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A PROGRAM TO DESIGN AN ADVANCED TECHNOLOGY
THERMOELECTRIC MODULE FOR A 2 KW POWER
SYSTEM AND TO FABRICATE AND TEST A HEAT
PIPE/THERMOELECTRIC MODULE

JULY 29, 1970

MASTER

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Space and Defense Products Department
ELECTRICAL PRODUCTS GROUP

5 August 1970

In reply refer to: CA-1291

Union Carbide Corporation
Nuclear Division
P. O. Box M
Oak Ridge, Tennessee 37830

Attention: Mr. E. T. Henson
Contract Administrator
Purchasing Division

Subject: 3M Company Proposal for "A Program
to Design An Advanced Technology
Thermoelectric Module for a 2 KW
Power System and to Fabricate and
Test a Heat Pipe/Thermoelectric
Modules"

Gentlemen:

3M Company respectfully submits a firm fixed price
of \$64,945 to perform the workscope detailed in
the attached 3M proposal entitled "A Program to
Design An Advanced Technology Thermoelectric Module
for a 2 KW Power System and to Fabricate and Test
a Heat Pipe/Thermoelectric Modules", dated
29 July 1970.

This proposal is submitted as an offer to negotiate
any resulting contract and is valid for a period of
sixty (60) days.

If there are any questions, please contact Mr. E. C. Lund,
Contract Administrator, 612/633-9400.

Very truly yours,

W. L. Merrill
Contracts Manager

WLM:la

cc - Mr. A. Fraas - ORNL
LCDR W. Holloman - AEC/RET

A PROGRAM TO DESIGN AN ADVANCED TECHNOLOGY
THERMOELECTRIC MODULE FOR A 2 KW
POWER SYSTEM AND TO FABRICATE AND TEST A
HEAT PIPE/THERMOELECTRIC MODULE

Submitted to:

OAK RIDGE NATIONAL LABORATORY
OAK RIDGE, TENNESSEE 37830

July 29, 1970

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Submitted by:

SPACE AND DEFENSE PRODUCTS DEPARTMENT
MINNESOTA MINING AND MANUFACTURING COMPANY
ST. PAUL, MINNESOTA 55101

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SECTION I

INTRODUCTION AND SUMMARY

The Minnesota Mining and Manufacturing Company (3M Company) is pleased to submit this proposal to the Oak Ridge National Laboratories (ORNL) to prepare a reference design of a thermoelectric/heat pipe module using advanced technology (TPM-217) thermoelectric materials and to design, fabricate, test, and deliver a thermoelectric/heat pipe module for evaluation of the basic features of this combination using current state-of-the-art thermoelectric materials. The module design is to be consistent with the general objectives of the 2 kw(e) radioisotope power system program. Reference is made to ORNL draft report number TM-2959 entitled "Reference Design for Thermoelectric Isotope Power Unit Employing Heat Pipe Modules". The concept presented therein is one in which the heat produced by the radioisotope is transferred to a number of thermoelectric energy converters by means of heat pipes.

For large radioisotope power systems of this type, efficiency has a very significant importance. With a program schedule of starting the prototype power system phase in about 2-1/2 to 3 years, an advanced thermoelectric technology which is projected to be ready for application at that time period should be considered now for reference design and overall system evaluation. The TPM-217 type thermoelectric materials developed by 3M and evaluated under an ongoing AEC

program have high temperature operating capability, long-term stability, and significantly higher conversion efficiency than state-of-the-art materials. Theoretical thermoelectric conversion efficiency of about 13% is projected under temperature conditions of 1300°F at the hot junction and 150°F at the cold junction. A reasonable goal would be to develop a thermoelectric module with an overall efficiency in the range of 10-11%. The planned development of the TPM-217 technology will make this a realistic goal in 2-1/2 to 3 years hence.

It is proposed that a reference design be prepared based upon the TPM-217 thermoelectric materials. This can then be used to evaluate and project system performance and cost characteristics. The design will include a layout of the module based upon mutually agreed upon interface parameters, a list of components and materials, and thermal and electrical performance characteristics. The module design will be configured to accept state-of-the-art PbTe alloys as a backup. Performance projections will also be made for this backup design.

