AI-67-132

SUMMARY REPORT
FOR
ALUMINUM BONDED LEAD TELLURIDE THERMOELECTRIC MODULE RESEARCH AND DEVELOPMENT

(1 May 1966 – June 1967)

Contract No. NAS5-10184

Prepared by
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SUMMARY

The objective of this program has been to investigate experimentally the Atomics International process for contacting PbTe couples with directly bonded aluminum straps, and to demonstrate the potential of this method for the construction of non-magnetic thermoelectric conversion modules suitable for long-term operation in space applications.

All measurements and test results on contacted single elements, couples, and 3-couple modules have demonstrated excellent initial power density (0.40 watt/couple) and efficiency (calculated 5.7%) for the temperature interval 750°F to 325°F. The test modules were constructed wholly of non-magnetic materials.

Life-test data, up to 3000 hours, show very promising stability, with power degradation rates ranging from zero at T_H = 700°F to 3.2%/1000 hrs. at T_H = 800°F. It is expected (from data available from other programs) that the degradation rate will decrease significantly with longer operation.

It is concluded that this 14-month program has demonstrated high potential for aluminum-bonded PbTe modules in space environment. It is recommended that life-testing be carried on for 2 to 5 years, to better determine stability; also, that additional experiments be performed to obtain improved data on contact resistance, efficiency, and mechanical integrity.

I. INTRODUCTION

This program, begun at Atomics International in May, 1966, was based on a proprietary process for contacting PbTe thermoelectric couples with aluminum, using particulate tungsten at the contact interface to penetrate the oxide normally present on the aluminum. The contacting process has not been further developed on this program.

The purpose of the program has been primarily to demonstrate that the reference process can be used to fabricate couples and multi-couple modules, using all non-magnetic components, which will give moderately high efficiency and power density, and very high stability in long term operation. A useful operational life of 5 years is desired; therefore, a reasonable goal is a degradation rate of a small fraction of 1% per 1000 hours. The program plan has been to determine the initial characteristics of aluminum-contacted N and P elements, and then to life-test small modules (of 3 couples each) in simulated space vacuum at hot-junction temperatures in the range of 700°F to 800°F. It had been proposed that the desired degree of stability could be achieved at a temperature in this range. Determination of the maximum practical operating temperature has been a prime objective. Three preceding reports, AI-66-158, AI-66-260, and AI-67-99, summarized progress of the first three quarters of the program. In February, 1967, it was apparent that the test modules were more stable on life-test than anticipated, so that tests much longer than originally proposed would be required in order to determine degradation rates. By mutual agreement, the last quarter was extended by two months; this allowed an additional 1500 hours of testing and the initiation of two additional life-tests.

This report summarizes a 14-month program, and includes also some life-test data taken during the writing period. Some of the more important technical data and conclusions previously reported in the quarterlies are reiterated for the sake of continuity.

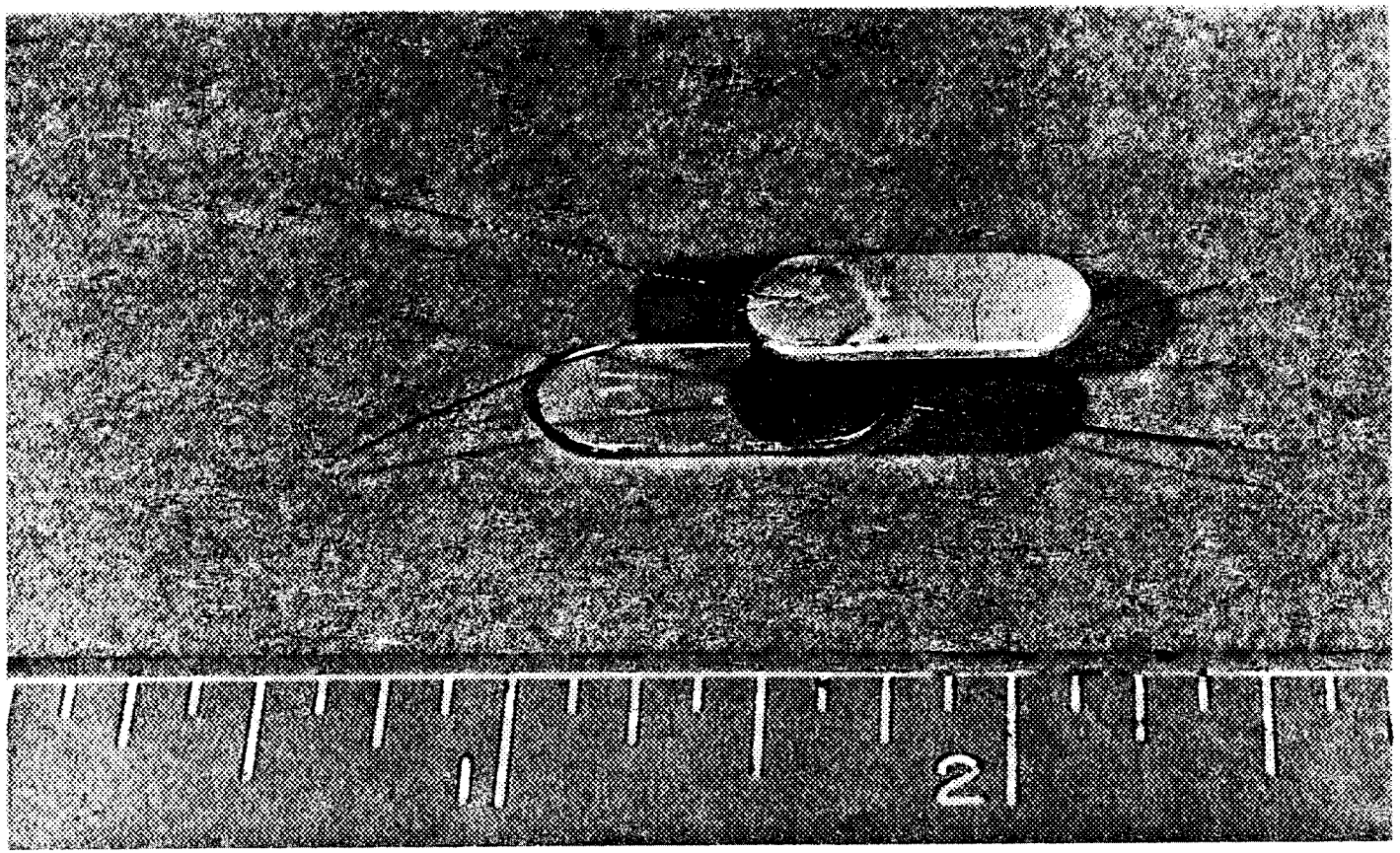

FIGURE 1. STANDARD TEST ELEMENT

II. DISCUSSION

A. BASIC ELEMENT DATA

1. Standard Test Element

The program scope did not include determination of contact resistance. Moreover, prior in-house work had shown that the aluminum PbTe contact resistivity was so small as to be very difficult to measure. Therefore, it was decided to make all basic measurements on a standard contacted test element. One such is shown in Figure 1, and its dimensions are listed in Table 1. All subsequent couples and modules in this program use the same element dimensions.

