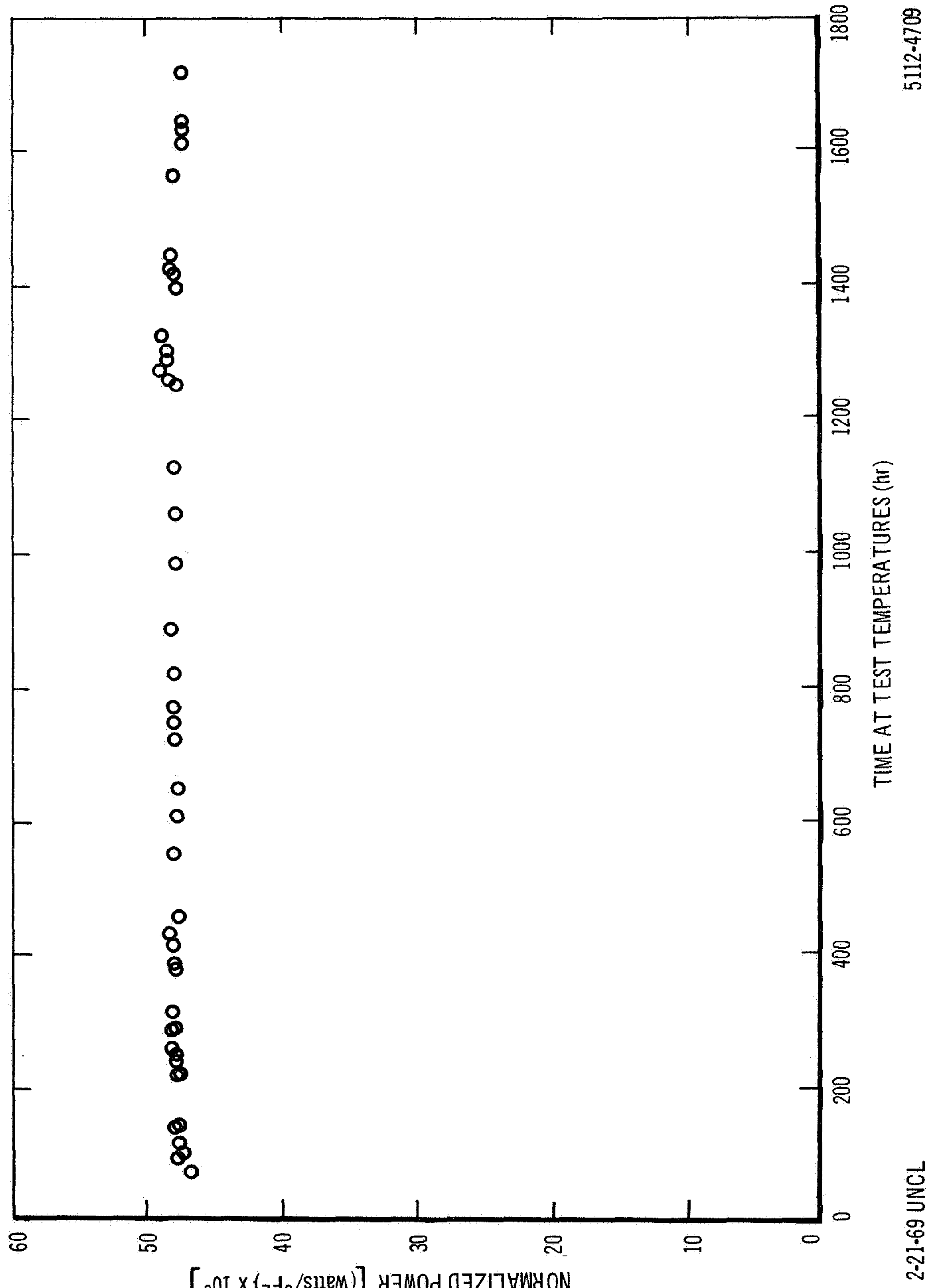


Figure 14. Normalized Power vs Time at Test Temperature for LMT-1

6-1/4 watts. Figure 15 shows the longitudinal temperature profile of each "side" of LMT-1. The temperature fall-off at the ends is related to the increasing view-angle of the heat sink, which includes the end surfaces of the test tank.

Figures 16 and 17 show, on greatly expanded scales, the basic electrical characteristics of Seebeck voltage and internal resistance, together with normalized power, efficiency, mean temperature, and temperature interval (ΔT), as they have varied with time. Aside from normal "reading" scatter, these plots follow consistent and expected trends; i.e., the Seebeck goes up when the mean temperature is raised, the efficiency follows the ΔT, etc. A degradation of about 1% at 1600 hours is apparent, but not well defined, in the normalized power. The rate should decrease with time, according to prior experimental data.

E. VIBRATION TEST

1. The Module

A standard SMT type 3-couple module was built and mounted on an extended hot wall piece. This permitted bolting to a commercial electrical strip-heater, and to ceramic stand-off insulators. The sides and ends of this assembly were thermally insulated to a reasonable extent, and a framework was added to support the current leads, voltage probes, and thermocouple instrumentation. The assembly was mounted on a heavy and rigid base which would be attached to the table of the vibration generator. Only one each hot and cold junction thermocouples were used. The module, at completion of all testing, is shown in Figure 18. It is noted that