In order to test and evaluate the basic concept of a heat pipe/thermoelectric module assembly early in the present 2 kw(e) technology development program, it is proposed that a module which uses state-of-the-art SNAP-21/23A technology be designed and fabricated. The design and fabrication of a module based upon the segmented "N" type PbTe and segmented "P" type PbSnTe/BiSbTe thermoelectric materials of SNAP-21/23A can be

accomplished within a relatively short time period. The objective of testing the heat transfer characteristics of the thermoelectric module/heat pipe combination can be undertaken with a minimum of unknowns within the thermoelectric module.

A full-scale module in which an array of 12 modules will provide 2 kw(e) for 5 years is proposed. The following Table I summarizes the preliminary design characteristics of this module.

Table I

Thermoelectric Module Design Summary

N-Leg Materials: Segmented PbTe

P-Leg Materials: Segmented BiSbTe/PbSnTe

Hot Junction Temperature:

BOL (Beginning of Life): 1000°F

EOL (End of Life): 939°F

Power Output:

BOL: 202 watts at 24 volts

EOL: 169 watts at 24 volts

Cold Junction Temperature: 150°F

Heat Input BOL: 2375 watts

Efficiency BOL: 8.5%

Number of Couples: 208 (16 rows of 13 couples each)

Outer Diameter: 4 inches

Length: 15 inches

It is planned that this module will be assembled onto a heat pipe outer case onto which is brazed a nickel sleeve. This assembly is to be supplied by ORNL along with necessary electrical heating equipment for processing and test at 3M.

Although the nominal BOL design hot junction temperature is 1000°F, the module will be designed to be capable of operation up to 1300°F for short durations in order to evaluate the heat transfer characteristics comparable to that associated with a higher temperature TPM-217 module design. The PbTe materials with a melting point of 1691°F can be operated at this temperature level for short periods.

The following Section II discusses the module design, instrumentation, and recommended tests.

Section III outlines a six-month program to prepare an advanced technology module reference design, and to design, fabricate, and deliver a thermoelectric test module.

SECTION II

TECHNICAL DISCUSSIONS

A. Objective

The initial objective of this program is to prepare a reference design of a heat pipe/thermoelectric module based upon advanced technology TPM-217 type thermoelectric materials. Performance characteristics will be estimated in order to provide information to establish overall system features. The approach is to select the best potential thermoelectric technology which will be available for use at approximately the time when prototype system design and fabrication will begin. The development schedule for the TPM-217 materials fits into this time period of 2-1/2 to 3 years hence.

The second objective of this proposed program is to design and fabricate a thermoelectric module/heat pipe assembly for test and evaluation of the heat transfer and overall performance characteristics of this combination. A full-scale module is proposed which will use state-of-the-art SNAP technology. There is confidence in the state-of-the-art technology that this module will perform as predicted by the design, thus allowing one to evaluate other unknowns that may result due to the combination of the heat pipe and the thermoelectric module.

B. Design Objectives

The following reference data for a 2 kw(e) radioisotope power unit is used in the design of the thermoelectric module.

Life: 5 years

Power System Output: 2 kw(e) EOL

System Load Voltage: 48 volts

Fuel: SR90

Number of Modules: 12

Module Output Voltage: 12, 24, or 48 volts

Hot Junction Temperature: up to 1300°F

Environment: Sea Water at 40°F

C. Advanced Technology Module

1. Design Approach

The basic concept to the preparation of a reference design of an advanced technology thermoelectric module is that it will incorporate a cylindrical array of thermoelectric legs arranged around a heat pipe. It will be very similar to the test module discussed in detail in the following section "D".

The "P" type and "N" type TPM-217 materials will be used in this design. The "P" material is well characterized with much 3M developed data as well as that generated under the on-going AEC sponsored program which was initiated in January 1968. The "N" material is in the early stages of optimization and characterization. Performance predictions will be based upon the present properties of the "P" and "N" materials and upon a prediction of improvement in the "N" material characteristics as it is optimized to the extent that it is equivalent in performance to the present "P" material. This will bracket the expected final performance.