TABLE 1

STANDARD ELEMENT NOMINAL DIMENSIONS

Element Diameter Before Contacting	0.257 in.
Length before contacting	0.208 in.
Diameter after contacting	0.2575 in.
Calculated ratio, length/area	1.59 cm^{-1}
Length with straps	0.258 in.
Strap Length	3/4 in.
Width	0.312 in.
Thickness before contacting	0.030 in.
Reduction in thickness at element location	0.005 in.
Overall Length, Assembled	1-3/8 in.

The test elements have chromel-alumel thermocouples embedded in each aluminum contact so as to give as exact as possible measurements of junction temperatures, and accurate measurements of Seebeck voltage and resistance at temperature (by ac voltage drop method).

2. Seebeck and Resistance Measurement

A special but simple apparatus was built to accept the standard test element, and elaborately instrumented for accurate and consistent simultaneous measurement of junction temperatures, Seebeck voltage, and in-gradient resistance. This equipment and its use are fully described in AI-66-150. Accuracy and precision are of the order of ±2%.

3. Thermal Soak Test and Data

Six 2N elements and five 2P elements, all aluminum contacted, were measured as above both before and after an isothermal soak at 800°F for 500 hours, in a vacuum of about 10^{-5} torr. Table 2 lists element power data, calculated from the Seebeck voltage and resistance at temperature, for these elements before and after thermal soak. The power capability of the contacted elements, with no correction for contact losses, is compared with power calculated for bare elements, using published vendor data.

TABLE 2

STANDARD TEST ELEMENT POWER DATA, BEFORE AND AFTER THERMAL SOAK

Calculated From	Lower Temperature Interval	Higher Temperature Interval
2N - Before	0.161 watt, 293-654°F	Not measured
After	0.166 watt, 310-660°F	0.221 watt, 323-750°F
Vendor	0.158 watt, 310-660°F	0.212 watt, 323-750°F
2P - Before	0.133 watt, 296-655°F	Not measured
After	0.135 watt, 307-650°F	0.197 watt, 325-752°F
Vendor	0.147 watt, 307-650°F	0.207 watt, 325-752°F

It is observed that the 2N elements are 4.2% above, and the 2P elements 4.8% below the vendor data, after thermal soak. On this basis, it was concluded that contact resistance in this size of element could be considered

negligible, and contacted element performance could be predicted from vendor curves without adjustment.

Element weights, also taken before and after thermal soak, showed an expected slight evaporative loss, which was the same for both 2P and 2N elements within experimental error. The calculated rate was -1.8×10^{-5} g/cm^2-hr. This rate, if constant with time, could cause a loss of about 1/3 of the hot junction area in 50,000 hours at 800°F in vacuum, calculated on the most pessimistic assumptions. Evaporation was considered not to be a limiting factor in operation up to 800°F, if fitted thermal insulation is used to retard evaporation by establishing a vapor-solid equilibrium.

Metallographic and electron-beam microprobe data suggested reaction and/or migration of Te at the aluminum PbTe interface, in the thermally soaked 2P elements. The extent was slight, but is evidence of a possible mode of contact degradation. It was clear that the particulate tungsten placed in the interface probably serves as an electrical bridge for this reaction zone, as well as for Al_2O_3. (The standard test element data are detailed in AI-66-260.)

B. DESIGN OF LIFE-TEST MODULES

1. Basic Considerations

In order that future operational data should be relevant to the test element data described, it was decided to vary as little as possible from the test elements in designing test couples. All dimensions, including aluminum strap thickness, were held identical; the only changes would be in strap length and configuration, as required to join the N and P elements into electrical series in a practical form.

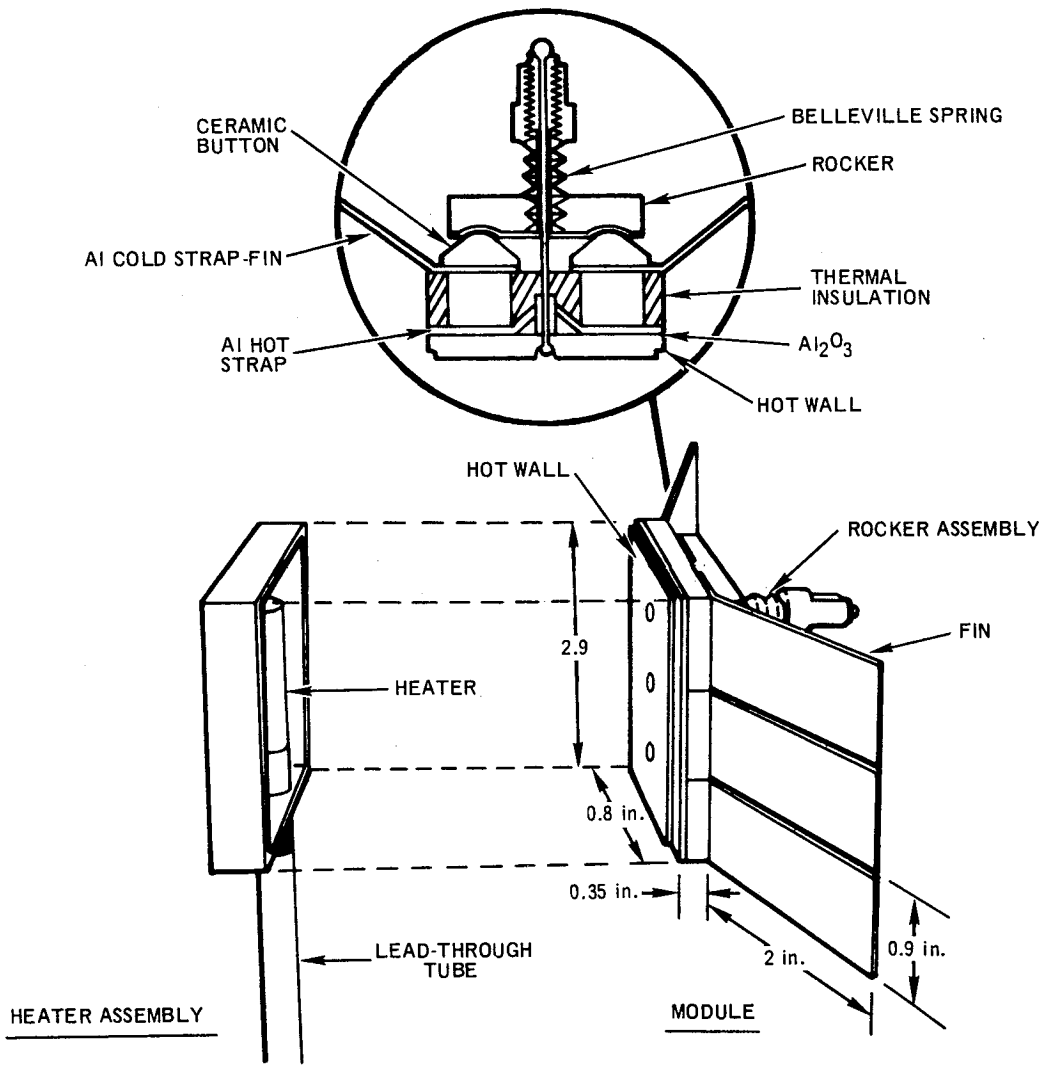

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FIGURE 2. SMALL MODULE TEST SCHEMATIC

