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THERMAL BATTERIES USING POLONIUM-210
(Information Report)

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(Information Report)

BY

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ABSTRACT

The general theory of thermal batteries of the thermopile type is developed and this theory is applied to the design of batteries powered by Po^{210} heat sources. The physical characteristics, performance, and fabrication of two experimental batteries with 57 and 146 curies, respectively, of Po^{210} are described. Maximum electrical power delivered to a load was 0.1 and 0.2 per cent of the power developed as heat by the polonium. The work capacity of the thermal batteries is 1.8 and 9.2 times, respectively, that of a commercial mercury battery (RM4Z) and the weight is 30 per cent less.

INTRODUCTION

Thermal batteries using a thermopile (more than one thermocouple) to convert heat to electricity were investigated many years ago and abandoned as economical means of generating commercial amounts of electrical power. However, such batteries are used extensively in many special applications.

When powered by radioactive substances these batteries become even less economical, but they may have special applications as a result of the high specific amounts of energy released by all radioactive materials. The rate at which this energy is released, and consequently the life of the battery, is determined by the half-life of the radioactive material which can not be changed by physical conditions.

To illustrate the amount of power developed by radioactive substances, the theoretical temperature which is attained by a solid sphere of polonium-210 in a vacuum can be calculated. The density of polonium-210 is about 9 grams per cubic centimeter, the specific activity is 2.2×10^{-4} grams per curie, and the power generated is .032 watt per curie. A solid sphere 0.1 inch in diameter could contain 350 curies of polonium-210 which will generate 11.3 watts of heat. If such a sphere were suspended in an evacuated chamber its surface (assuming an emissivity of 0.25) would reach a temperature of $2,200^{\circ}\text{C}$. Polonium must be hermetically sealed in very strong and dense capsules, consequently the surface temperature of the capsules will be much cooler than the temperatures calculated above.

Thermal batteries consist of a quantity of polonium or other radioactive material contained in a capsule which is thermally insulated from and positioned within a secondary container of about three times the dimensions of the capsule. The hot junctions of thermocouples with high thermoelectric power are thermally, but not electrically, connected to the capsule; and the cold junctions are connected in the same way to the outer container. The thermocouples are electrically connected in series on the surface of the outer container. After a time the temperatures of the system reach a steady state with a difference of temperature existing between hot and cold junctions, which temperature difference causes a voltage proportional to the temperature difference to be generated in the thermocouples.

NOMENCLATURE

a, a_1, a_2 - radius of wire and of each type of wire, cm

β - thermoelectric power of each thermocouple, volts/ $^{\circ}\text{C}$

e - voltage generated per couple, volts

f - ratio of R_t to $(R_s + R_c)$

H - surface radiation from wires, $\text{cal}/(\text{sec})(^\circ\text{C})(\text{sq cm})$

$\rho_t, \rho_{t,1}, \rho_{t,2}$ - thermal resistivity (inverse of conductivity) of wire and of each type of wire, $(^\circ\text{C})(\text{sq cm})/(\text{cal/sec})(\text{cm})$

l - length of each lead (distance between hot and cold junctions), cm

m - ratio of R_c to R_s

n - number of hot (or cold) junctions

q - lateral loss of heat per lead, cal/sec

$\rho_e, \rho_{e,1}, \rho_{e,2}$ - electrical resistivity of wire and of each type of wire, $(\text{ohms})(\text{sq cm})/\text{cm}$

R_c - thermal resistance from capsule to hot junctions, $(^\circ\text{C})(\text{sec})/\text{cal}$

R_t - thermal resistance of all the leads from hot to cold junctions, $(^\circ\text{C})(\text{sec})/\text{cal}$

R_s - thermal resistance of the space between capsule and outer container, $(^\circ\text{C})(\text{sec})/\text{cal}$

r - series resistance of both leads forming a junction, ohms

r_1, r_2 - resistance per lead of each type of wire, ohms

T - absolute temperature, $^\circ\text{K}$

T_1, T_2 - temperature of hot and cold junctions, $^\circ\text{K}$

$\Delta T - (T_1 - T_2), ^\circ\text{C}$

W_L - power dissipated in load, watts

W_S - heat generated by radioactivity within the capsule, watts

THEORY OF THE THERMAL BATTERY

Equations for the efficiency of the thermal battery can be developed and the final equation (Eq 17) will be one in terms of the absolute temperatures of the hot and cold junctions.

