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TEST OF A COMBINED HEAT PIPE-THERMOELECTRIC MODULE

D. B. Lloyd

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D. B. Lloyd

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TEST OF A COMBINED HEAT PIPE-THERMOELECTRIC MODULE

D. B. Lloyd

ABSTRACT

A test was conducted on a heat pipe and thermoelectric module assembly to determine the performance characteristics of the combined system. The unit was built according to the ORNL reference design which was established to serve as the basis for a technology evaluation in the Isotope Kilowatt Program. The thermoelectric module was designed and fabricated by the 3M Company, and the heat pipe was designed and built by ORNL. Performance characteristics of the system were determined. The thermal efficiency varied from 7.2% at the beginning of the test to 5.1% at the end of the test. Because the loss in performance represents about 30% of the output, the module is not recommended for long-term use.

INTRODUCTION

The heat pipe-thermoelectric module employed in this test was based on a reference design for a thermoelectric isotope power unit which makes use of potassium-filled heat pipes to transport heat to the thermoelectric modules from a heat block-shield assembly. The heat block-shield assembly employs seven capsules of ^{90}Sr in the form of SrTiO_3 encased in Hastelloy C. The reference design system is described in more detail in Ref. 1, and Fig. 1 shows a schematic arrangement of the overall system.

Twelve thermoelectric modules, each with a 1-in.-diam 79-in.-long heat pipe, are designed to produce 167 W each at 24 V and can be coupled in a series-parallel arrangement to yield 2 kW(e) at 48 V. The thermoelectric module, evolved from design efforts at ORNL and the 3M Company, is shown in Fig. 2. The design is based on the use of lead telluride thermoelectric junctions, but is equally suited to use with advanced materials yielding a higher thermal efficiency. The heat pipe design, based on information received from Los Alamos Scientific Laboratory,^{2,3} employs a composite wick made according to techniques developed there.

ORNL-DWG 71-11982

Fig. 1. Reference design for a 2-kW(e) isotope power unit with thermoelectric converters.

ORNL-DWG 71-5147

Fig. 2. Sections through a typical thermoelectric module.

The test setup is shown in Fig. 3. Power was supplied to the heat pipe by a bank of electric heaters, and the thermoelectric module was cooled by circulating water through a calorimeter jacket on the outside of the module. The entire system was protected from the atmosphere by a shield can attached to the lower end of the calorimeter jacket.

HEAT PIPE

A heat pipe is a device that makes use of the latent heat of vaporization to transfer large amounts of heat with very small temperature differences. A simple heat pipe consists of a sealed tube lined with a capillary structure or wick. The wick contains the liquid phase of the working fluid and the remaining space contains the vapor phase. When one part of the system is heated and another part is cooled, a counter-current, two-phase flow is established. The vapor generated in the heated section flows to the cooled section, where it condenses and releases its latent heat of vaporization. The heat is removed from the condenser section by a cooling medium, and the condensate is returned by capillary action in the wick to the heated section.

Design

The type of heat pipe employed in this test is shown schematically in Fig. 4. In this design the annulus between the porous tube and the inner wall of the containment tube serves as the flow passage for the return of liquid from the condenser to the evaporator. The capillary pumping system used in this design is based on a technique (discussed below) developed at Los Alamos for fabricating thin, rigid, porous tubes that can have an extremely fine pore structure. Since the capillary force that maintains fluid circulation is developed at the liquid-vapor interface, it is desirable to have a fine pore at this interface; the pore structure in the remainder of the wick can be very coarse, or, as in our case, an unobstructed annulus, thereby reducing the pressure drop in the liquid flow path. The Los Alamos fabrication technique permits this sort of tailoring of the pore structure into what is commonly called a "composite wick."

Fig. 3. Heat pipe-thermoelectric module test configuration.

Fig. 4. Heat pipe structure.

Fabrication

Two heat pipes were fabricated in the first phase of the program. The first pipe was built and tested to evaluate our fabrication techniques and to familiarize ourselves with the operational characteristics of the design before operating the combined heat pipe-thermoelectric module. After the first heat pipe was found to be satisfactory, the second was assembled and filled in preparation for assembly with the thermoelectric generator at the 3M Company.

The porous tubes were fabricated by wrapping 12 layers of 400-mesh type 304 stainless steel cloth around a hollow copper mandrel, slipping another copper tube over the wrap, and then drawing the assembly through a reducing die to compact the wire cloth. The copper was then dissolved away with nitric acid and the compacted wrap sintered in vacuum at 1700°F for 15 min. The resulting tube is capable of withstanding normal, careful handling. The effective pore size in these tubes was determined to vary between 20 and 30 μ in diameter.

After the porous tube had been fabricated and cut to the proper length, end plugs were inserted into the tube, and an additional sintering operation was performed in vacuum to seal the wick to the plug. The subassembly was then installed in the tube that served as the outer wall of the heat pipe, and the closure welds were made. Figure 5 shows the final arrangement of the components.

After the heat pipe had been assembled, it was connected to a distillation pot which was subsequently filled with the proper amount of potassium. Figure 6 shows the arrangement of the heat pipe and loading apparatus. When the necessary heaters and thermocouples had been installed, the unit was insulated and connected to a vacuum system for outgassing. It was then heated to 940°F and outgassed for 16.5 hr. (The distillation pot was not heated during this period.) The indicated system pressure at the end of the period was 1.7×10^{-7} torr.