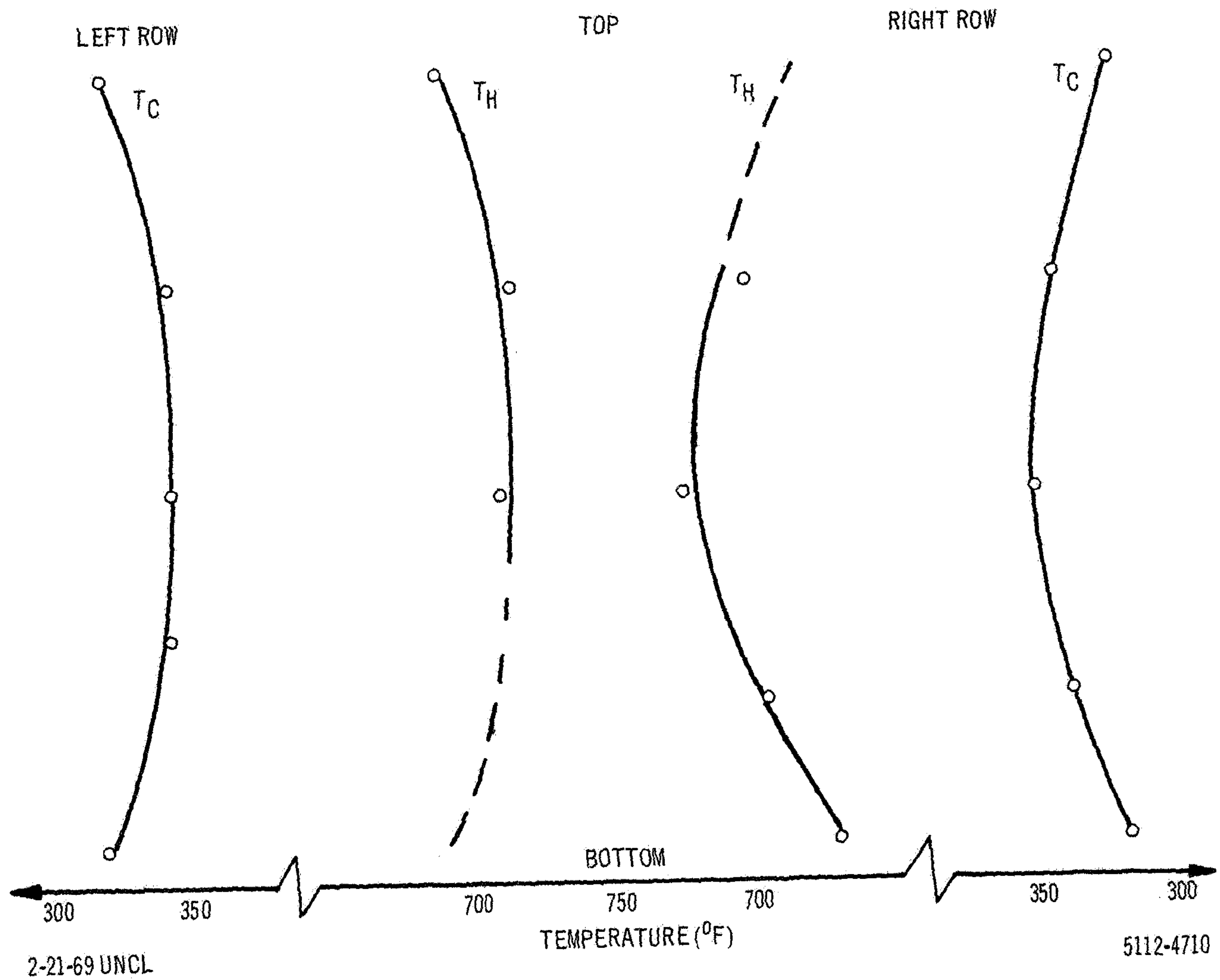


Figure 15. Thermal Profile along the Two Rows of Module LMT-1 after 1560 Hours of Testing

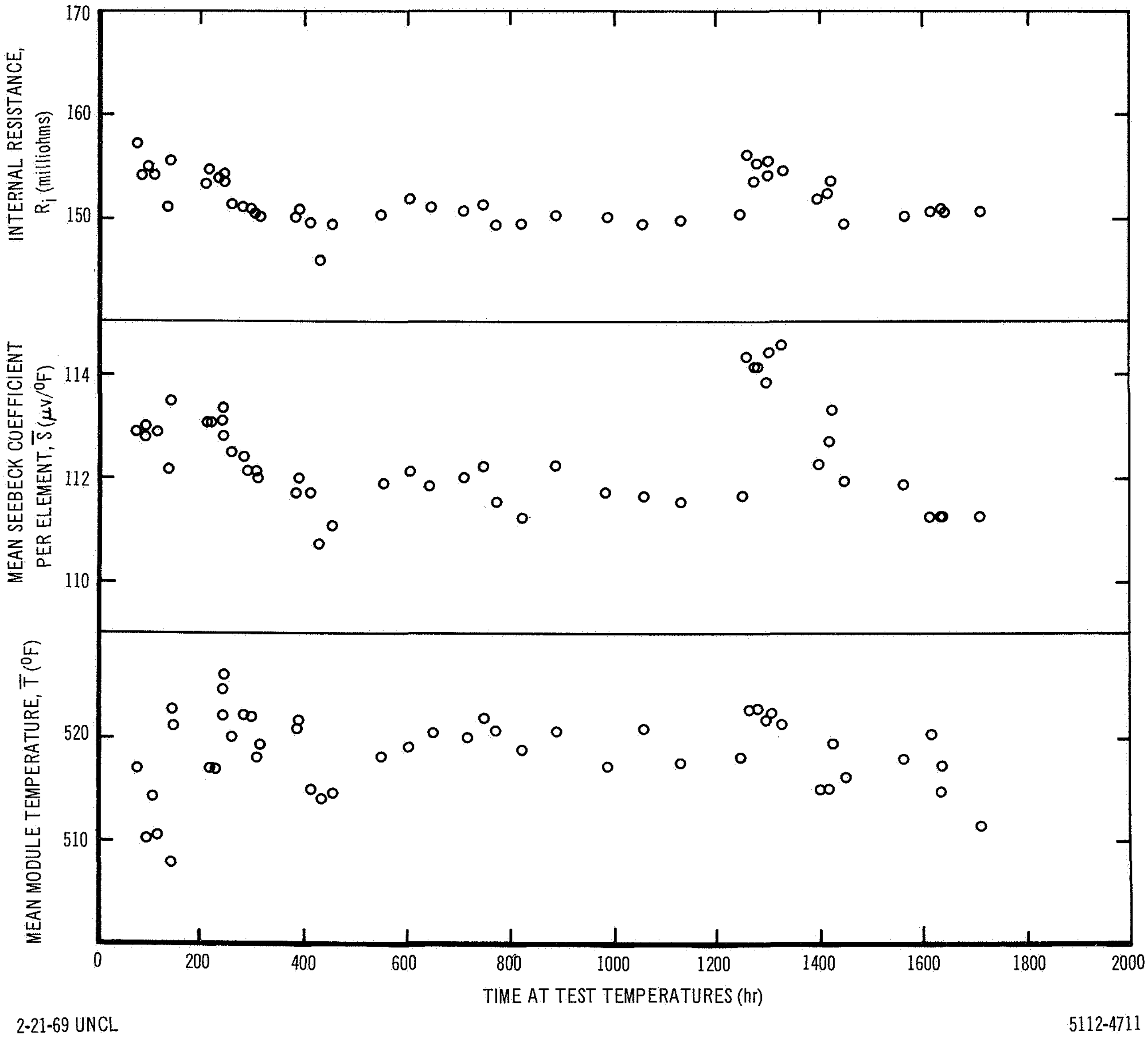


Figure 16. Internal Resistance, Mean Seebeck Coefficient, and Mean Temperature vs Test Time for LMT-1