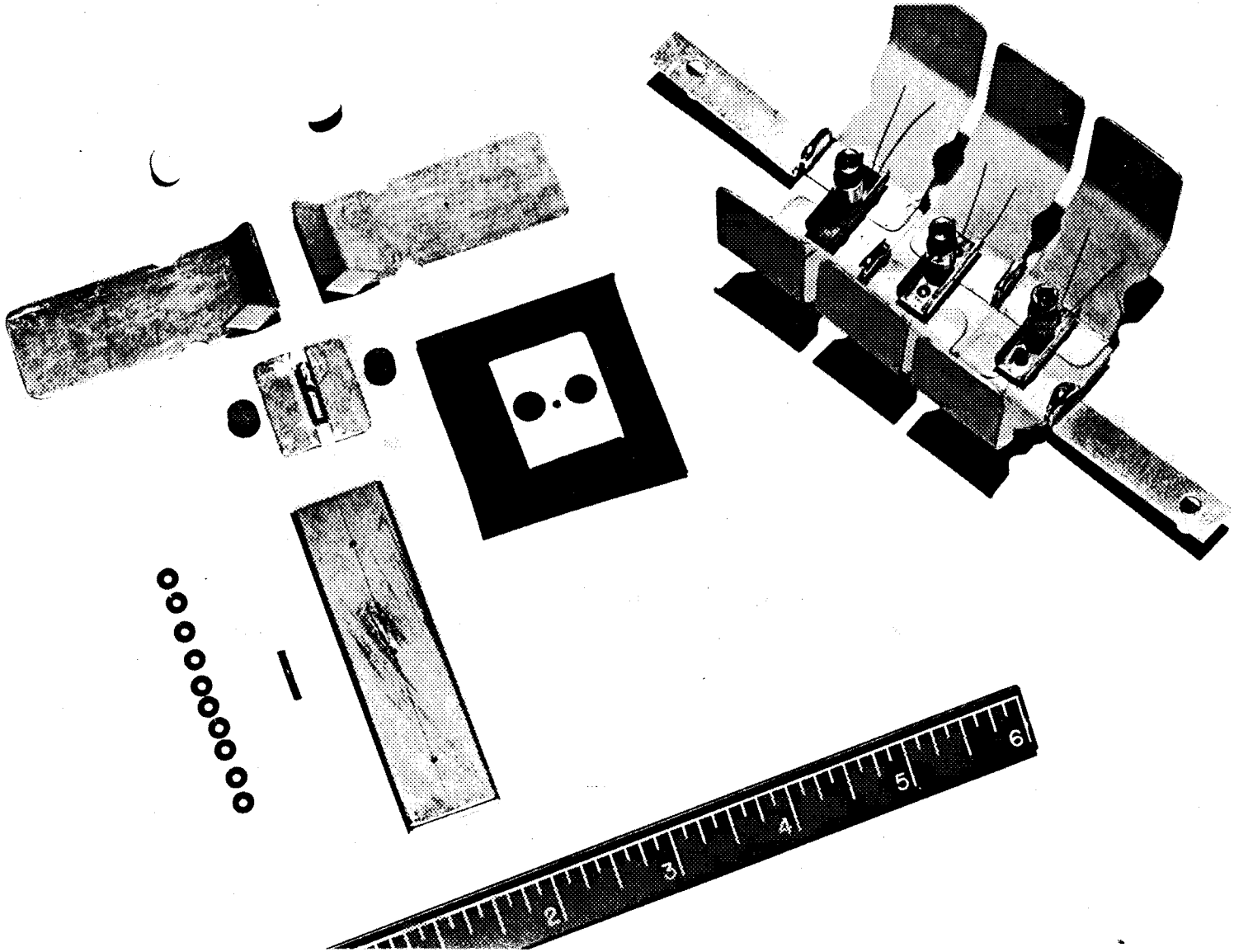

FIGURE 3. 3-COUPLE TEST MODULE AND PARTS

Although there was no mission or system requirement, it was decided to design couples that realistically represent practical use. The considerations here were:

1) Vacuum environment.

2) Heat rejection by radiation.

3) Heat collection by radiation.

4) Realistic mechanical structure.

2. Description of Modules

The circular insert in Figure 2 shows a section through a couple. The couple is tied to a Hastelloy "hot wall" and held in compression by a stud and rocker assembly. A special screw and nut compress a short stack of Belleville washers against a channel-shaped "rocker", which bears on two ceramic buttons, which bear in turn on the aluminum cold straps of the couple. The aluminum hot strap is convoluted to permit thermal compliance, and perforated to allow passage of the tension stud. The stud is of Rene' 41, an alloy chosen for low creep rate at high stress and temperature, and for low thermal conductivity. It contributes less than 2% heat shunt. The cold straps are configured to form heat rejection fins, when coated with a high-emissivity coating. The thermal insulation block is molded of commercial potassium titanate. The photo, Figure 3, shows a layout of couple components (except for stud, rocker, screw and nut) and a finished 3-couple module ready for attachment to a heater assembly. Note the cold straps and current leads T.I.G. welded to form the series circuit; also the thermocouple stub leads, which are provided at every hot and cold junction.