Figure 1 is a diagram of the electrical circuit of a thermal battery and Figure 2 is an equivalent circuit diagram of the thermal circuit.

Maximum power will be developed with a load resistance equal to the internal resistance of the battery, Figure 1:

$$W_L = \frac{(ne)^2}{4nr} = \frac{ne^2}{4r} \quad \text{watts} \quad (1)$$

The efficiency is:

$$\text{Eff} = \frac{100 W_L}{W_S} = \frac{25 ne^2}{W_S r} \quad \text{per cent} \quad (2)$$

5742

7

fig 1

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ELECTRICAL CIRCUIT

FIGURE 1

THERMAL CIRCUIT

5743

FIGURE 2

The voltage generated by each thermocouple is:

$$e = \beta (T_1 - T_2) = \beta \Delta T \quad \text{volts} \quad (3)$$

In the steady state the thermal circuit, Figure 2, can be represented by R_s in parallel with R_c and R_t connected in series. All of the heat from the source less that converted to electrical power ($W_s - 2 W_L$) flows through the parallel network. The temperature difference ΔT is measured across R_t and is:

$$\Delta T = \frac{R_t R_s (W_s - 2 W_L)}{4.185 (R_t + R_s + R_c)} \quad ^\circ\text{C} \quad (4)$$

Since W_L is small in comparison to W_s , Equation (4) can be adequately represented by:

$$\Delta T = \frac{R_t R_s W_s}{4.185 (R_t + R_s + R_c)} \quad ^\circ\text{C} \quad (5)$$

Substituting Equations (3) and (5) in Equation (2):

$$\text{Eff} = \frac{25 \pi \beta^2 R_t^2 R_s^2 W_s}{(4.185)^2 (R_t + R_s + R_c)^2 r} \quad \text{per cent} \quad (6)$$

Electrical and thermal resistivities are related by the Wiedemann-Franz law:

$$\frac{\rho_e}{\rho_t T} = \text{a constant} = \alpha \quad (\text{cal/sec})/(\text{}^\circ\text{C})(\text{ohms})(\text{}^\circ\text{C}) \quad (7)$$

The constant α has a value of about 5.5×10^{-9} for all pure metals and is usually higher for alloys, especially for alloys of higher electrical resistivity. It has a value of about 8×10^{-9} for constantan and about 10×10^{-9} for chromel. (As will be evident below, the increase in efficiency resulting from the greater thermal power of chromel-constantan over iron-constantan is partially cancelled by the greater Wiedemann-Franz constant of chromel.) The electrical resistance per lead of each type of wire is:

$$r_1 = \frac{\rho_{e,1} l}{\pi a_1^2}; \quad r_2 = \frac{\rho_{e,2} l}{\pi a_2^2} \quad \text{ohms} \quad (8)$$

The diameter of each type of wire should be chosen such that the thermal resistance of each lead is the same

$$r_t = \frac{\rho_{t,1} l}{\pi a_1^2} = \frac{\rho_{t,2} l}{\pi a_2^2} \quad (9)$$

Dividing Equations (7) and (8) by Equation (9) and making use of Equation (7) gives:

$$\begin{aligned} r_1 &= r_t \alpha_1 \left(\frac{T_1 + T_2}{2} \right) \\ r_2 &= r_t \alpha_2 \left(\frac{T_1 + T_2}{2} \right) \end{aligned} \quad (10)$$

Adding both parts of Equation (10) gives:

$$r = r_1 + r_2 = r_t (\alpha_1 + \alpha_2) \left(\frac{T_1 + T_2}{2} \right) \quad (11)$$

Since all of the leads are in parallel thermally,

$$r_t = 2 n R_t$$

which, substituted in Equation (11), gives:

$$r = n R_t (\alpha_1 + \alpha_2)(T_1 + T_2) \quad (12)$$

Solving Equation (12) for R_t and substituting for one R_t in the numerator of Equation (6) gives:

$$\text{Eff} = \frac{1.43 \beta^2 R_s^2 R_t}{(\alpha_1 + \alpha_2)(R_t + R_s + R_c)^2(T_1 + T_2)} \quad (13)$$