The selection of appropriate materials for electrodes and thermal insulation will be made on the basis of the presently existing information and engineering judgment. Methods of assembly and electrode-leg contacting will similarly be based upon present indications of anticipated acceptable techniques. A completely pressure contacted approach at both the hot and cold junctions similar to that presently planned for the test module may be a suitable contacting technique.

The hot junction temperature for the reference design will be assumed to be in the range of 1250° to 1300°F. The theoretical efficiency of the present "P" type TPM-217 in the temperature interval of from 1300° to 150°F is 12.8%. With the assumption that the "N" type material will also yield this efficiency, the overall module efficiency can be estimated. Electrical losses in electrodes and contacts of 10% can be assumed. As presented in the following sections, the thermal losses with an 850°F ΔT are about 7%. With the 1150°F ΔT of this reference design, the thermal losses will be in the range of 10-12%. The resultant module efficiency will then be in excess of 10%.

As a backup approach, the reference design will be configured to accept state-of-the-art PbTe technology. The hot junction temperature will be reduced to near 1000°F to assure long life based upon present test data. Electrode materials

and other components will be selected based upon past experience.

2. Background Information

An ongoing advanced technology program related to the TPM-217 materials at the 3M Company has been supported by the United States Atomic Energy Commission, Division of Space Nuclear Systems, since January 1968. This program, to evaluate the basic technology of these advanced thermoelectric materials, has as objectives to establish the chemical, physical, and mechanical properties and to generate long-term electrical and mechanical performance data. Electrical and thermal properties have been measured establishing the high efficiency of these materials. Mechanical tests have established the high strength characteristics of these materials.

Chemical compatibility tests are being conducted with thermal insulation and junction electrode materials. Long-term ingradient tests are being run with several having completed over 10,000 hours under various temperature and environmental conditions. Excellent long-term stability is being demonstrated. Based upon the point function property data, the following expected absolute optimum efficiencies for terrestrial applications have been calculated for the "P" type TPM-217 material:

<u>Temperature Interval</u>		<u>Optimum Absolute Efficiency</u>
<u>T_h (°C)</u>	<u>T_c (°C)</u>	
700	50	12.9
700	75	12.6
600	50	11.2
600	75	10.8
500	50	9.3
500	75	8.9

A majority of the development effort to date has been related to the "P" material. The "N" material is in early stages of optimization and characterization. It is anticipated to be at the same level of development as the "P" type by about the end of 1972. Total couple performance characteristics for long-term operation are expected by the end of 1973.

D. Test Module

1. Conceptual Design

The module concept shown in Figure 1 is basically similar to that shown in ORNL report TM-2959. It is a cylindrical array of thermoelectric legs arranged around a heat pipe. Conventional compression spring/follower construction is proposed. This structure can be enclosed within approximately the same overall envelope as that presented in the referenced report.

A one-inch diameter heat pipe is planned for this system. The desire for a somewhat larger diameter hot frame in order that the module will not be excessively long which could result

in significant axial expansion problems was expressed at a meeting between ORNL and 3M personnel on May 26, 1970. The concept of brazing a two-inch O.D. nickel sleeve to the heat pipe was proposed and is incorporated into the present design.

A flame-sprayed coating of alumina will be applied to the nickel sleeve to provide electrical insulation at the interface between the hot junction electrodes and the heat source. This can be ground to a smooth finish and allow the possibility of sliding to occur at this interface during thermal expansion. Experience has been gained in the use of this material in some of 3M's commercial development generators. Successful operation has been achieved under thermal cycling conditions indicating that this provides a suitable solution to this anticipated problem area.

The cold frame for this device will be formed of aluminum bars which contain the springs and followers. These will be hardcoated to form the required electrical insulation at the cold side.

The module is assembled by building up a row of thermoelectric couples on an aluminum cold frame follower bar. This subassembly is compressed against the alumina coated nickel sleeve. Assembly fixture rings at each end of the unit hold these subassemblies in position while the outer case is slipped into position. Release of the assembly fixture allows the follower bars to expand into proper position against the I.D.

of the outercase. This compressive force along with a coating of Dow Corning #340 heat transfer compound at this interface provides good thermal contact.

