**NUCLEAR BATTERY - THERMOCOUPLE TYPE
THIRD QUARTERLY REPORT**

July 1, 1957 to September 30, 1957

U. S. Army Signal Corps Contract No. R-50-799965-SC-01-91

**U. S. Army Signal Engineering Laboratories Technical
Requirement No. P.R. and C. 57 ELS/D 3704**

U. S. Department of the Army Project No. 3-99-09-00

**U. S. Army Signal Engineering Laboratories
Project No. 99-15-102**

**U. S. Army Signal Corps Engineering Laboratories
Ft. Monmouth, N. J.**

MOUND LABORATORY

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**NUCLEAR BATTERY - THERMOCOUPLE TYPE
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The object of this contract is to conduct research on radioactive materials and thermocouples suitable for the direct conversion of heat to electrical energy.

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U. S. D. A. Project No. 3-99-09-00

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Date: 30 September 1957

Bertram C. Blanke

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PURPOSE

Radioactive materials and thermocouples suitable for use in a system for the direct conversion of heat to electrical energy will be investigated.

Isotopes having suitable characteristics will be determined, heat sources will be designed, and a prototype battery will be constructed.

ABSTRACT

A second thermoelectric generator consisting of unitized rings of Chromel-Constantan thermopiles was designed and constructed. The generator was first assembled with three rings having a total of 122 junctions and was tested with an electrical heater. Another ring of 39 junctions was added, and the tests were repeated. The final generator (4 rings and 161 junctions) had a maximum efficiency of 0.67 per cent. At a power input of 17 watts, the temperature difference between hot and cold junctions was 400°C, and the efficiency was 0.56 per cent. The theoretical maximum efficiency at this temperature difference and with this type of thermocouple is 0.83 per cent.

REPORTS AND CONFERENCES

On July 30, 1957 a conference was held at Mound Laboratory between Mr. George Hunrath and Dr. Emil Kittl of U. S. Army Signal Engineering Laboratories, and Mound Laboratory personnel assigned to the project. Current progress was reviewed, and current and future thermoelectric generator designs were discussed. It was agreed that a complete discussion of power source feasibility would be included in the third quarterly report or the final report for each isotope selected at power levels of 1, 10, 100, 1000 and 10,000 watts. Sources of the isotopes, reactor space required, radiation hazards involved, shielding necessary, present and future separation costs, and a rating system for the isotopes were to be reported on.

On August 13 a conference was held between B. C. Blanke of Mound Laboratory and Mr. Arthur Rupp, Assistant Director, X-10 site, Oak Ridge National Laboratory, Oak Ridge, Tennessee. The availability of fission isotopes listed in the first quarterly report (MLM-CF-57-4-34) as having proper nuclear properties for heat sources was discussed. Oak Ridge is constructing a new fission product separation facility. However, use of most of the desirable isotopes to be separated has been committed. Mr. Rupp was of the opinion that strontium-90 and strontium-89 could be processed at Oak Ridge National Laboratory if they could be made available from the MTR reactor program. Roughly 100,000 curies could be processed as glass pellets of strontium fluoride and 16,000 curies could be sealed into a stainless steel tube 7/8-inch in diameter and eight inches long. He suggested that Mound Laboratory use him for liaison if further reactor information be required.

On September 13, a conference was held at Ft. Monmouth between B. C. Blanke of Mound Laboratory and Mr. George Hunrath and Dr. Emil Kittl. Results obtained with a thermoelectric generator built at Mound Laboratory were discussed. In view of the lack of facilities for metallurgical study at Mound Laboratory, it was decided that only commercially-available materials will be investigated in this contract. Arrangements were made for Mound Laboratory's use of Franklin Institute reports on production of thermocouple alloys under U. S. Army Signal Corps contract.

FACTUAL DATA

A second thermoelectric generator has been designed and constructed. This device consists of a stack of thermopiles mounted one above the other. Each thermopile consists of about 40 thermocouples in series arranged like the spokes of a wheel, as shown in Figure 1. A thin polystyrene ring forms the hub during construction and is dissolved out prior to final assembly. This method permits testing of each thermopile before assembly of the generator.