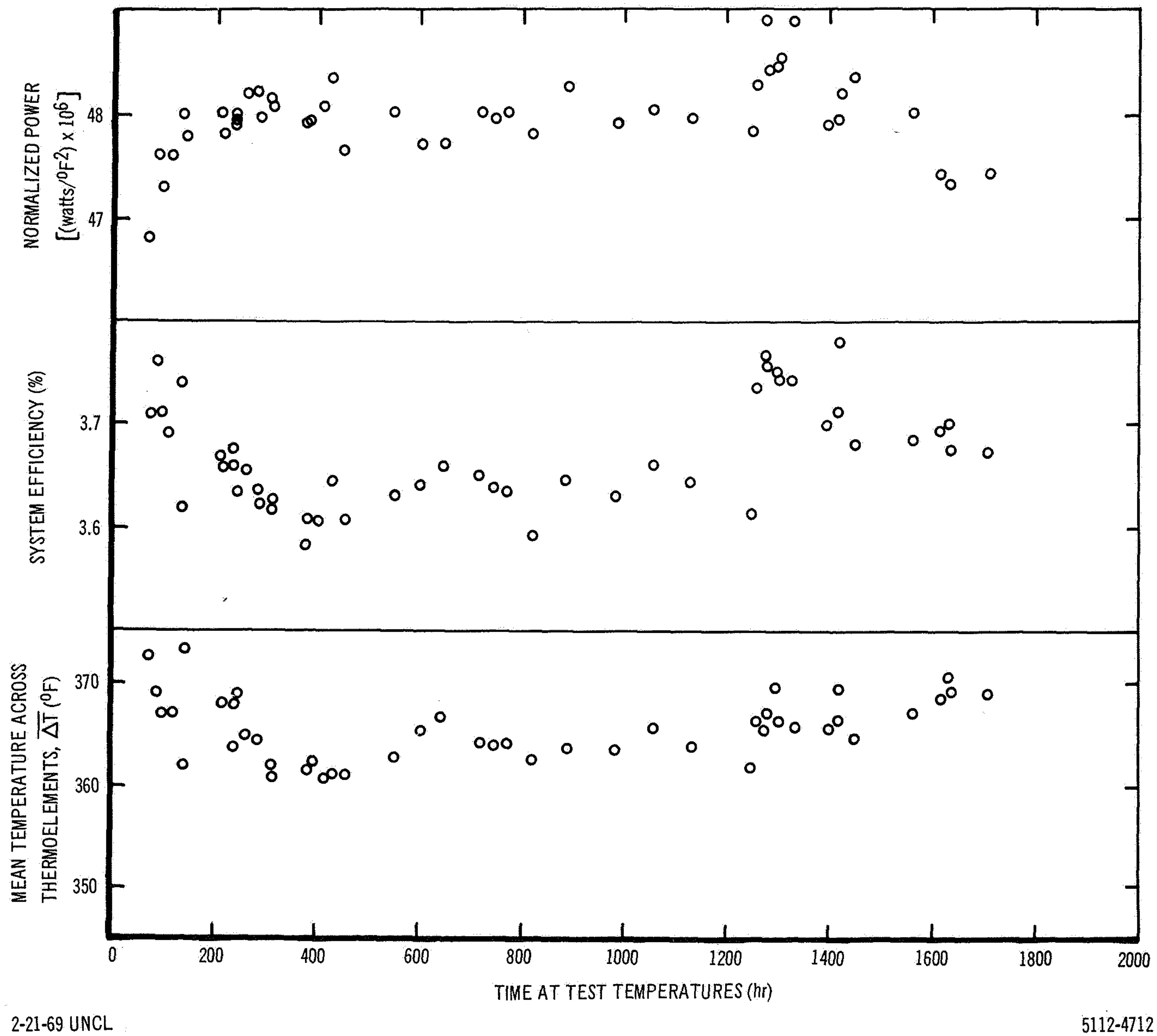


Figure 17. Normalized Power, Efficiency, and ΔT vs Test Time for LMT-1

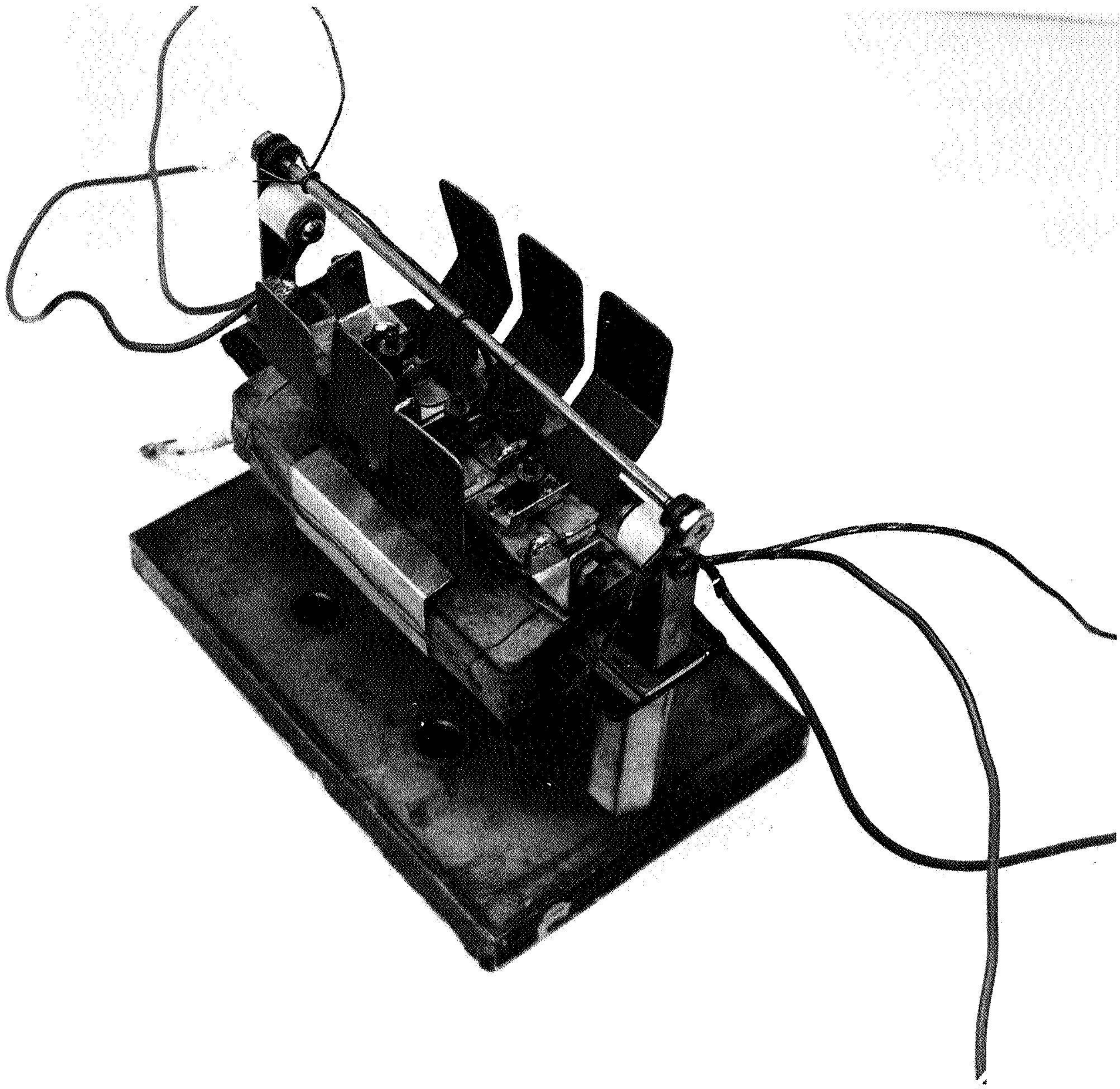


Figure 18. Vibration Test Module after Completion of all Tests

almost no permanent deformation (bending) of the fins occurred.

2. Test Procedure

The following procedure was outlined and applied:

Sequence A - bench testing

(a) Measure room-temperature resistance

(b) Outgas and operate in vacuum test station - determine Voc, R, and normalized power at $T_H \simeq 720°F$

(c) Operate in flowing argon - determine same quantities at $T_H \simeq 650°F$.

Sequence B - vibration testing

(a) Ship to Field Laboratory

(b) Vibrate at room-temperature, 3 axes

(c) Vibrate at $T_H \simeq 650°F$ in flowing argon, 3 axes

(d) Ship to headquarters laboratory

Sequence C - bench testing

(a) Measure room-temperature resistance

(b) Operate in vacuum and measure at $T_H \simeq 750°F$

The rationale for the above is based on the intention to simulate reality insofar as might be possible in testing a 3-couple experimental model.

Sequence A represents initial check-out and acceptance testing of a converter; the testing in argon recognizes that a converter intended for use in vacuum must be muffled with inert gas during pre-launch testing, and again after fueling.

Sequence B represents the mechanical shock loading of transportation, as well as launch-induced vibrations.

Sequence C represents operation finally in high vacuum, and checks total change effected by the prior experiences.

Muffling with argon was accomplished very simply by enclosing the test assembly in a round stainless steel can, over the top of which a plastic bag was attached by an elastic band. Flowing argon at about 2 psig inflated the bag; leakage was provided by a pin-hole at the end of the bag, and by penetrations made in the can to pass instrumentation leads and mounting bolts. This procedure probably did not exclude all air, but apparently was quite adequate. The flow rate was approximately 50 cfh. The hot-junction temperature of the module was approximately 70°F lower in the flowing argon than in high vacuum, for the same heater input.

During vibration testing, the cold-junction thermocouple was lost on the fourth shake, and hot-junction thermocouple on the sixth shake. However, the cold-junction thermocouple could be, and was repaired by splicing. By measurement of the open-circuit voltage, which amounts to using the 3 couple-module as a thermocouple, the ΔT could still be accurately determined.

The vibration test procedure used was as follows:

1. Attach test assembly to table of shaker using four 3/8-inch bolts.
2. Attach accelerometer to table adjacent to test assembly.
3. Adjust input to give 1/2-inch double-amplitude displacement at 9 Hz.
4. Sweep from 5 to 9 Hz at 1 octave/min.
5. Sweep from 9 to 400 Hz at 2.3g, 1 octave/min.
6. Sweep from 400 to 2000 Hz at 7.5g, 1 octave/min.

This schedule is approximately the same as used for the SNAP-10A converter qualification test.

TABLE 3

VIBRATION TEST DATA

Test Conditions	T_H, °F	T_C, °F	ΔT, °F	Voc, mv	R* mΩ	Normalized Power watts/°F^{2}x10^{-6}
Initial, room temp.	75	75			6.60 (5.00)	
Initial, hot, in vacuum	712	375	337	220	20.3	5.25
Initial, hot, in argon	651	336	315	200	17.8	5.66
After shipment, room temp.	80	80			7.76	