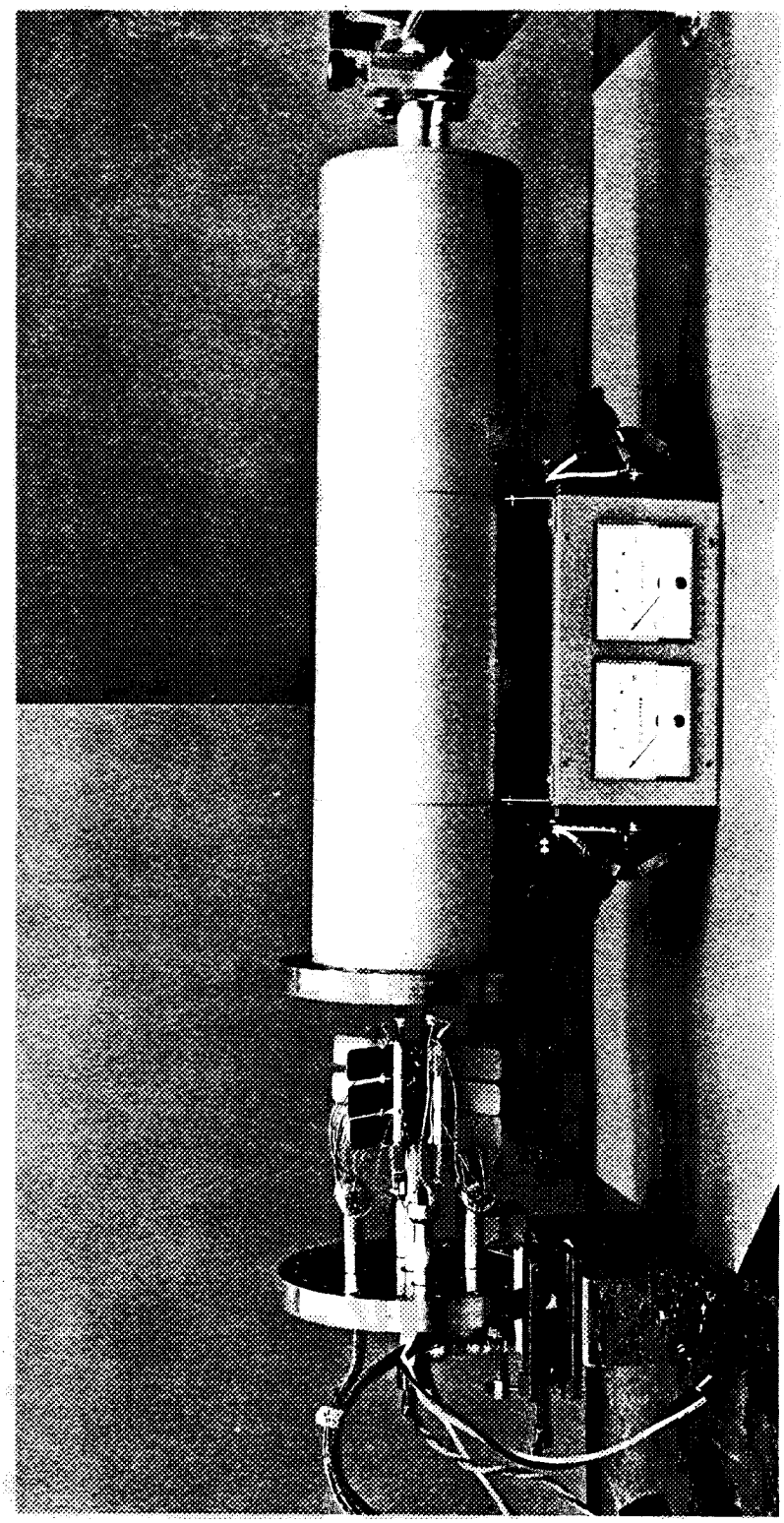

FIGURE 4. 6-INCH VACUUM TEST CHAMBER AND TEST MODULES

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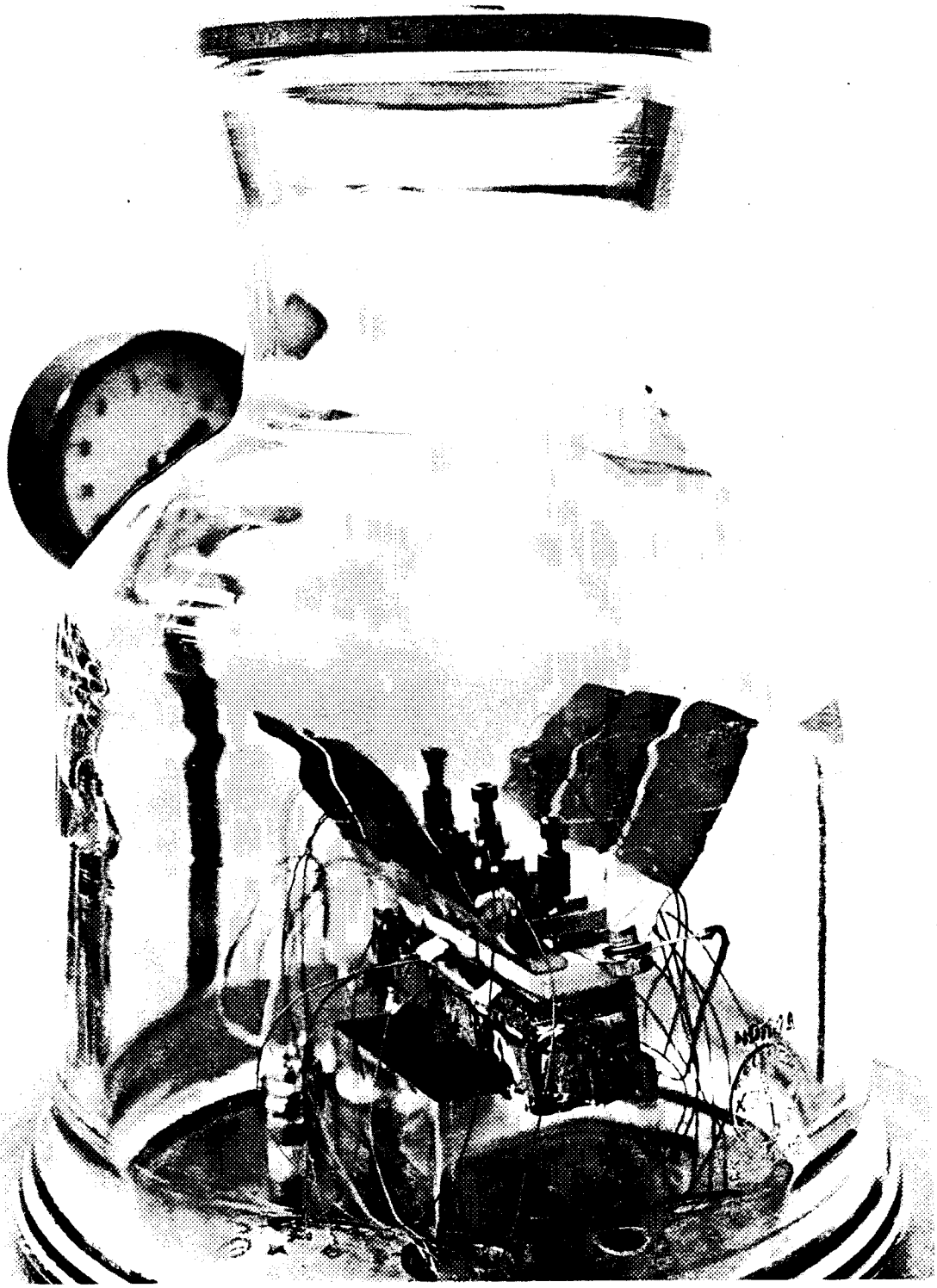