To transfer the 75-g charge of potassium from the distillation pot to the heat pipe, the temperature of the heat pipe was lowered to about 300°F, and the temperature of the distillation pot was raised to about 800°F. The potassium was transferred from the pot to the pipe in 25 min

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Fig. 5. Heat pipe construction.

Fig. 6. Potassium loading arrangement.

under these conditions. A dynamic vacuum was maintained on the system during the transfer. After transfer the tube between the heat pipe and the distillation pot was flame heated to a red heat to assure a potassium-free inner surface and then pinched shut, cut, and seal-welded. The heat pipe was then "wetted in" by heating to 800°F for 24 hr.

The fabrication procedure for the second heat pipe (the one to be used with the thermoelectric module) was the same as for the first except that, prior to mating the outer tube to the porous tube-end plug assembly, the nickel hot shoe (shown in Fig. 7) was machined and furnace brazed to the outer tube.

The second heat pipe was tested briefly to assure proper performance before shipment to 3M Company for assembly with the thermoelectric module. It was then heated in vacuum to 1800°F for 24 hr to allow any hydrogen that might have diffused into the pipe during the "wetting in" or testing phases to diffuse out of the system.

Testing

At design operating temperature (1060°F), the theoretical capacity of the heat pipe (21 kW) is far in excess of that required for satisfactory operation of the thermoelectric module (2.7 kW). It is also greater than the amount of heat that can be readily supplied or removed from the system. Thus the object of this test was not to measure the capacity of the heat pipe, but simply to demonstrate that the system as built would indeed supply the anticipated head load of the thermoelectric module.

The test stand configuration is shown schematically in Fig. 8. Electric heaters simulate the heat input from the heat block shield, and a water-cooled calorimeter of the same length as the thermoelectric module removes the excess heat. A shield can mates with a flange on the calorimeter jacket to permit maintaining an inert atmosphere around the heat pipe. (This avoids the problem of moisture in the air reacting with the heat pipe wall at elevated temperatures and the resulting hydrogen diffusing into the heat pipe.⁴) The shield can is fitted with a pivot mount that allows the entire test bed to be rotated through 180° to check the performance of the heat pipe in different attitudes. Thermocouples are

Fig. 7. Heat pipe-nickel sleeve assembly.

ORNL-DWG 73-222

Fig. 8. Heat pipe test configuration.

located along the outside of the evaporator end of the heat pipe. The close fit between the calorimeter jacket and the heat pipe made it necessary to provide a thermocouple well in the condenser end of the heat pipe in order to obtain a temperature measurement at that end. A differential thermopile was designed, fabricated, and calibrated to measure the temperature rise of the cooling water. This, in conjunction with a calibrated rotameter in the cooling water circuit, allows us to calculate the heat rejection rate within about 2%.

When we attempted to start the first heat pipe, we encountered a problem common to all heat pipes - a sonic flow limitation at low vapor pressures. The cooling load on the condenser exceeded the axial heat transport capacity of the heat pipe at low temperatures. This led to an accumulation of frozen potassium in the condenser which depleted the inventory of potassium in the evaporator. When the evaporator dried out, the temperature rose rapidly with no corresponding increase in the condenser temperature, and it was necessary to melt the accumulation in the condenser to redistribute the potassium. On the second attempt to start, the heat-pipe condenser was heated slightly in excess of the melting point of potassium (146°F), and the rate of heating of the heat pipe was increased so that the heat transport ability exceeded the cooling load before the excess inventory in the evaporator had been depleted. A thorough discussion of sonic flow limitations may be found in Ref. 5.

Failure of the differential thermopile early in the test prevented us from obtaining any performance data during these runs. We did, however, make a series of startup tests to improve our understanding of the system prior to attempting a startup of the heat pipe-thermoelectric module at the 3M Company.

During the startup tests we encountered a problem as a consequence of absorbed moisture in the thermal insulation. The moisture was driven out of the insulation in the hot zone and condensed on the flange near the water-cooled calorimeter jacket. A sensitive relay in a protective, ground current sensing circuit was activated, and the heater power was shut off. When the system was disassembled, enough moisture had collected in the insulation near the flange that it could be wrung out by hand. New, oven-dried insulation was installed, and the shield can was also

modified to allow it to be connected to a vacuum system. The heat pipe was outgassed in a vacuum furnace for 4 hr to remove hydrogen that had diffused through the wall during the operation. When the test program was resumed, the volume enclosing the insulation was evacuated for several days to remove additional moisture that might be released during heatup. Provisions were also made to avoid a similar problem with the unit to be used with the thermoelectric module.

When the test was resumed, it was found that the heat pipe performed as expected as long as the evaporator end was not elevated above the condenser end. When the evaporator was elevated, the temperature of the condenser began to fall rapidly and soon thereafter (about 15 to 30 sec) the evaporator temperatures began to increase steadily. When the position of the heat pipe was restored to an "evaporator-down" position before the temperature in the condenser dropped below the melting point, normal operation of the system could be restored.

A number of theories were advanced to explain the anomalous behavior of the system, but none were proved to be definitive. The present supposition is that the malfunction of the system was caused by a buildup of oxygen impurities in the potassium in the evaporator. Such a buildup would cause the surface tension of the potassium to decrease and would explain the apparent loss of pumping power. This theory is supported by work at Los Alamos⁶ and Aerojet General Nuclear Division.⁷ A detailed account of the efforts made to determine the nature of the problem and to restore normal operation is contained in Appendix A.