After shake, axis A	80	80			7.73	
After shake, axis B	80	80			7.65	
After shake, axis C	80	80			7.71	
Hot shake, axis C	635				18.4	
Hot shake, axis B	648				18.4	
Hot shake, axis A	650				18.45	
Before shipment, room temp.	83	83			7.40	
After shipment, room temp.	80	80			8.17 (5.30)	
Final, hot, in vacuum	722	376	346	235	22.6	5.10 (5.25) **
Final, room temp.	76	76			7.38 (5.30)	

*Measured at bolt-fastened terminals. Figures in parentheses are measured at current lead tabs.

**Adjusted for resistance increase at bolted terminals.

For the "heated" shakes, the electrical heater was turned on approximately one hour prior to step 3 above, and the argon purge was started at the same time. For all the "shake" operations, the indication of damage was to be change in resistance, measured at the terminals. Since there was a bolted connection at each terminal, this invited a resistance increase extraneous to the module, which obviously occurred. The total resistance was seen not only to increase, but also sometimes to decrease again. The entry in Table 3, "R" column, third from the end, is noted to be 8.17 mΩ, verifying the source of variability.

3. Test Results

Table 3 summarizes all the pertinent data from the test schedule. It is noted that the module room temperature resistance, after all tests, had increased from 5.00 to 5.30 mΩ, or 6%. This increment, however, would be only 1.5% of the hot resistance. The calculationally adjusted final normalized power, marked by **, indicates essentially no change. It is concluded that the total effect of all handling, transportation, and vibration is in the range 0 to 2%. Since the measurement accuracy is of the order of 1%, the change can be stated as 1% ± 1%.

It is commented that attachment of the controlling accelerometer to the table of the shaker, rather than to the module, allowed high amplitude reasonances to occur in the module. These were indeed observed both audibly and visually by "flutter" of the module fins. Therefore, it is reasonably assumed that accelerations much greater than 7.5g were experienced by the test module. Again, this simulates a real situation.

It is concluded that the vibration test was realistic, and that it resulted in no significant damage.

F. POST-TEST EXAMINATION OF COUPLES

Thermocouples from modules SMT-3, -4, -5, -6 were mounted for polishing and metallographic examination. The final polished surface on each thermoelement was a plane containing the axis of the thermoelement. Hot side contacts on the P legs of the center thermocouples of SMT-3 and -4 had failed mechanically when the couples were removed from the modules. In this case, the contacts were held in place against the thermoelement for mounting. Each mounted and polished thermoelement was visually searched with an optical microscope for unusual features. Particular attention was given to the thermoelement contacts which failed mechanically, since the P legs of these thermocouples had shown about a 120% resistance increase during test. Because of the hot side temperature and length of test on SMT-3 and -4, the hot contact regions of thermocouples from these modules were examined with an electron microprobe for possible interdiffusion of thermoelement material and aluminum contact. The results of these investigations are discussed below.