FIGURE 5. PILOT MODULE LIFE-TEST

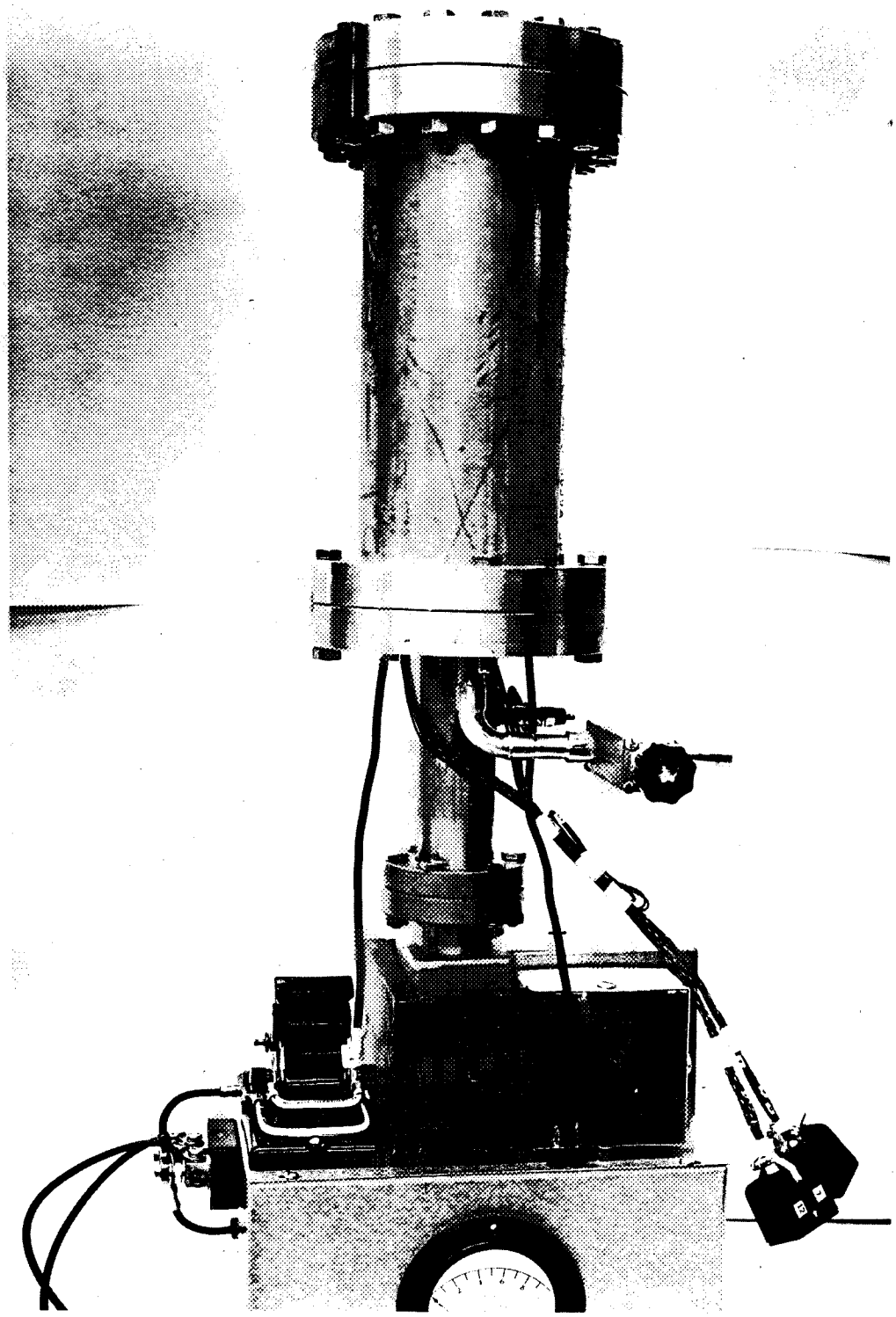

FIGURE 6. 4-INCH VACUUM TEST CHAMBER

In Figure 4 are seen two 3-couple modules assembled back-to-back on a common heater assembly, all instrumented, and ready for insertion in a vacuum test chamber.

All test modules were of the 3-couple size, fabricated exactly as shown. They were numbered serially SMT-1 through -6, the "SMT" standing for "small-module test". A subsequent program may include tests of larger modules.

C. TEST DESIGN

1. Test Chambers

The test chambers truly evolved with the program. The first test, SMT-1, was placed in a glass pipe reducer section, sealed with a Viton O-ring at top and bottom, and evacuated by ion pump through small-diameter tubing with 3 right-angled turns (see Figure 5). The vacuum at the module has perhaps been 2 or 3 decades poorer than the $\sim 10^{-6}$ torr measured at the pump. As components became available, a 4-inch diameter metal test chamber was built, Figure 6. It, also, accepted only one module, SMT-2. This chamber was, however, sealed by copper gaskets, and was directly open, through a 1-1/2 inch diameter pipe, into the ion pump. A vacuum of 10^{-6} to 10^{-7} torr is achieved at the module location.

The long test chamber already seen in Figure 4 is of 6-inch diameter. Improvements in lead-through seals have permitted vacuums better than 10^{-7} torr in two chambers of this size now in operation. A third test chamber of this size was nearly completed. The 6-inch diameter chambers are 24 inches long, intentionally designed to accommodate future modules of as many as 36 couples.

2. Instrumentation

Each test module is provided with electrical lead-throughs for 12 thermocouples, 2 current leads, and 2 heater leads. In some cases, heater voltage probes are also provided to allow monitoring heater power by watt meter. (A 6-inch test chamber has lead-throughs for 2 modules, totaling 36 lead-throughs, with 4 spares.) The thermocouple leads terminate in 12-point plugs, seen at the left in Figure 4. The mating plugs are wired to a jack-plug panel in a mobile instrumentation rack, as well as to a printout temperature recorder. The rack also contains a preamplifier - digital voltmeter combination; this can be plugged to any set of chromel thermocouple leads to measure voltage produced by individual elements, couples, or 3-couple modules.

Referring again to Figure 4: a switch is provided on each module current circuit to permit open and closed-circuit voltage measurements; a calibrated shunt (hidden by switch) is used with a potentiometer to measure current accurately; the panel ammeters are used only to show proper operation at a glance; the electrical load for each module is the sum of lead-wire and lead-through resistances, switch resistance, and ammeter resistance.

3. Test Method

For each new test module, the electrical load resistance is adjusted (by length and size of external lead wire) to be 1.25 to 1.50 times the module resistance at operating temperature.

The sealed chamber is evacuated and outgassed until it can be placed in steady operation on the ion pump, with the test at design temperature.

The heater input is adjusted, as required, to hold the average hot junction temperature of the hottest couple at 0 to 10°F above design temperature (700°F, 750°F, or 800°F).

The startup period usually includes one or more shutdowns to correct reversed leads, seal leaks, and clean out ion pumps. Therefore, some modules experience several thermal cycles and up to 100 hours operation before the test is considered to be satisfactory and stable. When the test is stable, the general procedure has been to read current, loaded voltage, and open-circuit voltage on the following schedule:

Up to 200 hours:	Daily, each element, each couple, and the module.
200 to 1000 hours:	Daily, each couple and the module; and weekly, each element.
Above 1000 hours:	Weekly, each couple and the module; each element only for special information, on occasion.

These data are recorded in log books. From them are calculated resistance, power, maximum power, and adjusted maximum power, as required for purposes of charting performance or making special interpretations.

D. LIFE-TEST RESULTS

1. Narrative

SMT-1, which originally was meant to be only a pilot test of slight duration, was permitted to remain in operation, because to do so cost so little. It has been unsatisfactory because of poor vacuum, and also because of erratic module contact with the cartridge heater, and consequent varying temperature distribution. The first 800 hours included 6 shutdowns for various corrective purposes and ion-pump clean-outs. Furthermore, at 2884 hours, a vacuum valve was accidentally opened, and left open for about 1-3/4 hours before the accident was discovered. This test, at nominal 750°F, is therefore given some reduced significance up to the 2884 hour point, and no significance thereafter. SMT-5 and -6, both at 750°F, are considered good replacements for SMT-1.

SMT-2, at 700°F, and SMT-3 and -4, at 800°F, are considered also to be very satisfactory tests.

The N elements of SMT-2 through 6 are segmented; i.e., they are hot-pressed of 2N and 3N materials, simply layered in the pressing die. The segmentation temperature is approximately 600°F.

2. Initial Performance Data

The term "initial" is taken here to mean the first set of data taken during the stable operational period. Table 3 gives initial power data for all 6 modules now on test, arranged in order of ascending temperature.

TABLE 3

INITIAL POWER DATA, SMT-1 THROUGH -6

Test No.	Time, hrs.	(T_H) max., °F	(T_H) av., °F	ΔT av., °F	P max. watts	P max/ΔT^2, watts/°F^2 x 10^{-6}
SMT-2	125	711	679	325	.649	6.14
-5	22	750	716	355	.803	6.37
-1	84	752	727	347	.694	5.76
-6	22	759	710	343	.790	6.71
-4	39	797	752	377	.911	6.41
-3	39	802	764	385	.949	6.40

The last column is included to show that a ΔT^{-2} adjustment for P max is valid; the difference between SMT-1 and SMT-2 to -6 is explained by 2N/3N segmentation; also, gradual process improvement is evident from SMT-2 through SMT-6.