Additional heat pipe experience at ORNL also tends to support the above conclusion. In this work a "getter foil" of zirconium was included in the evaporator section of a heat pipe to absorb the oxygen from the potassium. A heat pipe with this design feature was operated satisfactorily with the evaporator end $2\frac{1}{2}^\circ$ above the condenser end. Further work is under way in an effort to determine the effectiveness of several combinations of hot trapping and cold trapping in reducing the impurities in the potassium and improving operation of the heat pipe when tilted from the vertical to less favorable attitudes.

A series of calculations were made using the code described in Ref. 3. Although the calculations were made for a slightly different heat

pipe geometry, of interest for a continuing development program, they do serve to show the interaction of the several parameters that govern heat pipe performance. The characteristics of the heat pipes used in the test program and those that were the subject of the calculations are given in Table 1.

Table 1. Comparison of heat pipes

	Pore size (μ)	Annulus width (in.)	Vapor region diameter (in.)	Length (in.)
First heat pipe	30	0.013	0.846	67.8
Heat pipe used with TEM	30	0.016	0.840	64.5
Calculated heat pipe	10-95	0.010-0.034	0.791-0.815	81

Figures 9 through 12 show the calculated performance envelopes of a heat pipe that was analyzed as a function of vapor temperature, annulus width, capillary pore size, and heat pipe inclination angle respectively.

THERMOELECTRIC MODULE

The heat pipe-thermoelectric module (TEM) design characteristics are summarized in Table 2. The module consists of a cylindrical array of thermoelectric legs arranged around a nickel sleeve that is brazed to the condenser end of a heat pipe. The nickel sleeve was machined to provide 16 flat surfaces about the circumference for mounting 16 rows of thermoelectric couples, and strips of alumina were placed on each flat surface to provide electrical insulation between the nickel sleeve and the hot junction electrodes. A compression spring was used to maintain contact between the hot junction electrode, the thermoelectric leg, and the cold junction electrode. A follower bar formed the cold frame structure for each row of couples. The outer case and the end closures formed a hermetic seal for the system. The voids are filled with MIN-K 1301

Fig. 9. Calculated performance envelope for a heat pipe as a function of vapor temperature.

Fig. 10. Calculated performance envelope for a heat pipe as a function of annulus width.

ORNL DWG. 72-1628

Fig. 11. Calculated performance envelope for a heat pipe as a function of pore diameter.

Fig. 12. Calculated performance of a heat pipe as a function of orientation.

Table 2. Thermoelectric module design summary

N-leg material	Segmented PbTe
P-leg material	Segmented BiSbTe/PbSnTe
Hot junction temperature	BOL (beginning of life): 1000°F EOL (end of life): 939°F
Power output	BOL: 202 W at 24 V EOL: 169 W at 24 V
Cold junction temperature	150°F
Heat input BOL	2375 W
Efficiency BOL	8.5%
Number of couples	208 (16 rows of 13 couples each)
Outer diameter	4 in.
Length	15 in.

insulation and purged and backfilled with argon. The power and instrumentation leads are brought out through ceramic-to-metal headers. A more detailed design description may be found in Refs. 1 and 8.

Fabrication

The heat pipe with the machined nickel sleeve was built and tested at ORNL and then shipped to the 3M Company for assembly with the thermoelectric module and for preliminary tests. The calorimeter jacket, flange, and shield can were also fabricated by ORNL and sent to the 3M Company for assembly. After the module had been assembled and the calorimeter and flange had been attached, heaters, thermocouples, and insulation were applied to the heat pipe, and the assembly was enclosed by the shield can as shown in Fig. 3.

Shakedown Tests at 3M

Before starting the heat pipe, the temperature of the entire system was raised above the melting point of potassium (146°F) to reduce the

load on the heat pipe during startup and avoid freezing of the condensing potassium. Power (~2 kW) was applied to the internal heaters, and the heat pipe was started without difficulty. As part of a proprietary processing step in the manufacture of the module, 3M intended to raise the temperature of the hot junction to 1200°F within an hour after starting the system. This proved to be impossible because they had not installed sufficient power to give the required heating rate.

While the temperature of the system was still climbing, there was a step decrease in the maximum input power level from 2.7 to 2.2 kW about 3 hr after the startup. It was surmised (and later determined) that a lead to one of the internal heaters had failed. From this point on, temperatures in the system began to drop slowly until the temperature at the condenser end of the heat pipe was about 1100°F.

At this point the decision was made to shut down the system, allow it to cool, and check the heaters and wiring to determine the cause of the trouble.

When the shield can was removed, it was discovered that the nickel lead wires to the internal heaters had become severely embrittled, and one had broken. (The broken lead accounted for the sudden decrease in input power.) A yellow deposit had also formed on the surface of the Microquartz insulation next to the shield can. In searching for the source of the yellow deposit, it was found the 3M Company had substituted hose clamps to hold the two halves of the clamshell heaters together instead of nickel or Nichrome wire. Unfortunately, the adjusting screws and screw housings on these clamps were cadmium plated. The high temperature encountered during the startup caused the cadmium plating to evaporate from the hose clamp screws and condense on cooler portions of the equipment.