Gross visual examination of the polished thermoelement sections with an optical microscope showed the difference normally noted between hot-pressed 2N or 3N and 2P thermoelements. Generally, the N elements are relatively free of flaws, while the 2P elements are more porous and show some crack development. The cracks appear mainly on the periphery of the thermoelement at its intersection with the contact. A typical example of this feature is shown in Figure 19. Such cracks are suspected to arise during the original processing of the element, and may be broadened by the stresses produced in the element during mounting and polishing.

All thermoelements showed slightly the effect of evaporation at the hot-side contact. Those operating for longer times showed correspondingly more material removal. An example of this evaporation is shown in Figure 20, an N type thermoelement operated at about 750°F for more than 11,000 hours. It will be noted that the material evaporated from the element condenses on the cooler insulation surrounding the thermoelement. The reduction in contact area of the element at the hot contact due to evaporation is only a few percent at most, and would not account for any significant portion of the observed resistance increases.

A representative contact region between the PbTe thermoelements and aluminum contact is shown at two magnifications in Figures 21a and 21b. The particles discernible at the interface are tungsten. Because tungsten particles are pulled out during the polishing operation, what appears to be voids at the interface are merely positions from which tungsten particles have been removed. The contact region shown here is a P element cold-side contact and, because of its low operating temperature, it should represent to a good approximation, the bond as fabricated.

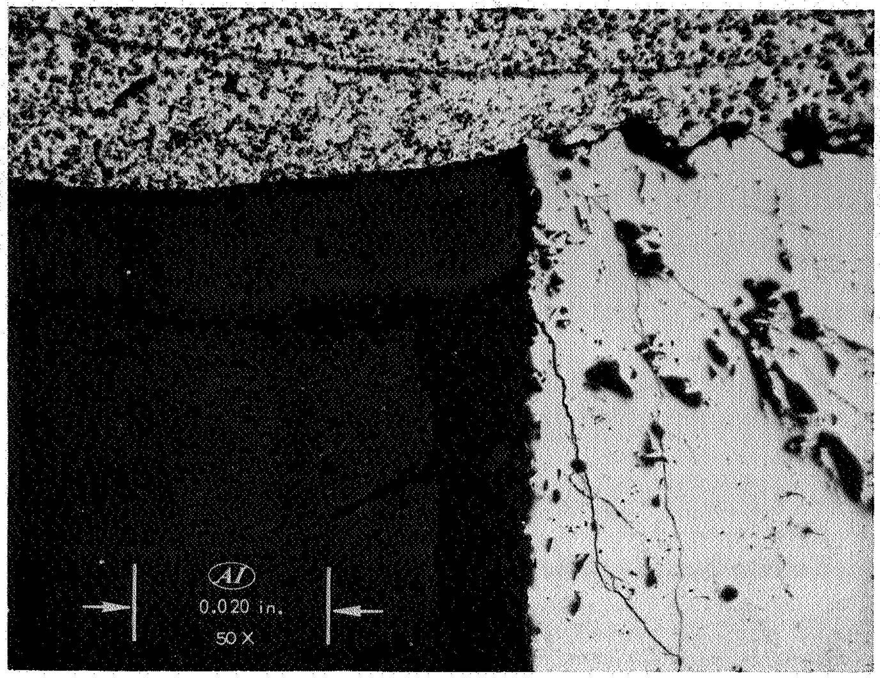


Figure 19. Crack Development at Intersection of 2P Thermoelement and Aluminum

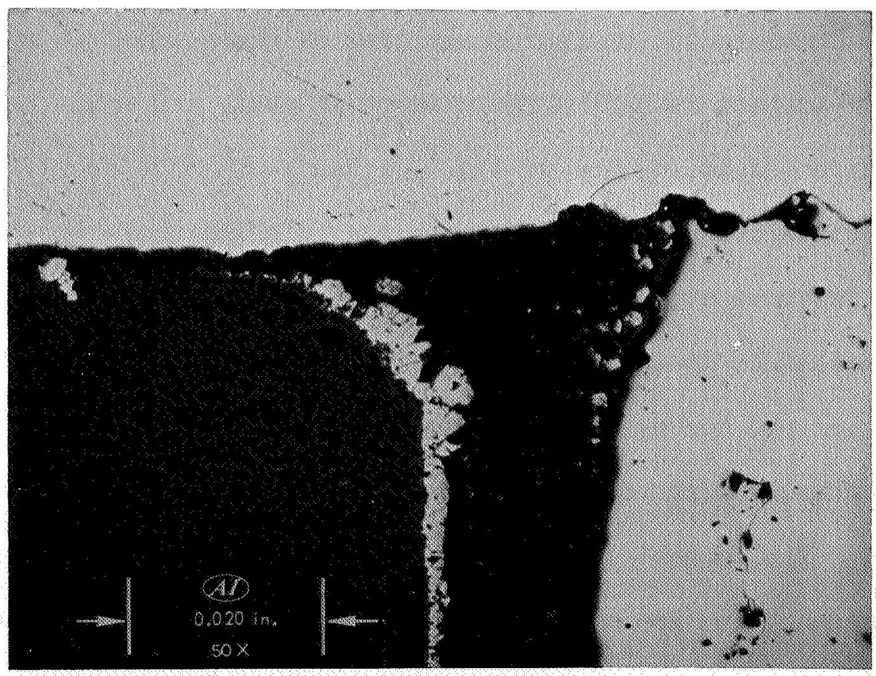
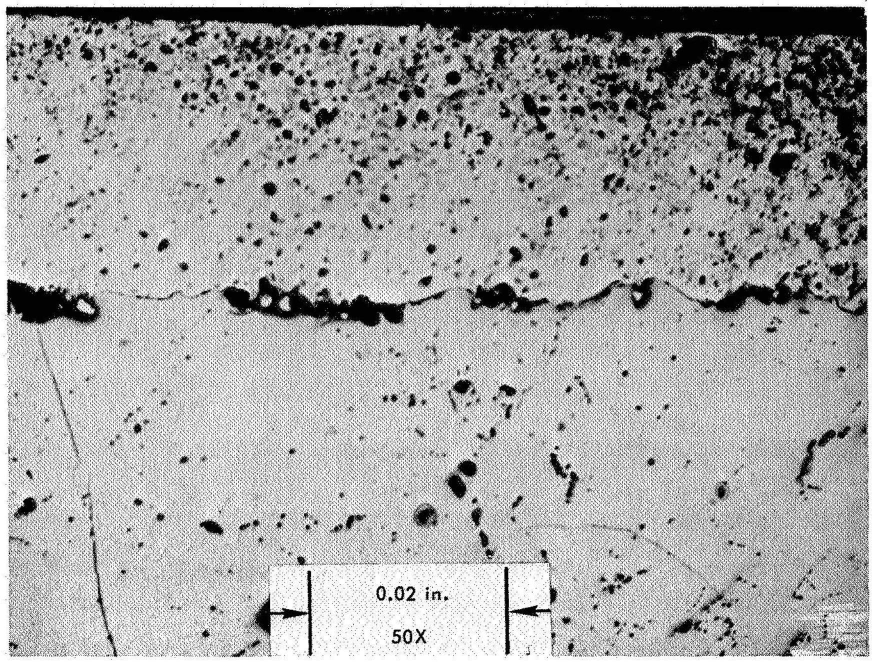


