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TECHNIQUES FOR THE MEASUREMENT OF HEAT RELEASED ON PLANAR SURFACES OF SMALL AMOUNTS

M.S. Thesis

February 1968

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TECHNIQUES FOR THE MEASUREMENT OF SMALL AMOUNTS OF HEAT RELEASED ON PLANAR SURFACES

Ramesh Chander K. Gupta
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TECHNIQUES FOR THE MEASUREMENT OF SMALL AMOUNTS
OF HEAT RELEASED ON PLANAR SURFACES

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ABSTRACT

The measurement of small quantities of heat released on planar solid
surfaces has been attempted by the use of thermopile and pyroelectric
elements. Calculations to relate the heat released to the output of
these devices have been performed. Considerable simplification is
achieved for cases where the time constant of heat liberation on the
surface is either very large or very small as compared to the time con-
ostant of the probe. The experimentally observed output of two thermo-
piles under different conditions of heat release has been used for
deriving calibration parameters which have been found to be independent
of the calibrating conditions. Thus, under certain conditions, charac-
teristic time constant and magnitude of the heat release on a surface
can be derived from the measurement.
I. INTRODUCTION

The measurement of heat liberated on a solid surface is of interest in the study of different interfacial phenomena, such as adsorption of gases and solutes, wetting process or electrochemical reactions.

Conventionally, the measurement of heats of adsorption and wetting processes are carried out by using powdered materials which because of their large surface area per unit mass give rise to relatively large temperature changes. Such determinations have the disadvantages of poorly defined surface structure, difficulties in removing and preventing contamination of the surface and uncertainty about the surface area measurement. These difficulties could be reduced greatly by the use of well defined planar solid surfaces. However, because of their small specific surface area, it is necessary to measure small amounts of energy released.

The direct experimental methods for measuring heat released on a surface can be divided into two classes, namely static and dynamic methods. In static methods, the heat content of the calorimeter and its contents is measured before and after the heat release has taken place. In dynamic methods, the heat liberated is allowed to be conducted away in a well defined way and the temperature at the surface, which is changing with time, is related to the heat released by means of the heat transfer equation.

It should be noted that both methods involve measurement of temperature changes of either the surface on which heat effects are taking place or of the bulk. The temperature measured at any time may be either absolute or relative to a reference point, kept at constant temperature.
Static methods have the disadvantage that only total heat evolved can be obtained. Also, because of the large heat capacity of the system, the temperature changes are small and difficult to measure. A large number of methods are available to measure change of temperature with time. However, only a few appear to be suitable for measuring small temperature changes (of the order of $10^{-5} \, ^{\circ}C$) confined to small regions, which are encountered during the dynamic measurements of heat effects on a surface. These are listed below.

1. Resistance thermometers
2. Pyroelectric devices
3. Thermocouple devices

Resistance thermometry is based on the change in the electrical resistance of the probe due to change in temperature. The ultimate sensitivity of the resistance thermometry is limited by considerations of electrical noise of the circuit and heat dissipation in the probe. A theoretical analysis taking the above two factors into consideration, indicates a lower limit of $10^{-5} \, ^{\circ}C$ for the measurement of temperature changes. The reported value of smallest temperature change measured by resistance thermometry is also about $10^{-5} \, ^{\circ}C$.

Pyroelectric crystals provide an accurate means of measuring temperature changes. A change in temperature produces a spontaneous polarization of the crystal which gives rise to a pyroelectric voltage between two electrodes placed perpendicular to the polar axis. This pyroelectric voltage can be measured by an electrometer. Using a barium titanate crystal, Lang and Steckel\textsuperscript{3} reported the measurement of bulk temperature changes of
the order of $10^{-6^\circ}C$.

Thermocouples are probably the most widely used temperature measuring device and have been used successfully for measuring both large and small temperature differences. Increased sensitivity is obtained with multiple junctions in series.

The object of the present work is to examine theoretically the possibility of using thermocouples and pyroelectric crystals to measure small heat effects on solid surface, to fabricate probes of optimum design and to study their performance under known rates of heat release on the surface.
II. THERMOCOUPLE DEVICE

In order to arrive at a definite relationship between the heat liberated on the surface and its change in temperature with time measured by thermocouples relative to a reference point kept at constant temperature, it is necessary to know exactly how the heat is being dissipated from the surface. Heat is lost from the surface by conduction through the thermocouple wires, by conduction through the solid providing the surface, and by conduction and convection through the surrounding fluid medium. The accuracy of the relationship between the heat liberated on the surface and the change of temperature of the surface depends on the accuracy with which these heat losses from the surface can be predicted. By using a definite shape of the solid which provides the surface for heat release and by embedding the thermocouples in the solid, it is possible to obtain a relationship between the heat liberated on the surface and its temperature change.

For this work, a rectangular shape has been selected for the solid because of the relative ease of fabrication. Heat lost by conduction and convection in the surrounding fluid has been neglected in the theoretical analysis. The relative amount of heat dissipated by conduction through the solid containing the thermocouples, the thermopile, and through the fluid medium depends upon their relative thermal conductivities and thermal diffusivities, and can be approximately calculated for different cases of heat release on the surface.