At our request, 3M forwarded to ORNL samples of (1) the embrittled nickel lead wire, (2) unused nickel wire from the same lot, (3) the Microquartz insulation with the yellow deposit, and (4) a clamp similar to those used on the heaters. (Those actually used had been discarded.)

In view of the potentially serious consequences, 3M was also requested to hold the system at room temperature until we had an opportunity to evaluate the results of the various analyses.

Preliminary results of the analysis of the samples at ORNL indicated the following:

Insulation. The yellow deposit contained both sulfur and cadmium in roughly equal concentrations. (Cadmium sulfide is a yellow substance commonly used as a pigment.) There was no evidence of cadmium anywhere in the insulation except in the yellow deposits, but sulfur was found throughout the insulation.

Nickel wire. The unused wire showed no evidence of cadmium but about 200 ppm sulfur. The embrittled wire had 1000 ppm sulfur and a surface contamination of cadmium.

After these results were obtained, ORNL requested 3M to send a sample of unused Microquartz and, if possible, scraping or filings from the heat pipe-nickel sleeve assembly that would permit us to evaluate the severity of the sulfur contamination.

The primary concern at this time was the integrity of the weld lip on the nickel sleeve (Fig. 2) because if this should become embrittled and fail, the protective atmosphere inside the thermoelectric generator would be lost. However, the 3M Company stated that, in the event of a containment failure, the system could be operated satisfactorily if a blanket of pure argon were maintained around the thermoelectric elements. The 10,000-hr test objective for the module required that extremely low levels of oxygen and water vapor be maintained in the argon.

After analyses of the additional samples received from the 3M Company, the following account of the situation was made.

1. The unit operated about 9 hr. The nickel heater wire failed after about 3 hr.
2. The heater wire leads were buried in thermal insulation; hence their temperature was indeterminate.
3. The 3M test differed from the ORNL heat pipe test only in the use of cadmium-plated clamps instead of Nichrome wire on the clamshell heaters, Microquartz instead of Kaowool insulation, and omission of a cold trap in the line to the vacuum pump.
4. Yellow freckles containing cadmium and sulfur, probably CdS, covered about 80% of the outer surface of the thermal insulation.

5. The Microquartz insulation used by 3M had 300 ppm sulfur, whereas the ORNL Kaowool had 10 to 80 ppm.

6. The clamps on the heaters contained 375 ppm sulfur in the cadmium plating. (Note: Cadmium is commonly plated from a cadmium sulfate solution. This explains the presence of sulfur in the cadmium plating.)

7. The nickel lead wires looked all right, but they were very brittle, especially at the heater end.

8. The ORNL analysis for sulfur in scrapings from the unit showed 1000 ppm in the heater lead wire surface, 200 ppm in the unused heater lead wire, 110 to 440 ppm on the surface of the stainless steel heat pipe, 120 ppm in drillings 1/32 in. deep in the nickel heat block, and 7000 ppm on the surface of the nickel bus bar.

9. Less than 10 ppm of cadmium was found on the surface of the heater lead wire; none inside.

We concluded from the foregoing information that the most likely source of trouble in future tests of the unit in a sulfur-free setup would be the region of the weld between the nickel heat block and the hermetic end bell, which was made of 0.020-in.-thick stainless steel. The nickel in this region could have been embrittled by sulfur. Note: Sulfur attack is a common problem when nickel components are operated at high temperature in an atmosphere containing even trace amounts of sulfur. Sulfur and nickel react at temperatures above 315°C (600°F) to form nickel sulfide, which melts at 550°C (1022°F), congregates in the grain boundaries, and causes hot shortness.

It was decided that the least risky course of action was to avoid disturbing the weld region in question and to proceed with care in the continued test program.

Accordingly, the 3M Company was requested to:

1. Abrade all surfaces inside the shield can to clean off any sulfur or cadmium deposits.

2. Install new heaters furnished by ORNL.

3. Install a valve on the instrumentation header to permit pressurizing the thermoelectric module with argon in the event of a leak in the hermetic seal. (3M was to specify the argon purity required.)

4. Install a cold trap between the vacuum pump and the shield can.

5. Install a 50-A power supply line.

After the system had been reassembled, the 3M Company restarted the system. After about 6 hr a leak developed in the suspected weld and pressure in the module began to decrease. The system was operated for an additional 2 hr, and the electrical heaters were then turned off and the system allowed to cool down. The valve on the instrumentation header allowed 3M to maintain an argon atmosphere inside the generator, thereby avoiding damage to the junctions.

The leak in the seal weld between the stainless steel end bell and the nickel sleeve was repaired by grinding out most of the material in the trepanned projection from the nickel sleeve and rewelding the stainless steel end bell to the nickel sleeve using an Inconel 82 filler rod. In addition, an additional Inco 600 end bell was welded in to close off the region between the end of the nickel sleeve and the outer stainless steel jacket. Figure 13 shows details of the repaired region.

Performance Tests at 3M

After the above repairs, 3M initiated the comprehensive set of performance tests specified in their contract and completed them with no further difficulties on July 29, 1971.

The experimental points from these tests are plotted in Figs. 14 and 15 superimposed on a set of curves that were calculated for the module in the course of the design work. The difference between the performance given by the experimental points and the calculated performance apparently stems from a somewhat higher internal resistance in the unit which might be expected from the fact that, because of the difficulty with the sulfur embrittlement, 3M did not operate the unit at as high a temperature as planned during their run-in process in which the electrical bonds were effected. However, 3M's personnel were of the opinion that this bonding might improve with extended operation, and offered as evidence that they had observed some improvement in a similar module that they built and had been operating at 3M.