The thermocouples are spot-welded. B and S #28 Chromel and #30 Constantan wires formed to a U shape with the junction at the bottom of the U. The junction is deburred by grinding. The thermocouples are slipped on the polystyrene ring with the loose ends radiating from the center. Jigs are used to hold the thermocouples in place while the cold junctions are formed. After silver soldering, the cold junctions are confined between Teflon rings and are insulated by mica pressed between aluminum rings.

The wheels are shaped so that all the hot junctions can be assembled over a thin-walled nickel cylinder $\frac{1}{4}$ -inch in diameter and $\frac{5}{16}$ -inch long. As the rings supporting the cold junctions of each wheel total $\frac{1}{8}$ -inch in thickness, the length and angle of the leads from the cold to the hot junctions vary, and are preformed in the jig before silver soldering the cold junctions. A very light coat of #7 Sauereisen cement is placed on the hot junctions after electrical checks show no shorts. After the cement dries, the polystyrene ring is dissolved in benzene.

Three such wheels were assembled into a thermoelectric generator having a total of 122 junction pairs. The generator was placed in a close-fitting brass cylinder closed at the bottom by a brass plate. A small electric heater, shown in Figure 2, having potential leads for measurement of the voltage drop across the winding was constructed to simulate the radioactive heat source. It consisted of a $\frac{1}{4}$ -inch Lavite core cut to hold nine turns of helically-wound, 0.004-inch Tophet A wire having a total resistance of 180 ohms. A Chromel-Constantan thermocouple was mounted in the center of the core. This core was fastened to a three-inch piece of ceramic spaghetti which was supported by a small tube socket. The space between the brass jacket and the rings, and between the hot and cold junctions was filled with Santocel insulation. Leads from the generator, thermocouple, and heater were brought out through a Formica plug.

This entire assembly was then inserted in a close-fitting brass jacket immersed to a depth of about 15 inches in a calorimeter water bath regulated to 0.0005°C . Other calorimetry instrumentation used were Willard low discharge batteries, a heat control panel, a Wenner potentiometer, and a type E galvanometer. The generator and heater can be considered a calorimeter. The accuracy of the calculated power (voltage $\times$ current) supplied to the heater was determined to be 0.5 per cent by comparing the generator output using a polonium-210 source of known power with the equivalent power supplied to the heater.

The values given for runs 1 through 8 in Table 1 were obtained by supplying a steady current to the heater until temperature equilibrium was attained with no load across the generator. The potential across the heater was measured to give the correct power input (W_s). The output of the thermocouple inside the heater (with the reference junction at 0°C) and the battery output (E_0) were then measured. The generator was then loaded by means of a Leeds and Northrup four decade resistance box. After a new temperature equilibrium was established, the voltage across the resistance box was measured, and the power to the load was calculated. This was repeated for about five different loads, and from a plot of W vs R , the maximum power and internal resistance of the

generator were determined (the maximum power occurs when the load and the internal resistance are nearly equal). All temperatures were calculated from calibration curves of the thermocouple (Figure 4). The calibration curves were obtained on two sets of junctions made from the same wires and spot welded by the same techniques as were used in the junctions in the generator. These junctions were calibrated in the sample space of a high-temperature calorimeter which had previously been calibrated by a primary standard platinum resistance thermometer and a platinum/platinum-rhodium junction. The temperature differences between hot and cold junctions ($T_c - T_s$) were calculated in runs 1 through 12 from E_c/n , ($n=122$).

In run 9 the entire generator was evacuated. The lower efficiency (0.392 per cent vs 0.457 per cent at the same power level at atmospheric pressure) can be attributed to a larger increase in radiation losses because of the higher operating temperatures, or to the increase in thermal resistance between the cold junctions and the exterior wall. Conduction losses should have decreased.

The generator was then removed from the water bath and was operated at the following three different ambient temperatures: 0°C (run 10) -13.2°C (run 11) and 200°C (run 12).