The relative magnitude of the thermal resistance terms can be expressed by:

$$R_c = m R_s \quad (14)$$

$$R_t = f (R_s + R_c) = f R_s (1 + m) \quad (15)$$

Substituting Equations (14) and (15) in Equation (13) gives:

$$\text{Eff} = \frac{1.43 \beta^2 R_s W_s}{(\alpha_1 + \alpha_2)(1 + m)(T_1 + T_2)} \cdot \frac{f}{(f + 1)^2} \quad \text{per cent} \quad (16)$$

The term $f/(f + 1)^2$ has a maximum of .25 for $f = 1$, which means that for maximum efficiency the leads should have a thermal resistance equal to $R_s + R_c$. If this condition is always satisfied:

$$T_1 - T_2 = \Delta T = \frac{W_s R_s}{2 \times 4.185} \quad (17)$$

Substituting Equation (17) in Equation (16) and letting

$$f/(f+1)^2 = .25$$

gives the maximum efficiency:

$$\text{Eff}_{(\max)} = \frac{3.0 \beta^2}{(\alpha_1 + \alpha_2)(1+m)} \frac{T_1 - T_2}{T_1 + T_2} \quad \text{per cent} \quad (18)$$

Several general statements can now be made concerning the design of thermal batteries and similar devices:

1. When the hot junctions are in good thermal contact with the source of heat ($m \rightarrow 0$), the maximum efficiency is obtained when one-half of the heat flows through the leads.
2. Like all heat engines, the efficiency of a thermal battery is dependent upon absolute temperatures (Eq 18).
3. The efficiency is independent of the number of junctions. However, from practical considerations, the thermal resistance between capsule and hot junctions tends to increase slightly with the number of junctions which increases the term $(1+m)$, and so decreases the efficiency. Metallic contact between capsule and junction is possible for a single junction ($m \rightarrow 0$).
4. The internal resistance of the battery increases as the square of the number of junctions (Eq 12).
5. The voltage of the battery is proportional to the number of junctions.

Another factor which tends to lower the efficiency and has not been included in the derivation of Equations (16) and (18) is the lateral loss of heat by radiation along the wires. This loss of heat has the same effect as an increase in the Wiedemann-Franz constant. As a first approximation the loss of heat per lead is:

$$q \approx \pi a H l (T_1 - T_2) \quad \text{cal/sec} \quad (19)$$

Where

q - is the lateral loss of heat along each lead, cal/sec

a - is the radius of each lead, cm

H - is the surface radiation, cal/(sec)(°C)(sq cm)

l - is the length of the leads, cm

The total lateral loss of heat can be included by assuming a thermal resistance of $1/\pi n a H l$ in parallel with all of the leads of each type of wire, and therefore the adjusted Wiedemann-Franz constant for each type of wire is:

$$\alpha_{(\text{adj})} = \alpha (1 + \pi n a H l R_t) (\text{cal/sec})/(\text{°C})(\text{ohms})(\text{°C}) \quad (20)$$

When the capsule size, the insulating material, dimensions of outer container, and type of wire have been determined, all of the variables in Equation (20) are fixed except n and a . Since a is inversely proportional to the square root of n , Equation (19) can be written:

$$\alpha_{(adj)} = \alpha (1 + K\sqrt{n}) \quad (21)$$

This is another reason for a decrease in efficiency with an increase in the number of junctions. With a fixed number of junctions the radius of the wire changes with the Wiedemann-Franz constant and with the resistivity. The relative radii of the two wires can be determined by substituting Equation (8) in Equation (10):

$$\frac{a_1}{a_2} = \sqrt{\frac{\alpha_2 \rho_{e,1}}{\alpha_1 \rho_{e,2}}} \quad (22)$$

Still another loss of efficiency might occur if the cold junctions were somewhat hotter than the outside container. This can be minimized, without introducing other losses in efficiency, by using cold junctions having a large surface area.

If R_s were very large, a condition which could be realized by placing the radioactivity within the junctions, the efficiency with the same working temperatures would be twice as great and $m = 0$:

$$\text{Eff} = \frac{6.0 \beta^2 (T_1 - T_2)}{(\alpha_1 + \alpha_2)(T_1 + T_2)} \quad \text{per cent} \quad (23)$$

Also the adjusted Wiedemann-Franz constant as given by Equation (20) would apply to Equation (23) and give a more exact statement of the efficiency. Such a junction must be fabricated in order to determine the effect of the activity upon the thermoelectric power and also upon the life of the junction.