Fig. 13. Repair of thermoelectric module hermetic seal.

Fig. 14. Comparison of calculated and experimental thermoelectric module performance characteristics at beginning of life conditions.

Fig. 15. Comparison of calculated and experimental thermoelectric module performance characteristics at end of life conditions.

Testing at ORNL

The thermoelectric module was received from 3M and installed in the ORNL test stand. The heater arrangement was similar to that developed for the heat-pipe test shown in Fig. 8. Cooling for the TEM was provided by a closed-circuit demineralized water system. The water was circulated by a small centrifugal pump with the flow going from the reservoir, through a rotameter, the generator cooling jacket, a heat exchanger, and back to the reservoir. In the event of a coolant flow stoppage caused by either an electrical power outage or a pump failure, a solenoid drain valve would open and allow coolant from the reservoir to continue to flow through the module cooling jacket. At the same time an automatic control would interrupt the power to the heaters. (These provisions were necessary to prevent the soldered connections in the module from becoming overheated and melting.) The reservoir contained approximately 130 gal of water, which was enough to provide emergency cooling until the system temperatures had all dropped below 200°F.

The system was started without incident on August 24, 1971, and operated satisfactorily. Initially a maximum hot junction temperature of about 870°F was employed with a heat pipe temperature of 1000°F. This temperature level was maintained until the instrumentation necessary for determining open circuit voltages was calibrated and installed. Then the power input was increased to bring the hot junction temperature to 1000°F.

The system was operated with a vacuum in the shield can around the heaters and the lower portion of the heat pipe in an attempt to remove an accumulation of hydrogen from the interior of the heat pipe. (Apparently the thermal insulation still contained some moisture when the test was initiated at 3M.) The presence of the hydrogen in the heat pipe was manifested by a nonuniform axial temperature distribution evident from the thermocouples mounted along the length of the module.

A 150°F difference existed between the points measured by two thermocouples which were 5/8 and 4 5/8 in. from the upper end of the nickel sleeve. The difference in temperature decreased as the system temperature was increased. (This type of behavior is typical when a noncondensable gas is present in the system. As the temperature increases, the working-

fluid vapor pressure increases and compresses the noncondensable gas to a smaller volume.) Figure 16 is a plot of this temperature difference as a function of the potassium vapor temperature in the heat pipe.

The output of the TEM was connected into the circuit shown in Fig. 17. Normally the module was allowed to charge a bank of lead-acid batteries, but once a day a timer circuit closed a switch that allowed the batteries to discharge into a resistance load. A shunt regulator was incorporated in the circuit to prevent overcharging the battery bank. [The shunt regulator (described in Appendix B) diverted a portion of the generator output through the shunt load if the output voltage rose above a preselected level.]

When it was necessary to make a performance map of the generator, it was possible to switch the output from the battery bank to a variable load. Another switch in the circuit made it possible to interrupt the current flow to obtain data from the generator under open circuit conditions. When the switch was activated the data logging system automatically began recording the data.

As the test progressed, the temperature difference between the two thermocouples began to decrease. The data points on Fig. 16 show the decrease in ΔT with the passage of time. It appears that the hydrogen gas that entered the heat pipe during the initial testing at the 3M Company gradually diffused back out of the system into the vacuum surrounding the heat pipe.

During a routine test of the protective circuitry, power to the heaters was interrupted for about 15 to 20 min. During this period the high-temperature portions of the system experienced a 400°F drop. When the power had been restored and the temperatures had returned to normal, it was noticed that some of the module internal instrumentation had failed, and it was no longer possible to monitor the ΔT at the end of the nickel sleeve.

The internal resistance of the generator was measured periodically. Prior to the above temperature cycle, the internal resistance had been exhibiting a steady increase as shown in Fig. 18. Measurements made immediately after the transient showed that there had been a further sharp increase in the internal resistance. It continued to increase for

Fig. 16. Temperature gradient along nickel sleeve as a function of heat pipe vapor temperature.

Fig. 17. Thermoelectric module external connections.

Fig. 18. History of internal resistance of 3M thermoelectric module.

two days and then began to decrease. The seemingly random variation in internal resistance shown in Fig. 18 has not been explained at this writing.

Figure 19 is a plot of a daily variation in some of the key heat pipe and TEM parameters. The data relating to a current output of 5.673 A are plotted but have been disregarded as far as drawing the curves is concerned. These data were all taken for the conditions in the load cycle that prevailed shortly after the battery bank began to discharge. Prior to that time the battery bank was fully charged, the current output was about 4.7 A, and the vapor temperature, hot junction temperature, and voltage were at the high end of their range. The particular data points which all fall above the curves were taken before the system had "leveled off." For the same reason the output power also falls above its expected position.

A common failure mode in a TEM is the loss of the inert gas protective atmosphere and subsequent deterioration of the thermoelectric materials. Figure 20 is a plot of the module internal pressure during the test program. When the gas pressure had fallen to a dangerously low level, it was necessary to repressurize the system to continue the test. Accordingly, a supply of very pure argon (see Table 3) was connected to the valve on the TEM and was admitted to the generator through a dry ice and alcohol cold trap to minimize the water content. However, eventually the leak rate became so great that it became necessary to repressurize the module every few days in order to maintain a satisfactory pressure.