A fourth ring having thirty-nine junctions was added to the generator. This generator, with the electrical heat source in place, is shown in Figure 3. To obtain directly the hot and cold junction temperatures, thermocouples were added by extending both wires of one cold junction and a single wire of the adjacent hot junction. T_c and T_s could then be determined without taking the junctions out of the generator circuit. However, only T_s could be measured under load conditions. By quickly measuring T_c after removing a load and obtaining a curve of T_c vs time on semi-log paper, the temperature under load conditions was obtained for several runs. Because of thermal conduction of the heat away from the junction by the leads, it was thought that the temperatures of these junctions might not be representative of the other junctions. However, $T_c - T_s$ values for these junctions were within a few per cent of the temperatures obtained from E_c/n , ($n=161$). Runs 13 through 19 in Table 1 give the results of the tests with 161 junctions and give the values of T_c and T_s .

In Figure 5 the efficiency of the second generator is plotted vs the heater input in watts for both three and four rings. If there were no differences in per cent loss of heat, and if the thermal resistances were independent of temperature, the efficiency at the same power input would be expected to be 1.33 times greater for three rings than for four rings. The ratio of efficiency between the two generators starts near 1.25 at 2 watts, decreases to 1.13 at 10 watts, and falls to 1.05 at 17 watts.

In Figure 6, a more useful plot of efficiency is presented. Here the efficiency is plotted vs the temperature difference between the hot and cold junctions. At the same per cent loss of heat, the efficiency for any given $T_c - T_s$ should be entirely independent of the number of junctions. Since the efficiency in Figure 6 is greater for four rings than for three rings at all values of $T_c - T_s$, the per cent loss of heat is less for four rings at all temperatures. This was not entirely unexpected because the difference between the temperature of the heat source and the hot junctions would be larger for four rings even though the temperature of the heat source would be lower. From calculation, it appears that most of the heat loss occurs from the surface of the wires and not directly from the heater and surface of the source container. The relative surface areas are three square centimeters for the sample container and 56 square centimeters for the wires on the 161-junction generator.

The generators were very stable. No noise could be detected in the fifth decimal place once the loads and currents were adjusted.

Table 2 shows the measured parameters as well as some of the calculated parameters for Generator 2 with 161 junctions and a power input of 16.42 watts. The thermal resistance can be considered tentative to the extent of 10 per cent. The magnitude of thermal resistance from the cold junctions to the bath (about $4^{\circ}\text{C-watt}^{-1}$) is larger than expected and deserves more attention in future designs.

CONCLUSIONS

The second thermoelectric generator design has been successfully built and tested. Results show that a design is available for a battery which can supply a voltage of over six volts and a current of 80 milliamperes with a power input of about 16 watts.

PROGRAM FOR NEXT INTERVAL

The program for the next interval is to complete the evaluation of isotopes and the hazards associated with the production of heat sources of various sizes.

Designs of thermoelectric generators will be reviewed in order to obtain one of optimum efficiency based on data collected, and a final model will be constructed.

IDENTIFICATION OF KEY PERSONNEL

Bertram C. Blanke (285 hours)

Kenneth C. Jordan (480 hours)

John H. Birden (528 hours)

Elton L. Murphy (528 hours) was added to the program. He served with the U. S. Army Air Corps (ETO) from January, 1943 to October, 1945. After his discharge, he worked about two years for the Vall Decker Company, Piqua, Ohio before attending Ohio Northern University where he was graduated in 1951 with a B.A. in chemistry. He joined Mound Laboratory in October, 1951 and has worked on physical properties of high-temperature materials and on radium and actinium processing.