There is a considerable temperature gradient in the test modules, caused by heat drain through the heater assembly support, and through the exit current leads. A representative complete set of power data is given to show this for one module,

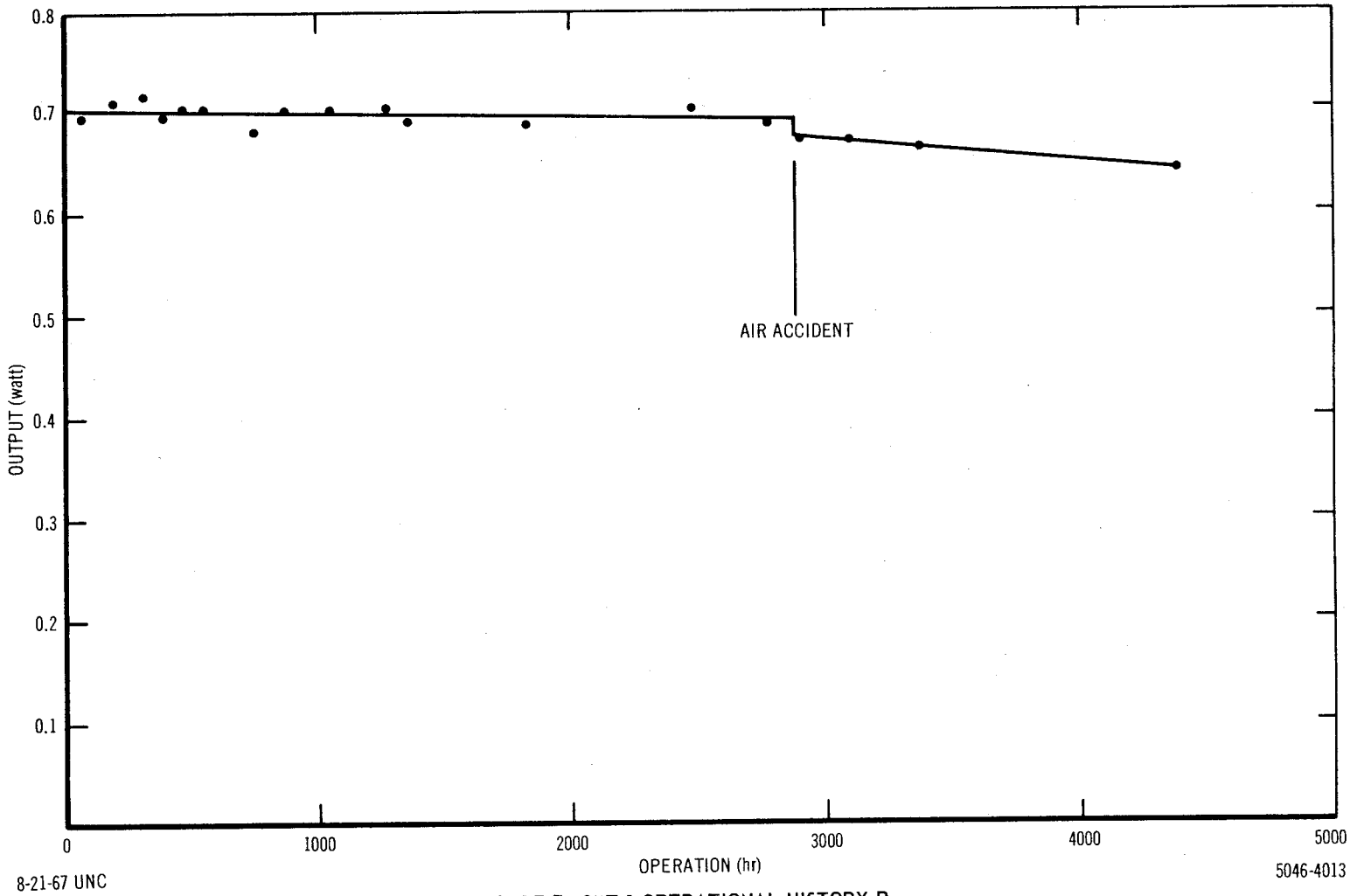

FIGURE 7. SMT-1 OPERATIONAL HISTORY P_{MAX} ADJUSTED TO INITIAL ΔT, VS TIME

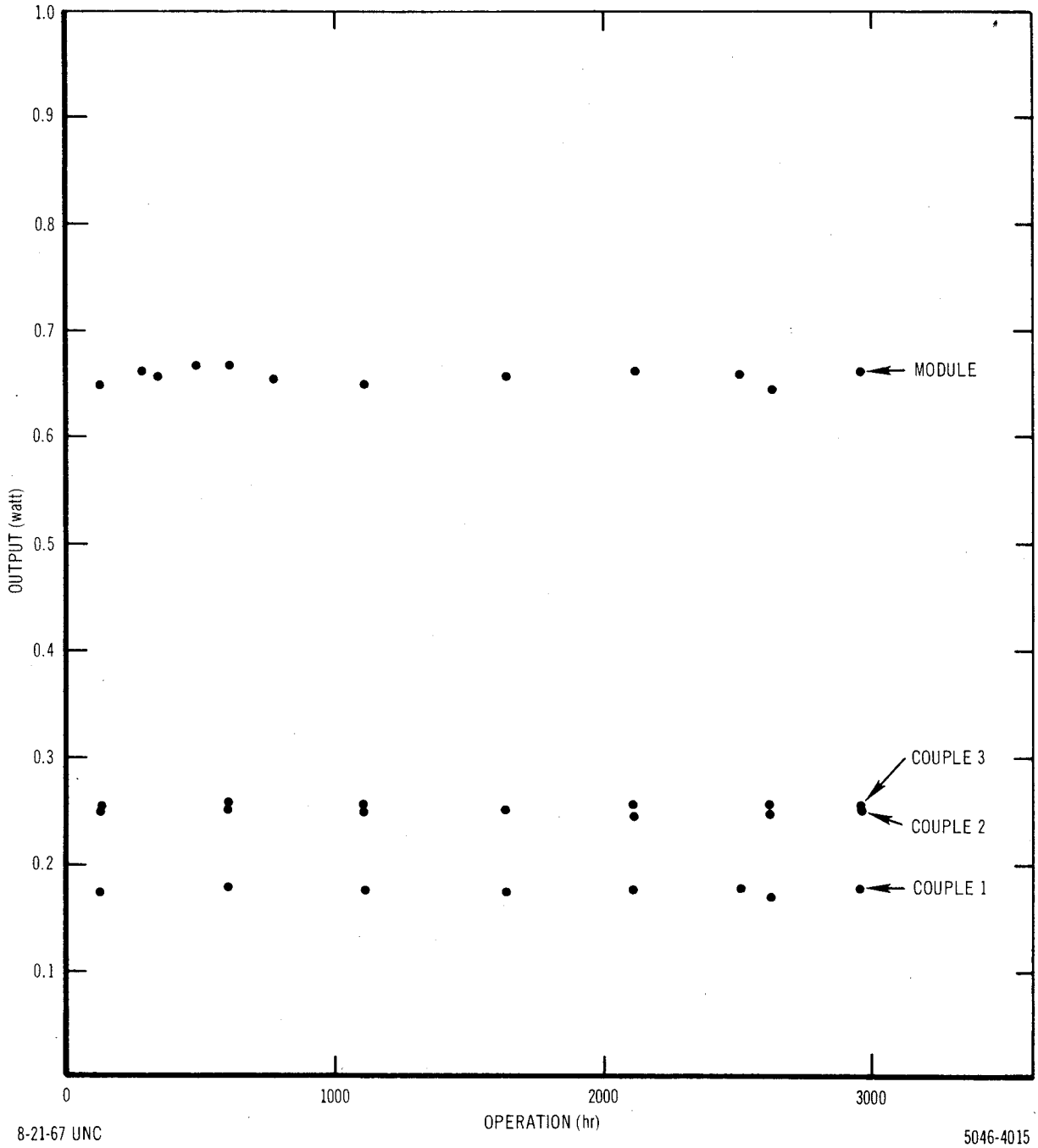

FIGURE 8. SMT-2 OPERATIONAL HISTORY P_{MAX} ADJUSTED TO INITIAL ΔT, VS TIME

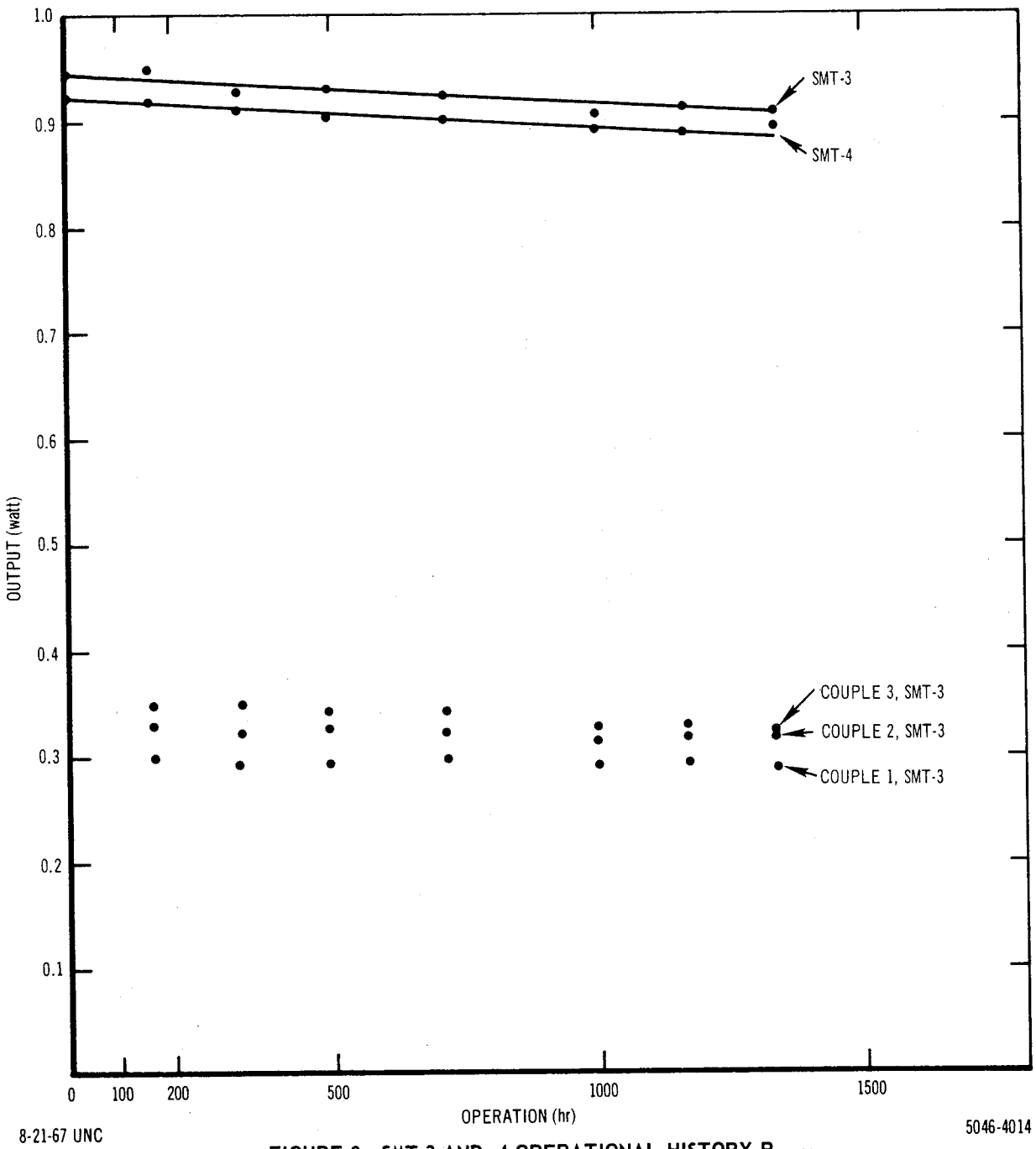

FIGURE 9. SMT-3 AND -4 OPERATIONAL HISTORY P_{MAX} ADJUSTED TO INITIAL ΔT, VS TIME

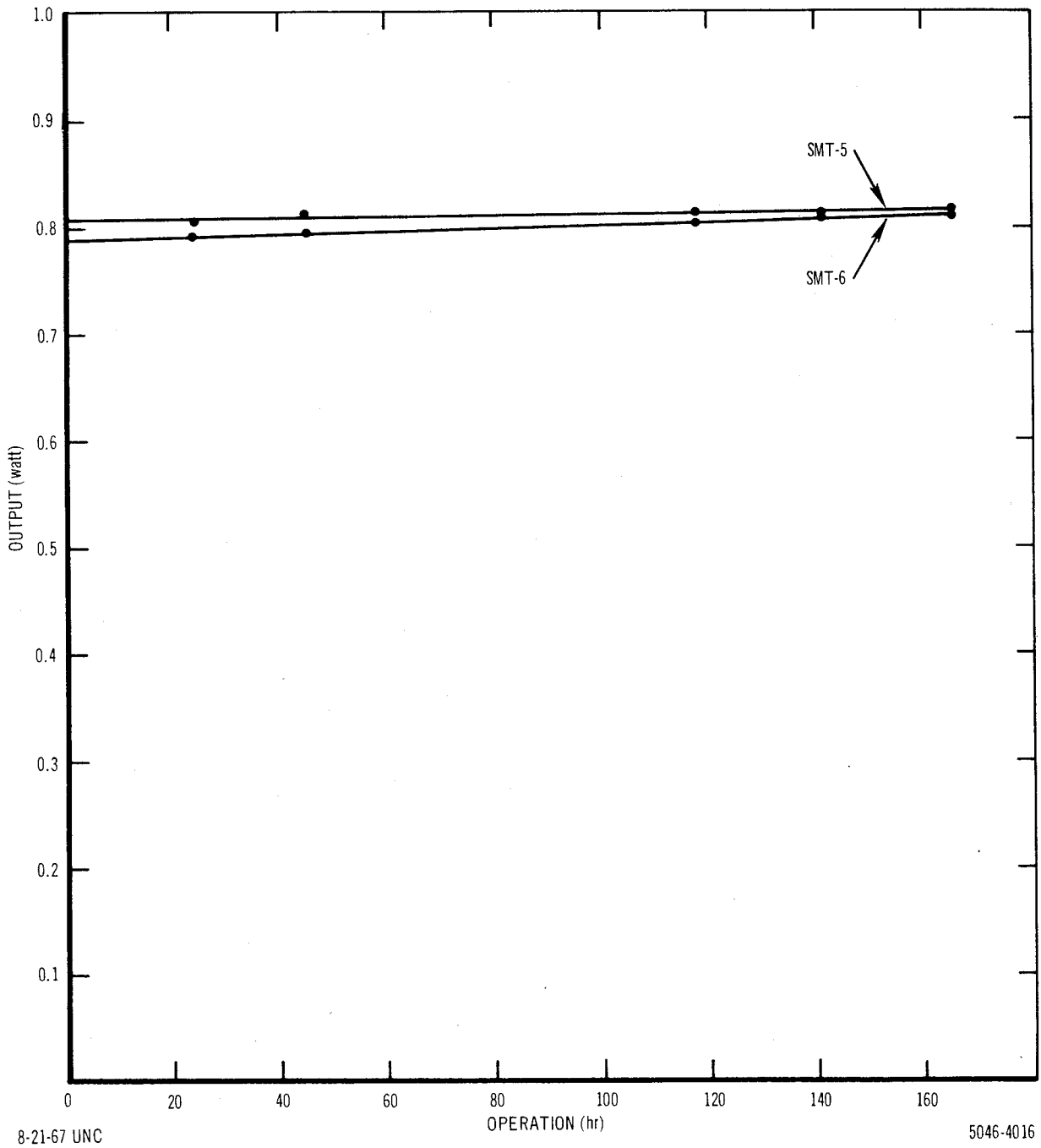

FIGURE 10. P_{MAX} ADJUSTED TO INITIAL ΔT, VS TIME

SMT-3 at 159 hours, in Table 4. From these data, it is calculated that the segmented couple will give about 0.40 watts at $T_H = 750°F$, $T_c = 325°F$, a practical set of temperatures for a space mission. From the data, also, it is calculated that the total strap losses are about 5.8%, of which 2.6% is in the cold straps. In a fully engineered system, the strap losses could be substantially reduced.

TABLE 4

POWER DATA, SMT-3 AT 159 HOURS

Element No.	Couple No.	T_H, °F	T_c, °F	ΔT, °F	P max, watts	P max/ΔT^2 watts/°F^{2}x10^{-6}
N_1		726	353	373	.169	1.21
P_1		727	384	343	.138	1.17
	1	727	369	358	.298	2.33
N_2		795	388	407	.190	1.15
P_2		799	391	408	.182	1.09
	2	797	390	407	.357	2.16
N_3		752	379	373	.167	1.20
P_3		753	372	381	.162	1.12
	3	753	376	377	.321	2.26
Module		759	378	381	.950	6.54

3. Stability

The Figures 7 through 10 show in graph form the operational history of the 6 tests now in operation. P max., adjusted to"initial" ΔT, is plotted against time. Brief comments on these are as follows:

SMT-1, Figure 7: The relatively large initial scatter in data points results from experimental temperature variations. Up to the air-exposure accident at 2884 hours, the degradation was at a steady rate, within experimental error.

SMT-2, Figure 8: The output is plotted for each couple, as well as the 3-couple module, to show consistency with time.

SMT-3 and -4, Figure 9: These modules are operating together in one chamber and are read in close sequence, so they follow the same experimental error variations. The 3 couples of SMT-3 are plotted also.