Table 3. Argon analysis

Argon	99.998%
O ₂	<0.0001 (1 ppm)
N ₂ + CO	0.0005 (5 ppm)
CO ₂	<0.0001 (1 ppm)
H ₂ O	<0.0004 (4 ppm)
H ₂	0.0011 (11 ppm)
CH ₄	<0.0004 (4 ppm)

Fig. 19. Daily variation of thermoelectric module parameters as a function of module output current.

Fig. 20. History of gas pressure of 3M thermoelectric module.

In an attempt to determine the location of the leak, a small quantity of helium was mixed with the argon gas used to repressurize the module. A helium mass spectrometer leak detector was used to locate the leak. No evidence of any leak was found at the lower end of the unit. (Apparently the repair made by 3M was still holding.) There were, however, several areas on the instrument header (on the top of the module) that were leaking. Each of the leaks was associated with an electrical feed-through. The area that gave the strongest signal to the leak detector was one of the feed-throughs for the module output; the rest of the feed-throughs all seemed to be leaking but at a slower rate.

The heat pipe-thermoelectric module continued to operate satisfactorily except for the leaks until March 19, 1972, when it was intentionally shut down to permit repair of the leaks that had developed in the instrument header. At the time of the shutdown the module had accumulated 4989 hr of continuous operation.

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Prior to potting the header it was necessary to disconnect the wiring between the feed-throughs and the terminal block to provide access to the header plate for cleaning. Because the success or failure of this type of operation depends, in large measure, on the cleanliness of the surfaces involved, it was imperative that the header plate be thoroughly cleaned prior to the potting operation. After the wiring was disconnected, the header plate was rinsed with ethyl alcohol, abrasively cleaned by grit blasting with alumina, washed with trichloroethylene, rinsed with ethyl alcohol and distilled water, and then dried with a hot air blast. After pouring the potting material the cover gas pressure was reduced slightly below atmospheric while the potting compound cured.

When the system was restarted, it was determined that no significant change in the leak rate had been effected, so no further attempt at solving the problem was judged to be feasible.

The system was operated for an additional 694 hr and then, at the request of the AEC, terminated. Total operating time on the module was 5683 hr. It was subsequently restarted briefly to obtain performance maps of the system so that its performance could be compared with that at the beginning of the test program.

Results and Recommendations

The performance of the TEM at the end of the test and at the beginning of the program is shown in Fig. 21. It may be seen that the internal resistance had risen from about 2.5Ω to between 4 and 5Ω . As a result, the output voltage and power of the module were drastically reduced. Also, the upturn at the end of the curves for the hot junction temperature and potassium vapor temperatures is not typical for such a system. Normally, Seebeck cooling serves to reduce the system temperatures as the current flow increases. In this case, the sharp increase in internal resistance with increased current flow causes enough I^2R heating inside the module to overshadow the cooling effect.

The peak efficiency of the module decreased from 7.2% at the beginning of the test to 5.1% at the end of the period.

No problems were encountered as a result of mating the thermoelectric generator to the heat pipe and no change in the heat pipe operation was detected over the period of the test.

The thermoelectric module tested in this program cannot be recommended for long-term use until the reasons for the severe loss in performance are determined and corrected. Additional effort must also be made to provide a reliable seal around the power leads from the module.

In addition, work should continue on the heat pipe system to determine cleanup techniques to be applied to the potassium. This appears to be necessary in order to assure proper operation of the heat pipe under accident conditions when it might be tipped on its side.

Fig. 21. Comparison of beginning of life and end of life for heat pipe and thermoelectric module parameters as a function of thermoelectric module output current with constant power setting on heat pipe heaters.

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APPENDICES

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Appendix A

HEAT PIPE INVESTIGATION

At the point in the test program where the anomalous behavior of the heat pipe was found, an extensive series of tests was initiated in an effort to resolve the problem. The nature of the problem and the various steps in the study are detailed here.

It was noted that the temperature distribution along the heat pipe was strongly dependent upon the inclination of the heat pipe. With the heat pipe in any position between vertical with the condenser end up and horizontal, the temperature was essentially uniform throughout the length and the heat-transport capacity was high. However, if the evaporator end of the heat pipe was raised above the condenser end, the temperature distribution diverged and the heat-transport capacity of the heat pipe dropped sharply. (See Fig. A.1.)

In attempting to diagnose the difficulty, the first thought was that there might be noncondensable gas, particularly hydrogen, present in the heat pipe. Another hypothesis advanced was that the wick structure was not adequately sealed to the wall at the evaporator end of the heat pipe, in which case the wick could lose its prime if the condenser were lower than the level of the boiler.

The presence of an inert gas bubble between the wick and the heat pipe wall could also act to break the suction formed by the capillary pores in the wick and thus would negate the strong pumping effect of the 30- μ pores in the wick. If this occurred with the condenser above the evaporator, gravitational forces would still act to recirculate the liquid, and there might be no observable effect on the heat-transport performance. However, if the condenser were below the evaporator, gravitational force would act to resist recirculation. The only force acting to produce recirculation from the condenser to the evaporator would be the capillary force acting on the free liquid surface between the wick and the heat pipe wall. (The nominal radial gap at this point was 0.017 in., but the wick was free to be eccentric. The gap on the close clearance side could be in the range of 3 to 5 mils. Inasmuch as the 30- μ pore size is about 1 mil,

Fig. A.1. Axial temperature distribution for the heat pipe in two typical positions, with the condenser end inclined upward with a slope of $+1.5^\circ$ and with the condenser end inclined downward with a slope of -10° .

the capillary force would drop to perhaps 25% and the heat-transport capacity of the heat pipe would be cut by the same factor.)