Figure 1. Wheel Thermopile

Figure 2. Heater

Figure 3. Thermoelectric Generator No. 2
161 Junctions; Heat Source in Place

Figure 4. Thermoelectric Power of Chromel - Constantan Junctions Used in Generator No. 2

Figure 5. Efficiency of Generator No. 2 vs Electric Power Input

Figure 6. Efficiency of Generator No. 2 vs Temperature Difference Between Hot and Cold Junctions

Table 1
PERFORMANCE OF GENERATOR NO. 2

Run	W_s Heater Power watts	T_s Temperature Inside Heater °C	T_1 Hot Junction Temperature °C	T_2 Cold Junction Temperature °C	$T_1 - T_2$	T_a Ambient Temp. °C	E_o Generator Voltage with No Load volts	W_L Maximum Power To Load milliwatts**	Eff. Efficiency %
1	1.149	125.7	(see note)	(see note)	48.5	25.9	0.3625	0.817	0.0711
2	2.383	189.4			189.4	25.9	0.7511	3.438	0.1443
3	3.605	269.9			142.2	25.9	1.1220	7.601	0.2108
4	5.013	384.2			188.8	25.9	1.5291	14.06	0.2805
5	5.085	408.8			189.5	25.9	1.5408	14.17	0.2786
6	9.814	657.8			323.2	25.9	2.7716	44.80	0.4565
7	15.24	838.6			446.0	25.9	3.9441	89.72	0.5887
8	18.27	904.0			503.2	25.9	4.4895	115.96	0.6347
9	9.856*	832.0			305.1	25.9	2.5841	38.67	0.392*
10	9.847	643.5	31.73	29.88	355.3	0.0	2.7852	45.62	0.463
11	9.851	640.5			330.3	- 13.2	2.8074	46.43	0.471
12	9.851	727.0			270.3	200.0	2.3682	32.15	0.326
13	0.0523	34.02			(see note)	29.52	0.016952	0.00135	0.0026
14	1.7337	157.0			35.9	29.52	0.56379	1.4990	0.0865
15	4.872	340.8			45.6	29.52	1.5570	11.436	0.2347
16	10.599	595.4			62.0	29.52	3.1920	45.35	0.4279
17	16.416	779.4			77.0	29.31	4.5986	93.48	0.5694
18	19.789	854.9			85.5	29.31	5.3258	124.60	0.6296
19	22.450	911.0	566.5	91.3		29.31	5.8448	149.40	0.6655

* Air evacuated from generator.

** Voltage can be approximated by $(E_o/2 \times 0.99)$

Note. In runs 1 through 12, generator had 122 junctions.

In runs 13 through 19, generator had 161 junctions.

Table 2

CHARACTERISTICS OF GENERATOR NO. 2 WITH 161 JUNCTIONS

W_s (heater input)	16.42 watts
T_a (ambient temperature)	$\sim 29.3^\circ\text{C}$
T_c (cold junction temperature, no load)	$\sim 77.0^\circ\text{C}$
T_h (hot junction temperature, no load)	$\sim 465.6^\circ\text{C}$
T_s (temperature inside heater, no load)	$\sim 779.0^\circ\text{C}$
E_o (generator voltage, no load)	~ 4.6000 volts
R_i (internal resistance of generator)	55.5 ohms
R_L (external load)	~ 55.5 ohms
E_L (voltage across external load)	~ 2.2778 volts
W_L (power dissipated in external load, E_L^2/R_L)	~ 0.09348 watts
L (average length of wire, hot to cold junction)	~ 1.90 cm
R_p (thermal resistance, hot to cold junctions, calculated, assuming no loss)	$\sim 33.2^\circ\text{C-watt}^{-1}$
R_s (thermal resistance to heat losses, (thermal resistance to heat loss by radiation)	$\sim 82^\circ\text{C-watt}^{-1}$ 164 $^\circ\text{C-watt}^{-1}$
(thermal resistance to heat loss by conduction)	$\sim 164^\circ\text{C-watt}^{-1}$
(thermal resistance, cold junctions to bath)	4.0 $^\circ\text{C-watt}^{-1}$
(thermal resistance, heater to hot junctions)	19.1 $^\circ\text{C-watt}^{-1}$
f (ratio of R_i/R_s)	0.40
f/f_1 (fraction of W_s lost)	0.29
β (thermoelectric power $E_o/n(T_c - T_s)$)	$\sim 73.4 \mu\text{ volts-}^\circ\text{C}^{-1}$
Eff (over-all efficiency $100 W_L/W_s$)	0.569 %
Eff (theoretical maximum efficiency $0.569/(1-f)$)	0.797%