SMT-5 and -6, Figure 10: On this short time scale, 165 hours, an initial small power improvement is observed. It is thought that this probably occurred in SMT-1 (also at 750°F) but was masked by data-point scatter. In SMT-3 and -4 (at 800°F), it perhaps was masked by the small but significant degradation rate. At 700°F (SMT-2), the power appears to peak at 500 hours.

The power degradation rates have been calculated from the graphs of Figures 7 through 10, and are listed in Table 5. It is noted here that these degradation rates correspond remarkably with those observed on in-house programs, in which tests were performed in different configurations and environments. From these, it is believed that the degradation in SMT-3 and -4, at 800°F, will level off gradually, and become negligible after about 12,000 to 15,000 hours.

TABLE 5

POWER DEGRADATION RATES, TO DATE

SMT-1 THROUGH -6

Test	(T_H) max, °F	Time Period, hours	Power Change %	Rate, %/1000 hrs.
SMT-1	750	2884	-1.7	-0.59
-2	700	2954	0	0
-3	800	1334	-4.1	-3.1
-4	800	1334	-4.2	-3.2
-5	750	165	+1.2	meaningless
-6	750	165	+2.5	"

4. Efficiency

It has been observed on SMT-3 and -4, which have been monitored continuously by wattmeter for heater input, that the heater power has had to be steadily reduced to maintain the correct (T_H) max. Three representative data points are given in Table 6.

TABLE 6

POWER AND EFFICIENCY, SMT-3+4, vs HEATER INPUT

Time, hours	Heater Input, watts	(T_H) max., °F	(T_H) av., °F	ΔT av., °F	P actual watts	Efficiency %
39	55	802	756	381	1.832	3.33
1166	50	798	754	380	1.764	3.53
1334	50	813	767	388	1.857	3.71

These data were thought at first to show an increase in module efficiency: it is now believed, however, that most of the change is in the test efficiency. Specifically, vacuum cleanup of heater assembly surfaces may have reduced emissive losses. The 39 hour and 1166 hour data show similar T_H and T_c,

suggesting that the module heat through-put is unchanged - assuming constant radiator fin efficiency. It is still possible that the module efficiency is rising; e.g., the thermal resistance may increase with increasing electrical resistance. This possibility could easily be investigated in a future program.