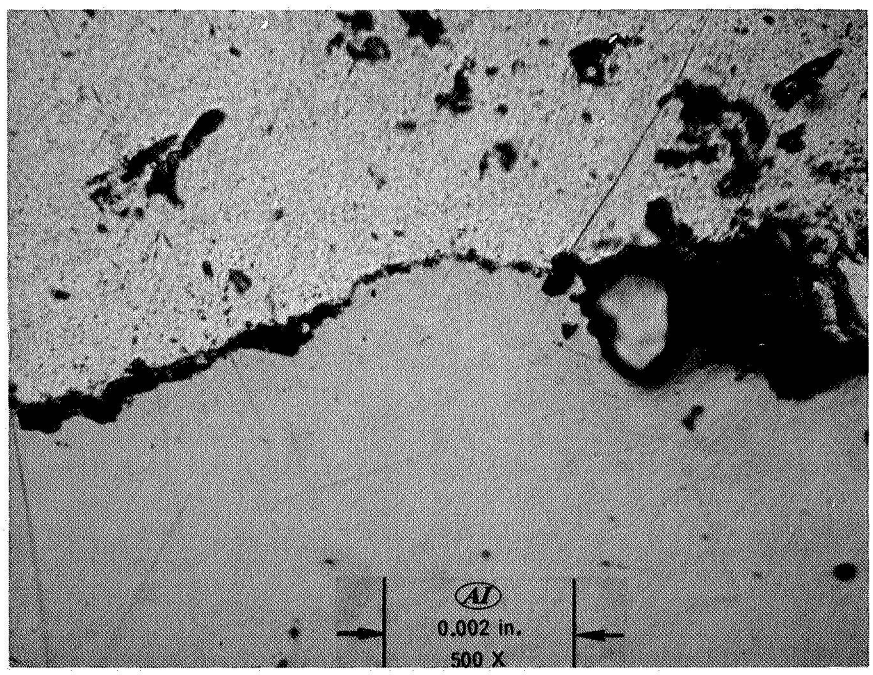
Figure 20. Thermoelement Evaporation at Hot Junction and Condensation on Insulation



Al SHOE

2P
PbTe

a.



Al SHOE

2P-
PbTe

b.

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Figure 21. Bond Region at Al-PbTe Contact

Al SHOE

2P
PbTe

Al SHOE

2P
PbTe

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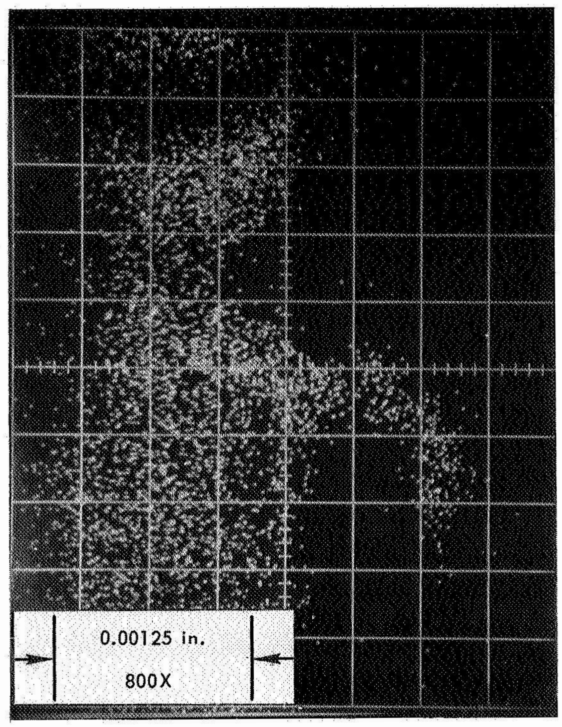
Figure 22. Failed Hot Shoe Bond Operated 11,600 Hours at 800°F (2P)

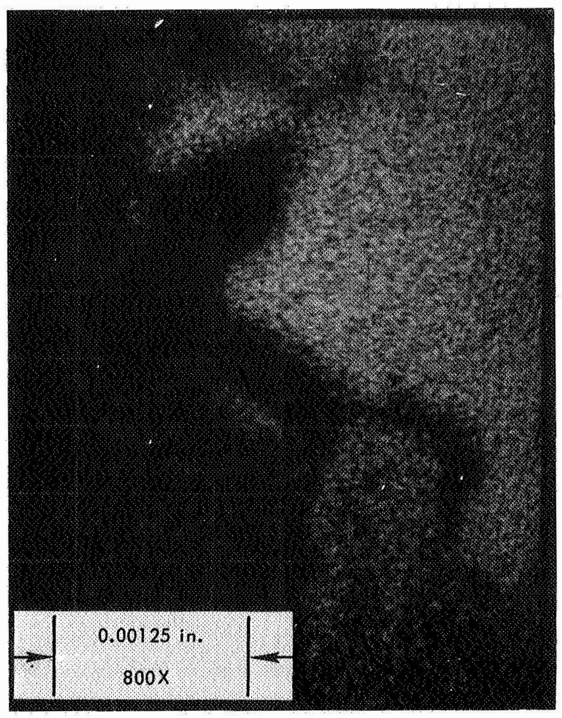
The structure at a hot-side contact which failed when the couple was being removed from the module is shown in Figure 22. This interface operated at about 800°F for more than 11,000 hours (SMT-3). Although this (separated) bond region was in poor condition for metallographic examination, it is observed that the tungsten particles in the center of the photomicrograph have material clinging to them which is made up mostly of another solid phase. This phase could not be analyzed, but, based on past experience, it is suspected to be aluminum telluride. Aluminum telluride has a high electrical resistivity and is very unstable in air, transforming to aluminum oxide or hydroxide and volatile hydrogen telluride, in the presence of water vapor. Therefore, direct verification of its presence is difficult. The hot-side contact region of thermocouples from the same module (SMT-3), operating at 750°F and lower temperatures, did not show the presence of this new phase.