If one examines Fig. A.1 with this rationale in mind, it appears that the experimental data can be explained as follows. The large increase in temperature of the evaporator, when it was raised above the condenser, implies a pronounced increase in heat losses from the heater region and consequently a reduction in the heat input to the heat pipe. Similarly, the drop in the condenser temperature (from 420 to 320°C) indicates a reduction in the heat-transport rate.

To investigate these problems it seemed eminently desirable to evolve a set of nondestructive tests that could be employed for inspecting heat pipes in production. The first step taken was to remove the heat pipe from the test stand and radiograph it. When examining the films, it was noted that the interior details of the heat pipe were easy to discern. The annulus between the porous tube and the outer tube could be seen, but it was not possible to determine whether or not the annulus and porous tube were filled with potassium. The pool of excess potassium at the bottom was easily seen, and the amount of excess potassium was estimated as ~ 0.9 in.³ more than one would expect if the wick and annulus were completely full, thus implying a void behind the wick.* The radiographs also revealed the presence of a void about 11/16 in. in diameter between the tapered plug and the end cap at the fill end, as shown in Fig. A.2.

The heat pipe was then subjected to a phase-sensitive eddy-current test, which measures the local conductivity of the material being tested. It was hoped that the test might show the effects of whatever was causing the anomalous behavior of the heat pipe. However, inquiries indicated that this technique had not been used to study heat pipes, and without prior experience or a test standard, it was difficult to analyze the results of such a test. The test did indicate that there was a basic eccentricity in the pipe (probably the eccentricity of the porous tube in the outer tube) with other eccentricities superimposed over various

*When the heat pipe was filled initially, it was loaded with 20% excess potassium to insure an adequate supply of the working fluid.

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Fig. A.2. Heat pipe — evaporator end.

portions of the length along the pipe. However, analyses have shown that eccentricity of the wick structure should not have an adverse effect on the heat pipe performance.

At this point it was decided that the time lost in installing and removing the heat pipe from the test facility could be reduced by conducting the performance check tests in a tilting muffle furnace. After an initial performance test to familiarize ourselves with the heat pipe operation in the muffle furnace, the joint between the wick and the outer wall at the evaporator end of the heat pipe was rerolled. The heat pipe was then reinstalled in the muffle furnace, but the performance was found to have deteriorated. A new radiograph showed that the second rolling operation had acted to drive the plug out from under the rolled region. Fortunately, there was still room for a second rolling operation farther toward the end of the heat pipe, and this operation was carried out. Retesting of the heat pipe showed that the performance was restored to the level obtained before the end plug had been dislocated.

The following operations were then carried out on the heat pipe in an effort to eliminate hydrogen and/or argon which (as a noncondensable gas) was thought to be the cause of the trouble:

1. Vacuum outgas at 1600°F for 13 hr to allow any hydrogen to diffuse through the heat pipe wall.
2. Performance test the heat pipe.
3. Vacuum outgas again at 1600°F for 13 hr.
4. Performance test the heat pipe.
5. Cut off the pinched-off portion of the fill tube and install a valve. Connect the heat pipe to a vacuum system and reevacuate to remove any noncondensable other than hydrogen, e.g., argon.
6. Performance test while connected to vacuum system.

Each of the above operations resulted in some improvement in the heat pipe performance, but none completely accounted for the loss in output that occurred when the condenser end of the heat pipe was dropped below the evaporator end.

It is possible that the above operations would fail to remove a gas bubble trapped in the annulus between the wick and the wall. (The surface tension of the potassium in the pores of the wick could easily

serve to keep the gas trapped in the annulus.) With this in mind, the heat pipe was evacuated and all the potassium distilled off prior to flushing with clean potassium. The heat pipe was then completely filled with potassium, heated to 800°C (1472°F) for 66 hr, and drained. It was refilled with potassium, again heated to 800°C for an additional 20 hr, and again drained. A working charge of 75 g of potassium was then distilled into the heat pipe and the fill tube sealed shut.

A preliminary performance test of the heat pipe indicated that the unit still exhibited the same behavior when the condenser end of the heat pipe was positioned below the evaporator section. Therefore, the factors which determine the heat pipe performance from a theoretical standpoint were reviewed together with the many tests that had been conducted in an effort to diagnose the problem.

It now appears that the most likely source of the problem is the accumulation of impurities in the evaporator section of the heat pipe. When the potassium vapor condenses in the condenser section of the heat pipe, it tends to remove all the impurities from the metal surfaces in the condenser and transport them to the evaporator section. The buildup of oxides in the potassium lowers the surface tension, thereby reducing the capillary forces available to return the liquid to the evaporator. This has never before been a problem in ORNL potassium systems, but the surface-volume ratio is over 100 times greater for the liquid charge in a potassium heat pipe than in any of the other boiling potassium systems that we have operated. Thus the amount of oxygen picked up from surface oxide films is inherently two orders of magnitude greater in the heat pipes, and consequently the effects to be expected are correspondingly greater.