The lower conical hermetic seal is then welded in place. MIN-K 1301 powdered insulation is then installed. Output and instrumentation leads are connected to headers in the upper hermetic seal which is then welded in place. Subsequently, the module is processed and acceptance tested.

2. Thermoelectric Materials

Segmented thermoelectric legs are planned of the same materials as used in the SNAP-21/23A thermoelectric converters. The hot and cold segments of the "N" leg are 3N and 4N PbTe, respectively. The "P" leg hot and cold segments are 3P PbSnTe and 10P BiSbTe.

3. Cold Junction Temperature

With a seawater environment of 40°F and with the significant efficiency gains made at lower cold junction temperatures, every reasonable effort should be made to minimize the temperature drop from the cold junction to the seawater.

An estimate of the temperature drop from the seawater to the wall of the module well is 65°F with no fins. It is suggested that fins be incorporated to reduce this to about 30°F. The temperature drop through the wall of the well which is assumed to be 1/4 in. thick steel is 5°F.

The gap between the O.D. of the module outer case and the I.D. of the well requires careful study. A gas filled gap will result in a sizeable temperature drop. An expandable outer case is attractive from the standpoint of achieving good thermal contact. There is, however, some apprehension regarding any movement of the thermoelectric components after the module has undergone processing and initial testing. Further consideration will be given to this approach during the detailed design of the module.

An alternate technique may be to use a heat transfer compound such as Dow Corning #340 to reduce the temperature drop across this gap. This material has a thermal conductivity of .24 BTU/hr ft°F which is over an order of magnitude greater than a gas such as nitrogen. A reasonable objective would be to achieve a temperature drop of no more than 25°F across this gap.

From past test data, an estimate of the temperature drop from the cold junction to the exterior of the outer case is 30°F. With consideration given to the cost of the components in this cold end hardware and the tight tolerances required to achieve a low temperature drop, a 50°F value is allowed in these design calculations. The resultant cold junction design temperature is:

Seawater =	40°F
Seawater to Well ΔT =	30°F
Well Wall ΔT =	5°F
Well-Module Gap ΔT =	25°F
Cold Junction-Outer Case ΔT =	<u>50°F</u>
Cold Junction =	150°F

4. Hot Junction Temperature

The presently established design hot junction temperature for this test module is 1000°F. A capability of short-term operation (of the order of one or two days) at 1300°F will be a requirement of the design. Since test modules have been operated at 1250°F for over 2000 hours before serious degradation due to material sublimation has occurred, operation at 1300°F for short periods is anticipated to present no major material problems. Thus, data which can be applied to the advanced technology reference design can be obtained. The data obtained of temperature differences that occur in the various components and interfaces of the module will be applicable to the reference design. Any expected differences in heat flux between the two designs can be used to proportion the measured temperature drops to give appropriate values for the reference design.

5. Number of Couples and Leg Geometry

The desired output voltage of a module is such that a total of 48 volts can be obtained by connecting one or more in series with the load. A 24 volt module was selected based

upon past experience of the number of couples required and the leg geometry.

The following Table II lists the integrated properties of the "N" and "P" legs in the temperature gradient of 1000°F at the hot junction and 150°F at the cold junction.

Table II
Summary of Thermoelectric Leg Properties

Material	3N/4N	3P/10P
T_H	1000°F	1000°F
T_C	150°F	150°F
Open Circuit Voltage	113.4 mv	90.6mv
Load Voltage at Maximum Efficiency	64.2 mv	51.5 mv
I L/A (current & length to area ratio)	38.6	35.1 amp/inch
Q_s L/A (heat input & length to area ratio)	24.9	21.9 watt/inch
Lead and Contact Resistance	10%	10%
Efficiency	9.96%	8.25%

The load voltage per couple is 115.7 millivolts. To obtain 24 volts a total of 208 series connected couples are required. An array of 16 rows with 13 couples each was selected giving a total of 208 couples per module.