An attempt has been made to calculate the true conversion efficiency of SMT-3 + 4. This involves identifying the experimental heat shunt losses and calculating their quantities. They are:

Conduction losses	
Heater leads	0.42 watts
MgO insulators	0.71
Heater tube	1.73
Radiative losses	
Heater tube	8.90
Heater box	7.30
Total	19.06 watts

Several approximations and estimates are involved in obtaining these numbers. Using them as best values, however, the initial true module conversion efficiency is calculated to be 1.832 watts/36 watts = 5.1%.

A confirmatory calculation of the module heat throughput, using vendor data for thermal conductivity, gave 31.9 watts for the elements, plus 4.0 watts for tension stud and thermal insulation heat shunts, totaling 35.9 watts. This improves confidence in the efficiency number.

If the module efficiency is 5.1%, at $\Delta T = 381°F$, then at $\Delta T = 425°F$ ($T_H = 750°F$, $T_c = 325°F$) the efficiency would be about 5.7%. Taking out the Carnot factor and module transmission (thermal and strap) losses, one obtains a very respectable <u>materials</u> efficiency of 19.3%, which includes contact losses.

E. CONCLUSIONS AND RECOMMENDATIONS

The summary conclusions are:

1) The aluminum contact can be used to form sound, practical PbTe couples.

2) The couple assembly can be made wholly of non-magnetic parts.

3) The couples can be operated in space vacuum environment for tens of thousands of hours, at 800°F or less, without special coatings.

4) Stability appears to be potentially excellent, Early data on degradation rates range from 0 at 700°F to 3.2%/1000 hrs at 800°F.

5) Data from other programs suggests that the initial degradation rates will decrease markedly with time.

6) The power density and efficiency data are excellent.

It is recommended that:

1) Life-testing of the present modules, and additional ones for confirmation, should be continued for 2 to 5 years.

2) Further or additional experiments should be performed to quantize more exactly certain basic engineering data, such as contact resistance, efficiency, mechanical integrity.