Appendix B

SHUNT REGULATOR DESIGN AND OPERATION

The external load and instrumentation connections to the thermoelectric generator are shown in Fig. 17 in the body of the report.

Lead-acid batteries are normally limited to about 2.4 V per cell at full charge. Since there were 12 cells in series across the generator output (a series-parallel arrangement of four 12-V automobile storage batteries comprised the battery bank), the maximum voltage across the bank had to be limited to about 28 V. Inasmuch as a thermoelectric generator should always be electrically loaded, a shunt regulator was required. This allowed us to preset the maximum voltage across the generator output terminals. When this level was reached the shunt regulator automatically connected an auxiliary load across the line and maintained the voltage at the preselected value. When the generator output fell below this level, the auxiliary load was disconnected and normal operation resumed.

The circuit for the shunt regulator (Fig. B.1) was provided by the Naval Nuclear Power Unit at Fort Belvoir, Virginia. This circuitry fired the output transistor when the preselected voltage was reached and maintained the firing until the voltage across the regulator was reduced to zero. The modifications to the output stage (Fig. B.2) were made by ORNL to allow the shunt to reopen when the voltage across the shunt fell just below the preset value.

No difficulty was experienced in the operation of the regulator during the course of the test.

Fig. B.1. Shunt regulator - Naval Nuclear Power Unit design.

Fig. B.2. ORNL-modified shunt regulator - Naval Nuclear Power Unit design.

Appendix C

EXPERIENCE AT 3M COMPANY

The 3M Company built and tested a thermoelectric module similar to the one tested at ORNL. The heat input to this module was supplied by an electric resistance heater.

A summary of the test data supplied to ORNL by the 3M Company indicates the following:

1. The test duration was 7076 hr.
2. The measured internal resistance increased from 2.28 Ω at the beginning of the test to 2.66 Ω near the end of the test.
3. The internal resistance due to contact resistance and lead and electrode resistance accounted for about 16% of the total internal resistance at the beginning of the test. During the test this resistance increased to the point where it amounted to 33% of the total internal resistance.

3M indicated that the electrical feed-throughs on the instrument header on their module also leaked, and that it was necessary to repressurize the system.

Although the module has not been analyzed, 3M stated that the increased internal resistance was likely associated with an increase in the electrode to thermoelectric material contact resistance. They also said that such an increase could be caused by a contaminating atmosphere. Thermal cycling, while mentioned as a possible contributor to the increase, was felt to be an unlikely cause of the trouble.

Internal Distribution

1. S. E. Beall	37. A. M. Perry
2. T. A. Butler	38. R. A. Robinson
3. W. B. Cottrell	39. M. W. Rosenthal
4. W. C. Cox	40. A. F. Rupp
5. F. L. Culler	41-46. G. Samuels
6. R. G. Donnelly	47. M. R. Sheldon
7-11. A. P. Fraas	48. A. C. Schaffhauser
12. J. H. Frye, Jr.	49. M. J. Skinner
13. J. H. Gillette	50. A. M. Smith
14. A. G. Grindell	51. I. Spiewak
15. K. W. Haff	52. D. A. Sundberg
16. H. W. Hoffman	53. D. B. Trauger
17. R. S. Holcomb	54. J. J. Tudor
18. P. R. Kasten	55. A. M. Weinberg
19. M. E. Lackey	56. G. D. Whitman
20. E. Lamb	57. J. V. Wilson
21. M. E. LaVerne	58. H. C. Young
22-31. D. B. Lloyd	59. Biology Library
32. M. I. Lundin	60-61. Central Research Library
33. R. N. Lyon	62. Y-12 Document Reference Section
34. R. E. MacPherson	63-87. Laboratory Records Department
35. H. C. McCurdy	88. Laboratory Records Department (RC)
36. A. J. Miller	

External Distribution

89. D. F. Cope, RDT Site Office, ORNL
90. W. M. Crim, Jr., Research and Technology Department, Building 322, Ft. Belvoir, Virginia 22060.
91. R. E. English, NASA, Lewis Research Center, Cleveland, Ohio 44135
92. E. E. Fowler, Division of Applied Technology, AEC, Washington, D.C. 20545
- 93-97. W. D. Holloman, Division of Reactor Development and Technology, AEC, Washington, D.C. 20545
- 98-99. M. Klein, Division of Space Nuclear Systems, AEC, Washington, D.C. 20545
100. J. J. Lynch, NASA Headquarters, Washington, D.C. 20546
101. S. V. Manson, NASA Headquarters, Washington, D.C. 20546
102. T. A. McLaughlin, Isotopic Auxiliary Power Branch, AEC, Washington, D.C. 20545
- 103-107. Nuclear Engineering Division, Naval Facilities Engineering Command, Washington, D.C. 20545
108. J. Pidkowitz, RDT Site Office, ORNL

- 109. W. B. Taylor, Technical Director, U.S. Army Mobility Equipment Research and Development Center, Ft. Belvoir, Virginia 22060
- 110. E. S. Wilson, Space Nuclear Systems, AEC, Germantown, Maryland 20767
- 111-113. Director, Division of Reactor Licensing, USAEC, Washington, D.C. 20545
- 114-115. Director, Division of Reactor Standards, USAEC, Washington, D.C. 20545
- 116-132. Manager, Technical Information Center, AEC (For ACRS Members)
- 133. Research and Technical Support Division, AEC, ORO
- 134-135. Technical Information Center, AEC

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