The end-of-life (5 years) power required per module is 167 watts. To allow for radioisotope decay and thermoelectric degradation, the beginning-of-life power level is estimated to be about 200 watts. At this power level, the current is 8.33 amperes with the load voltage at 24volts. With this current, one can now determine the leg length to area ratios.

From Table II it is observed that the optimum L/A for the "N" and "P" legs are approximately equal. Negligible sacrifice in efficiency results by making them equal. The legs then are of identical geometry and use common hardware. The resultant L/A ratio of the legs is 4.42 inches^{-1} .

With an available die of 0.281 inches in diameter, the leg length is 0.274 inches.

6. Thermal Losses

The thermal conductance of the MIN-K 1301 insulation is:

$$U_1 = \frac{2\pi KL (1-P.D.)}{\ln R_o/R_i}$$

Where:

PD = packing density = percentage of hot frame area
covered by thermoelectric material (34%)

K = .025 BTU/hr ft°F

L = 15 inches

R_o = 1.3 inch

R_i = 1.0 inch

U₁ = 0.145 watts/°F

The thermal conductance of the lower end hermetic seal is:

$$U_2 = \frac{2\pi K t}{\ln R_o/R_i}$$

t = thickness - 0.025 inches

K = 9.3 BTU/hr ft°F

R_o = 2 inches

R_i = 1 inch

$$U_2 = 0.052 \text{ watts/°F}$$

These factors are incorporated into the overall generator analysis computer code to determine the total heat input. The thermal losses are slightly over 7% of the heat input.

7. Performance as a Function of Time

A computerized program was used to predict the module performance as a function of time. This analytical tool takes into account the radioisotope decay as well as degradation in thermoelectric performance. With the module loaded at a constant 24 volts throughout life, as it might be in a battery charging application, the performance with time is as listed in Table III.

Table III
Module Performance as a
Function of Time

<u>Time</u>	<u>T_H</u>	<u>Power</u>	<u>Voltage</u>	<u>Heat Input</u>	<u>Efficiency</u>
0	1000°F	202 w	24 v	2375 w	8.50%
1 yr	987	196	24	2317	8.44
2	976	189	24	2260	8.36
3	964	182	24	2205	8.28
4	951	176	24	2151	8.18
5	939	169	24	2099	8.07

8. Instrumentation

Two major areas of interest in the evaluation of the module design are considered in selecting the points for instrumentation. The first is the heat transfer characteristics of the device. Two thermocouples will be installed within the nickel sleeve external to the hermetic seal. The outer case will be instrumented with two thermocouples external to the hermetic seal.

Within the hermetic seal, several thermocouples will be attached to various hot junction and cold junction electrodes. This will allow measurement of the temperature drop from the nickel sleeve to the hot junction electrodes and from the cold junction electrodes to the outer case. Temperature distribution within the module can be determined also.

The second aspect of concern is the effect upon couple performance due to axial expansion of the nickel sleeve. The thermoelectric couples at each end of two rows will be instrumented to measure their individual electrical performance. A comparison can then be made between couples which are subject to the maximum potential movement with those of minimum movement.

9. Testing

Following the processing of the module, the unit will be allowed to stabilize at its nominal BOL conditions of 1000°F and 150°F at the hot and cold junctions respectively. A performance map will be made as a function of load voltage. The various temperatures will be measured and the performance of individual instrumented couples will be determined.

The module thermal input will then be reduced to nominal BOL temperature conditions and the above data again recorded.

The module will be stabilized with a hot junction temperature of 1300°F. The various temperatures will be measured and electrical performance data will be obtained.

The unit will then be subjected to thermal cycles from room temperature to BOL temperature conditions and the performance measured. Four thermal cycles are planned. The rate of the thermal cycle will be approximately that to simulate the expected heat-up rate of the actual system.