In an attempt to verify the interaction of aluminum and tellurium, the bond regions of thermoelements were examined using an electron beam microprobe. In this analysis method, the bond region under study is scanned with an electron beam. Detectors are used to pick up back-scattered electrons and X-rays characteristic of the chemical elements present in the surface of the area being scanned. The detector signal is used to modulate the intensity of an oscilloscope beam which is scanning the oscilloscope screen in synchronization with the probe beam over the sample scan area. Photographs of the oscilloscope trace are used for evaluation. White areas on X-ray scans represent regions where a given chemical element is present, while white areas on back-scattered electron scans represent areas where heavy elements such as Pb and W are present.



b. Te X-RAYS

a. BACK SCATTERED ELECTRONS



c. Al X-RAYS

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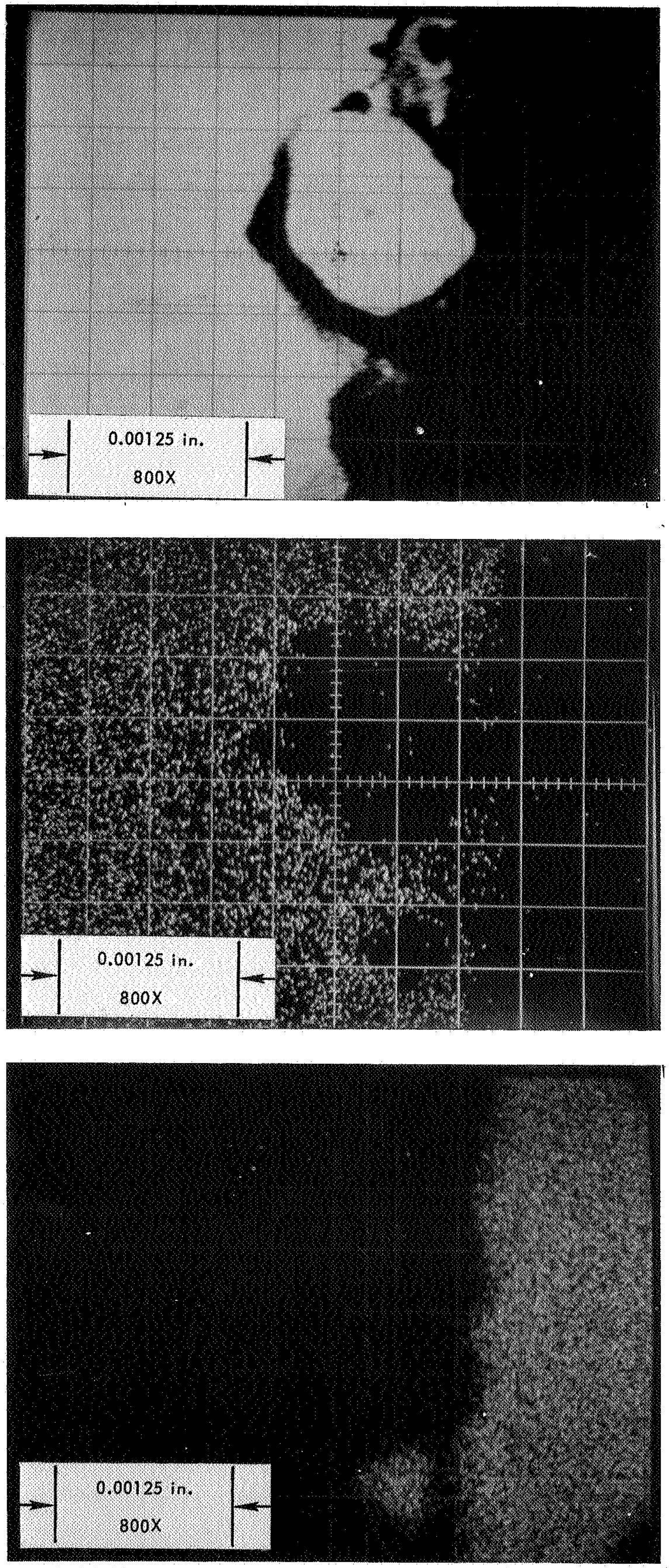
Figure 23. Electron Microprobe Scans at Region of Al-PbTe Bond

Figures 23, 24, and 25 present the results of two area scans at the hot bond region of the center couple N-thermoelement of SMT-3, and a single area scan at the hot bond region of an outside couple N-thermoelement of SMT-4, respectively. These scans are representative of those taken in bond regions which operated under similar thermal environments.

A comparison of Figures 23b and 23c (800°F, 3N) suggests that interpenetration of Al and Te has occurred, while a similar comparison for Figures 24b and 24c, another section of the same bond, throws doubt on this conclusion. Comparison of the Te and Al scans of Figure 25 (750°F, 3N) suggests that little, if any, interdiffusion of Al and Te occurred. In view of what has been noted about the instability of aluminum telluride, it is not surprising that these scans do not always show the expected interpenetration and compound formation between Al and Te. The microprobe electron beam penetrates into the surface to only about one micron, a depth to which aluminum telluride could easily react with atmospheric water vapor.

The conclusion to be drawn from this analysis is that aluminum-contacted PbTe operates with reasonable stability at 750°F, while operation at 800°F results in serious bond deterioration. The mode of deterioration at the higher operating temperatures is believed to be the formation of aluminum telluride, which causes the observed resistance increase in the thermocouple. It would also account for a reduction in the strength of the contact due to the brittleness of the aluminum-tellurium compound, and perhaps a tendency of the compound growth process to disrupt existing mechanical bonds.