The maximum theoretical efficiency may be readily calculated from Equation (23). Thus for $T_1 \gg T_2$, the maximum theoretical efficiency is:

$$\text{Eff} = \frac{6 \beta^2}{\alpha_1 + \alpha_2} \quad \text{per cent} \quad (24)$$

For bismuth-antimony couples

$$\beta = 12 \times 10^{-5}$$

$$\alpha_1 = 7.7 \times 10^{-9}$$

$$\alpha_2 = 5.8 \times 10^{-9}$$

the maximum theoretical efficiency would be 6.4 per cent. An efficiency of this magnitude probably will never be obtained.

DESIGN OF THERMAL BATTERIES

In the design of a thermal battery perhaps the first factor to be determined is the type of thermocouple which will be used. Because the efficiency is proportional to the square of the thermoelectric power, this will be the most important factor. However, other factors must be considered, such as melting points, welding or soldering properties, Wiedemann-Franz constant, and resistivity. Of the commercial varieties of thermocouple materials, chromel-constantan and iron-constantan have the largest thermoelectric power and are satisfactory in all other respects. Bismuth-antimony couples have an appreciably higher thermoelectric power, but are difficult to fabricate and their melting points are low. Other thermocouples have been developed recently with reportedly higher thermoelectric power but no definite information concerning them is available.

The relative merits of chromel-constantan and iron-constantan couples may be compared. The thermoelectric power of these couples increases with temperature and at 300°C chromel-constantan has a value of $\beta = 7.7 \times 10^{-5}$ volts per degree C and iron-constantan has a value of $\beta = 5.5 \times 10^{-5}$ volts per degree C. The Wiedemann-Franz constants are 5.5×10^{-9} , 8×10^{-9} , and 10×10^{-9} (cal/sec)/(°C)(ohm)(°C) respectively for iron, constantan, and chromel. From Equation (16) or (18) the efficiency of chromel-constantan relative to iron-constantan is:

$$\left(\frac{7.7}{5.5}\right)^2 \left(\frac{5.5 + 8}{8 + 10}\right) = 1.5$$

The electrical resistivities are 10×10^{-8} , 44×10^{-8} , and 65×10^{-8} (ohm)(cm) respectively, for iron, constantan, and chromel. The radius of a chromel wire relative to an iron wire is, from Equation (22);

$$\sqrt{\frac{5.5 \times 65}{10 \times 10}} = 1.9$$

The larger chromel wire will increase the adjusted Wiedemann-Franz constant given by Equation (20) by an amount depending on the number of junctions. This will further reduce the relative efficiency of the two types of thermocouples. However in most cases the efficiency of the chromel-constantan junctions will be higher than the corresponding iron-constantan couples.

CHARACTERISTICS OF EXPERIMENTAL BATTERIES

Two experimental batteries were constructed to check the validity of the theory and to develop techniques of fabrication. Physical and performance characteristics of these two batteries are listed in Table I. Details of construction of the two batteries are shown in Figures 3 and 4.

HEAT SOURCE

A heat source was required having dimensions of approximately 0.2 inch in diameter and 0.4 inch in length. Such a heat source was prepared by volatilizing polonium-210 into a capsule or container, closing the capsule with a plug, and coating the capsule with nickel. The finished heat source measured 0.21 inch in diameter by 0.45 inch in length.