The thermopile consists of a number of thermocouples connected in series and embedded in a block of some thermally insulating material such
as epoxy resin, with the alternate junctions being on the two opposite faces of the block. A cross-section of the thermopile is shown in Fig. 1. One of the faces of the thermopile containing the junctions is kept in contact with a constant temperature heat sink, heat is released on the other face containing the junctions.

A. Simple Relationship between the Heat Released on the Surface and the E.M.F. Produced by the Thermopile

Consider a thermopile of the construction described above connected to an instrument to measure the E.M.F. produced by the thermopile. Let \( q(t) \) be the rate of heat released per unit area of the surface at time, \( t \). Neglecting the heat conducted away by the surroundings and using the argument that part of the heat is stored in the thermopile and part is lost to the sink, the following equation can be derived.

\[
q(t) = \frac{1}{A g_1} (p z + \mu \frac{dz}{dt} + \frac{N^2 \beta \epsilon_o z}{(NL/r_1/A_1 + r_2/A_2 + R_f)}).
\]  

Here \( p \) is the effective thermal conductance of the thermopile, \( \mu \) is the effective thermal capacity of the thermopile, \( N \) is the number of junctions, \( \beta \) is the Peltier coefficient, \( \epsilon_o \) is the thermoelectric coefficient, \( L \) is the length of the thermopile, \( r_1 \) and \( r_2 \) are the specific electrical resistivities of the two materials of the thermocouples having cross-section areas \( A_1 \) and \( A_2 \) respectively. \( A \) is the area of the surface on which heat is released, \( R_f \) is the resistance of the external circuit, \( g_1 \) is a constant defined as the units of output, \( z \), of the measuring instrument produced at steady state by one degree centigrade difference in temperature between the two opposite faces of the thermopile. The first term
Fig. 1  Schematic diagram of thermopile probe.
A - Surface on which heat is released
B - Thermocouple wires
C - Thermal insulation
D - Insulation over the thermocouple junctions
E - Electrical leads
F - Constant temperature heat sink
on the right hand side of Eq. (II-1) denotes heat lost by conduction, the second term denotes heat stored in the thermopile and the third term is the correction term to take care of the Peltier effect. Generally, the Peltier effect is very small and can be neglected.

Equation (II-1), called Tian's equation, has been successfully used for cases where the heat is liberated over a large period of time which may extend over a few hours. However, for heat release over short time periods, this equation is not applicable because of the marked dependence of p and μ on the rate of heat release on the surface.

B. Theoretical Relationship between the Heat Released on the Surface and the Output of the Thermopile

For any thermopile, the electrical output is proportional to the instantaneous temperature difference between the junctions on the opposite sides of the thermopile. This electrical output may be related directly to a heat release on one of the thermopile surfaces, once the unsteady state temperature distribution is known. For this purpose, a theoretical derivation of the unsteady state temperature distribution is developed below.

Consider a thermopile of length L and cross-sectional area A with one face containing the junctions in contact with a constant temperature heat sink as shown in Fig. 2. Let q(t) be the rate of heat released per unit area of the surface at time t, and let all of it be conducted away through the thermopile. (Heat losses by conduction through the surrounding medium can be taken into account during calibration of the thermopile as given in Appendix B.) Let T be the temperature of any element of the thermopile relative to the temperature of the heat sink.

The flow of heat in the thermopile is essentially two-dimensional and can be represented by the two-dimensional heat conduction equation:
Fig. 2 Coordinate system for heat released on the surface of the heat probe.
where \( k \) is the thermal conductivity, \( \rho \) is the density and \( c \) is the specific heat. Because three different materials of the thermopile are conducting heat, the exact solution of the above equation would be very complicated. However, the problem is greatly simplified if only one-dimensional heat conduction is assumed with no flow of heat along the \( y \) axis (Fig. 2). Then, \( k, \rho \) and \( c \) become average properties of the thermopile.

For the simplified heat conduction equation, the initial and boundary conditions become:

\[
\begin{align*}
\text{at } t = 0, & \quad T = 0 \text{ for all } x \\
\text{at } x = 0, & \quad T = 0 \text{ for all } t \\
\text{and at } x = L, & \quad \frac{\partial T}{\partial x} = \frac{q(t)}{k}
\end{align*}
\]