SECTION III

WORK STATEMENT FOR THE PREPARATION OF AN ADVANCED TECHNOLOGY THERMOELECTRIC MODULE REFERENCE DESIGN AND FOR THE DESIGN, FABRICATION, TEST, AND DELIVERY OF A THERMOELECTRIC TEST MODULE

SCOPE

The 3M Company shall provide the necessary equipment, materials, and personnel to prepare a reference design of a thermoelectric module using high performance thermoelectric materials (TPM-217) and to design, fabricate, test, and deliver a state-of-the-art thermoelectric module for test and evaluation. The thermoelectric module design will incorporate heat pipe assembly as the hot frame structure of the module. An array of several of these heat pipe/thermoelectric module assemblies integrated with a large radioisotope heat source to yield two kilowatts of electric power is the ultimate goal. The reference design of the advanced technology module will provide information for overall system definition and evaluation. The test module will be designed and fabricated so that the heat transfer and electrical performance characteristics of this energy conversion concept can be tested and evaluated.

A six (6) month program is planned with the specific activities described below to be undertaken:

Task 1.0 Advanced Technology Module Reference Design

The 3M Company shall prepare a reference design of a thermoelectric module based upon using the TPM-217 type thermoelectric materials. The module design approach will be

consistent with the following objectives:

- System Power Output - 2 kw(E) EOL
- System Load Voltage - 48 volts
- Life - 5 years unattended
- Heat Sink - 40°F seawater during normal operation;
120°F still air during shipping
- Undersea Application at 6000 feet depth with
flexibility to go to 20,000 feet
- Isotopic Fuel - $^{90}\text{SrTiO}_3$
- Maintenance - field replacement of modules
- Number of Modules - 12 per system
- Heat Transfer System - Heat pipe

A cylindrical array of thermoelectric couples arranged around a cylindrical heat pipe is the basic concept planned. A layout will be prepared showing the constructional details of the module including a list of components and materials.

The design hot junction temperature planned is 1300°F. Performance characteristics will be predicted parametrically for hot junction temperatures in the range of 1000° to 1300°F.

The module design will be generally configured to accept state-of-the-art PbTe thermoelectric materials as a back-up design. Performance of the back-up design will be predicted with a hot junction temperature near 1000°F.

Task 2.0 Thermoelectric Test Module

2.1 Design

The 3M Company shall design a cylindrical thermoelectric test module with the following reference design objectives:

Thermoelectric Materials - PbTe, PbSnTe, and BiSbTe

Power - 200 watts at 24 volts

Hot Junction Temperature - 1000°F

Cold Junction Temperature - 150°F

Size - 4 inches diameter x 15 inches long

Instrumentation -

14 thermocouples internal

4 thermocouples external

The module will be designed to incorporate a customer furnished heat pipe/nickel sleeve assembly as the hot frame. The heat pipe/nickel sleeve assembly is to be supplied in a mutually agreed upon configuration to interface directly with 3M fabricated thermoelectric hardware.

2.2 Fabrication

One (1) thermoelectric module designed above will be fabricated, assembled, and processed with the goal of achieving the above stated design objectives. To accomplish the processing phase, a customer supplied electrical heating system for the heat pipe will be utilized.

2.3 Module Testing

The following tests will be conducted on the module prior to delivery:

a) Nominal BOL steady-state conditions with $T_H = 1000^\circ\text{F}$, $T_C = 150^\circ\text{F}$, and $V_{\text{Load}} = 24$ volts.

b) Performance map as a function of load voltage with a fixed heat input as established in (a) above.

c) Nominal EOL temperature conditions will be established and the module performance measured.

d) Elevated temperature conditions (1300°F hot junction) will be established and the module performance characteristics measured.

e) Thermal cycling tests from room temperature to full BOL temperatures will be conducted. Performance will be measured after each of four cycles.
(The total planned duration for the above module testing is 4 weeks.)

2.4 Liaison

Liaison will be conducted to assure proper definition of the interfaces of the module hardware with the heat pipe and other related test equipment.

DELIVERABLE ITEMS

- One Thermoelectric Module - 6 months ARO
- Reference Design Report - 2-1/2 months ARO

CUSTOMER FURNISHED EQUIPMENT

- Heat pipe with nickel sleeve brazed in place.
- Electrical heater system for the heat pipe.

90 days prior
to module
delivery