BATTERY NUMBER 1

FIGURE 3

540085

540212

fig 4

15

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FIGURE 4

BATTERY NUMBER 2

TABLE I
CHARACTERISTICS OF EXPERIMENTAL BATTERIES

	BATTERY NO. 1	BATTERY NO. 2
RADIOACTIVITY	57 CURIES, Po^{210}	146 CURIES, Po^{210}
CAPSULE CONTAINING Po^{210}	SPHERE, O.D. 0.4 IN.	CYLINDER, O.D. 0.21 IN., LENGTH 0.45 IN.
MATERIAL OF CAPSULE	COLD-ROLLED STEEL WITH 0.02 IN. NICKEL COATING	COLD-ROLLED STEEL WITH 0.02 IN. OF NICKEL COATING
THERMOCOUPLES	SILVER-SOLDERED CHROMEL- CONSTANTAN	WELDED CHROMEL-CONSTANTAN
NUMBER OF JUNCTIONS	7	40
LENGTH OF LEADS	1.2 CM	1.3 CM
WIRE SIZES	B AND S NO. 18 CHROMEL B AND S NO. 20 CONSTANTAN	B AND S NO. 29 CHROMEL B AND S NO. 30 CONSTANTAN
INSULATION BETWEEN JUNCTIONS AND CAPSULE	SAUEREISEN CEMENT	SAUEREISEN CEMENT
ESTIMATED m	0.3	0.2
INSULATING MATERIAL	SANTOCEL	SANTOCEL
OUTSIDE CONTAINER	LUCITE CYLINDER	ALUMINUM CYLINDER
INTERNAL RESISTANCE	0.25 OHM	15 OHMS
VOLTAGE AT NO LOAD	42 MILLIVOLTS	750 MILLIVOLTS
TEMPERATURE RISE FROM HOT TO COLD JUNCTIONS ($T_1 - T_2$)	$42/7 \times 7.7 \times 10^{-2} = 78^\circ\text{C}$	$750/40 \times 7.7 \times 10^{-2} = 244^\circ\text{C}$
TEMPERATURE OF COLD JUNCTION (T_2)	45°C	80°C
TEMPERATURE OF CAPSULE (BASED ON m ABOVE)	146°C	373°C
MAX POWER DELIVERED	1.8 MILLIWATTS	9.4 MILLIWATTS
ACTIVITY OF Po^{210} IN WATTS	1.8 WATTS	4.65 WATTS
EFFICIENCY	0.10 PER CENT	0.20 PER CENT
WEIGHT	34 GMS	31 GMS
WORK CAPACITY	1.5×10^4 JOULES	7.7×10^4 JOULES
CURRENT AT MAX POWER	85 MILLIAMPS	25 MILLIAMPS

373
80
293

COMPARISON WITH COMMERCIAL BATTERIES

Batteries powered by radioactivity cannot be compared directly with chemical batteries because the life of the thermal battery is independent of current drain. In particular, a rating in ampere-hours has no meaning so that a comparison of work, in joules, should be made.

The Mallory Mercury Cell (RM4Z) is considered to be an efficient small-size dry cell in terms of energy per unit mass. It is rated at 2,300 milliamperes-hours, which corresponds to a total output of 8,280 coulombs. The rated voltage is 1.3 volts, and the battery will probably average 1.0 volt over its life-time, so that the total work from an RM4Z cell is 8,280 joules.

The total work which the polonium battery can perform is

$$J = \frac{1}{100} \int_0^{\delta} \text{Eff} \times W_s dt \quad (25)$$

where

$$W_s = W_{s(\max)} e^{-\lambda t} \quad (26)$$

and λ is the radioactive decay constant. In one half-life the efficiency will not decrease appreciably, and one half-life probably represents the useful life of the battery. Then

$$J = \frac{\text{Eff} W_{s(\max)}}{100} \int_0^{T_{1/2}} e^{-\lambda t} dt = \frac{\text{Eff} W_{s(\max)}}{100 \lambda} \quad (27)$$

The radioactivity constant λ is related to the half-life:

$$\lambda = \frac{0.693}{T_{1/2}} \quad (28)$$

For polonium, with a half-life of 138.4 days,

$$\lambda = 5.8 \times 10^{-8} \text{ sec}^{-1}$$

From Equation (27) the total useful work of the two completed batteries would be 1.5×10^4 and 7.7×10^4 joules, respectively, or about 1.8 and 9.2 times that of the RM4Z cell. On a weight basis, the polonium batteries weigh 34 and 31 grams, respectively, in comparison to 45 grams for the RM4Z mercury cell.

CONCLUSIONS

A thermal battery, using polonium-210 as the heat source, has been made which has about 10 times the work capacity of ordinary dry cells of the same weight.

Another advantage of such batteries is the insensitivity of the output to ambient temperatures. From the theory output should increase slowly with decreasing ambient temperatures to absolute zero.

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