a. BACK SCATTERED ELECTRONS

b. Te X-RAYS

c. Al X-RAYS

Figure 24. Electron Microprobe Scans of Another Position of Bond of Figure 23

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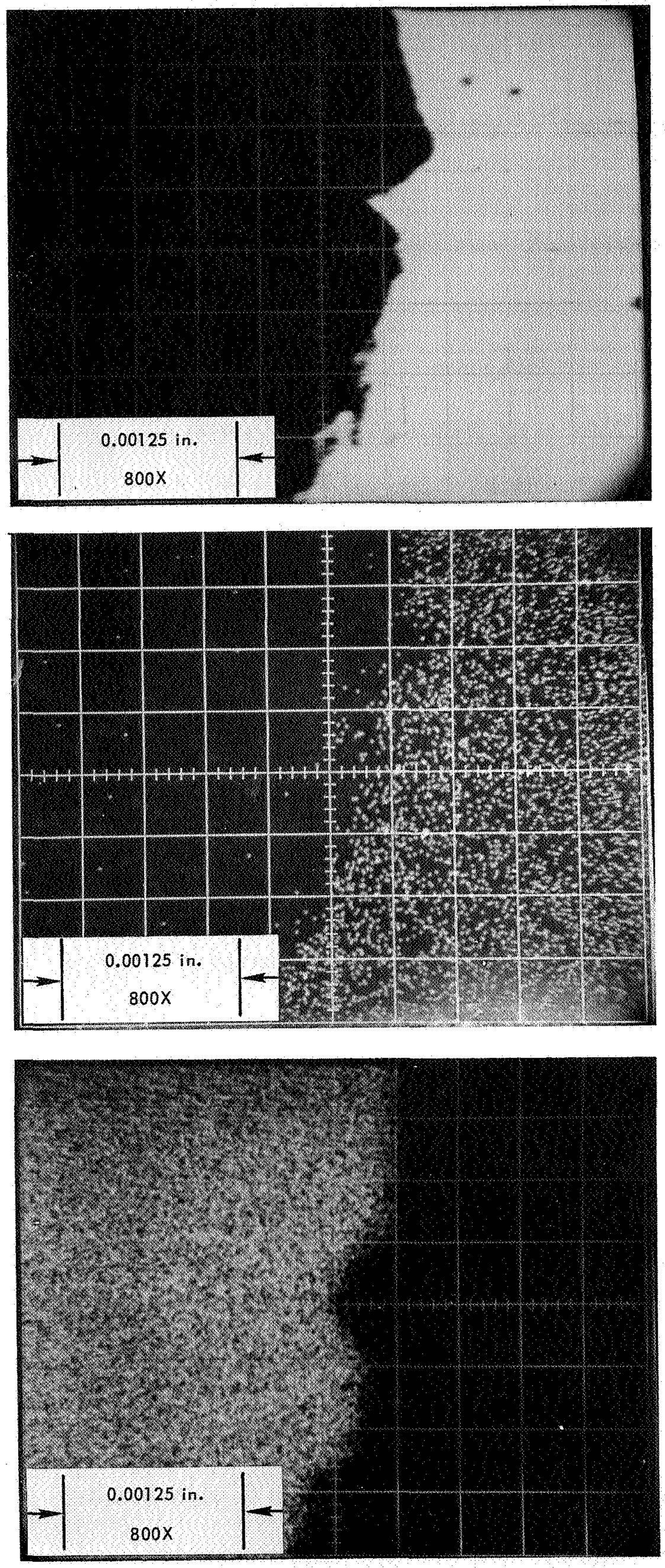

a. BACK SCATTERED ELECTRONS

b. Te X-RAYS

c. Al X-RAYS

Figure 25. Microprobe Scans of Hot Side Contact Between Al and PbTe for N-Thermoelement from SMT-4 Operated at 750°F for 11,600 Hours

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G. EVALUATION OF TOTAL EXPERIMENTAL FINDINGS TO DATE

1. Power Output

A review of all of the operational data reveals that the initial normalized power, $P_{max}/(\Delta T)^2$, is consistently in the range 2.0×10^{-6} to 2.4×10^{-6} watt/°F^2 per couple for the standard 0.257-inch-diameter by 0.200-inch-long elements. The values vary somewhat with T_H and ΔT. In a prior report (AI-67-132), power calculated from vendor date, for uncontacted elements, is given as .419 watt/couple at 427°F; this corresponds with 2.30×10^{-6} watt/°F^2 per couple. It is apparent therefrom that the total parasitic electrical loss of contacts and straps is at most very small.

2. Stability

It becomes increasingly clear that the standard aluminum contact, though initially very efficient, is suitable for 3-5 year operation only at hot-junction temperatures below 750°F maximum. In a practical, large converter, with reasonable assumptions as to temperature distribution, the 750°F limit might imply an average hot-junction temperature of 730°F to 740°F. However, if the early good behavior of SMT-9 and -10 continues, an increase of at least 50°F in future modules can be contemplated. The average T_H of SMT-9 is, in fact, approximately 785°F, and its two hottest couples are being kept at T_H = 800°F average.

From knowledge of the nature of the AI tungsten hot-junction contact used with 2P material, and various in-house experiments therewith, it is expected that good long-term stability will be achieved in a high-vacuum environment, even at temperatures higher than 800°F. The dominant degradation process will probably be evaporation of the thermoelectric material or its components. Extension to higher temperatures may therefore require a coating or other mechanism to retard evaporation. This has not been within the scope of the present program.

3. Efficiency

As shown in II.D.7. the efficiency that can be obtained is limited by the ΔT applied to the couples. It is further subject to thermal insulation and electrical-optimization efficiencies; which also determine, in part, the heat flux to be rejected and, hence, the cold-junction temperature. The prediction of efficiency in a space-radiating system requires, therefore, that a realistic estimate be made of whether or not the desired ΔT can be obtained. An iterative calculation is required. For example, if one assumes that, as in II.D.7., the converter efficiency will be 6%, at a ΔT of 450°F, the following applies:

1. The couple power will be 2.10×10^{-6} watt/°F^2 x(450°F)^2 = .425 watt.

2. The reject heat per couple will be .425 watt/.06 - .425 watt = 6.68 watts.

3. If the cold-fin area = 7.00 in.^{2}, the reject heat flux will be $\frac{6.68}{7.00}$ = 0.954 watts/in.^{2}.

4. If the overall fin efficiency (including emissivity and temperature distribution) is 80%, the cold-junction temperature in deep space will be about 305°F.

5. The hot-junction temperature will be 305°F + 450°F = 755°F.

A similar analysis based on the assumption of 5% converter efficiency yielded $T_C = 340°F$, $T_H = 790°F$. It is apparent that the ΔT of 450°F, or greater, can be achieved with a couple fin area of 7 in.^{2}; and that the ΔT consideration will not be limiting in the efficiency range above 5%.

H. CONCLUSIONS

1. The standard Al-PbTe electrical contact is sufficiently stable for long-term use (3-5 years) at temperatures below 750°F. The life-limiting factor is confirmed to be interaction between Al and Te at the P-element hot junction.

2. A modification appears to have been found a (a tungsten-faced barrier) which can greatly retard the degradation process, and may permit raising the hot-junction temperature limit to at least 800°F.

3. Initial power output and efficiency data are excellent.

4. A successful vibration test has shown that the basic design, with tension-stud mounting, is structurally sound and resistant to anticipated mechanical stresses in space system use.

5. The experiments and tests to date show that space thermoelectric converters can be engineered, using the subject technology, which will be made wholly of magnetically neutral materials, be mechanically rugged, provide excellent power/weight and power/area ratios, yield conversion efficiencies of 5% or higher, and operate with sufficient stability for 3 to 5 year missions.

6. The stability claimed above is based on less than 2 years' testing and is not felt to be completely substantiated, especially with regard to the latest (P-element hot-junction) technology. Therefore, considerably longer life-testing is required.