The rate at which heat is liberated on the surface can be represented by:

\[
q(t) = \sum_{i} a_{i} e^{-\omega_{i} t}
\]

where \( a_{i} \) and \( \omega_{i} \) are constants. The reason for selecting such a representation of heat liberation is that in the processes of interest the rate of heat release as a function of time shows generally a peak followed by a monotonic decay. Such a function can be described by the first two terms of the expansion (II-3). In such cases, using the condition that \( q(t) = 0 \) at \( t = 0 \), Eq. (II-3) reduces to:
where $a$, $\omega_1$ and $\omega_2$ are constants. By using Eq. (II-3), the last boundary condition becomes:

$$\frac{\partial T}{\partial x} \bigg|_L = \frac{\sum a_i e^{-\omega_i t}}{k}$$

The one-dimensional heat conduction equation can be solved for the above boundary conditions by means of the Laplace transform (Appendix A). The resulting solution is:

$$T = \sum_{i} a_i \frac{\sin \left( \frac{\omega_i}{\alpha} x \right)}{\cos \left( \frac{\omega_i}{\alpha} L \right)} e^{-\omega_i t}$$

$$= \sum_{i} \sum_{n=0}^{\infty} \frac{2a_i (-1)^n L}{k \left( \frac{2n+1}{2} \pi \right)^2} \frac{\sin \left( \frac{2n+1}{2} \frac{\pi x}{L} \right)}{1 - \frac{\omega L^2}{4n+1} \left( \frac{2n+1}{2} \pi \right)^2} e^{-\alpha \left( \frac{2n+1}{2} \frac{\pi}{L} \right)^2 t}$$

where $T$ is the difference between the temperature of any point inside the thermopile at a distance $x$ from the heat sink and the temperature of the heat sink, $k$ is an average conductivity of the thermopile, and $n$ is an integer.

In order to take the finite thermal capacitance of the surface into account, let the thickness of the extra insulation between the junctions and the surface on both faces of the thermopile be each equal to $\delta$. 
Then the two sets of junctions of the thermocouples lie at a distance \(8\) and \((L-8)\) from the face in contact with the constant temperature heat sink. The difference of temperature, \(T_s\), between the junctions of the thermocouples situated on opposite faces can be given by:

\[
T_s = \frac{a_1}{\frac{8}{L}} \left[ \sin \left( \frac{\sqrt{\omega_1}}{\alpha} (L-8) \right) - \sin \left( \frac{\sqrt{\omega_1}}{\alpha} 8 \right) \right] e^{-\alpha t}
\]

\[e^{-\alpha \frac{(2n+1) \pi}{2} t}\]

\[
\sum_{i=0}^{\infty} \sum_{m=0}^{\infty} \frac{2 a_1 (-1)^n}{L} \left[ \sin \left( \frac{2n+1}{2} \pi \frac{L-8}{L} \right) - \sin \left( \frac{2n+1}{2} \pi \frac{8}{L} \right) \right]
\]

\[
\frac{\alpha \frac{L^2}{(2n+1)^2 \pi^2}}{1 - \frac{\alpha \frac{L^2}{(2n+1)^2 \pi^2}}{\omega_1 L^2}}
\]

\[
\cdot e^{-\alpha \frac{(2n+1) \pi}{2} t}
\]

If \(N\) is the number of thermocouple junctions in the thermopile and \(\varepsilon_o\) is the thermoelectric constant, then the E.M.F., \(E(t)\), generated by the thermopile can be given by:

\[
E(t) = \varepsilon_o T_s N
\]

and the current, \(I(t)\), flowing in the external circuit is given by:

\[
I(t) = \frac{\varepsilon_o T_s N}{N L_w \left( \frac{r_1}{A_1} + \frac{r_2}{A_2} \right) + R_f} \equiv \lambda T_s \quad (II-7)
\]

where \(L_w\) is the length of thermocouple elements between the junctions, \(r_1\) and \(r_2\) are the specific electrical resistivities of the materials of the thermocouple wires having cross-sectional area \(A_1\) and \(A_2\) respectively, \(R_f\) is the external circuit resistance and \(\lambda\) is defined by Eq. (II-7). The
length, $L_w$, of the thermocouple wires can be taken as approximately equal to the length, $L$, at the thermopile. If $z$ is the output produced by the measuring instrument when a current, $I(t)$, passes through it, then one can write,

$$I(t) = gz$$  \hspace{1cm} \text{(II-8)}

where $g$ is the sensitivity of the instrument. Neglecting Peltier's effect, $z$ can be obtained as a function of time from Eqs. (II-6, II-7 and II-8) as:

$$z = \frac{\lambda}{g} \left\{ \sum_i \frac{a_i}{k \sqrt{\omega_i/\alpha}} \frac{[\sin(\sqrt{\omega_i/\alpha} (L-\delta)) - \sin(\sqrt{\omega_i/\alpha} \beta)]}{\cos(\sqrt{\omega_i/\alpha} L)} e^{-\omega_i t} \right\}$$

$$- \sum_{i} \sum_{n=0}^{\infty} \frac{2a_i (-1)^n L}{k(\frac{2n+1}{2})^2} \frac{[\sin(\frac{2n+1}{2} \frac{L-\delta}{L}) - \sin(\frac{2n+1}{2} \frac{\pi \delta}{L})]}{\left(1 - \frac{\omega_i L^2}{(\frac{2n+1}{2} \pi)^2 \alpha}\right)} e^{-\alpha(\frac{2n+1}{2} \frac{\pi}{L})^2 t}$$

The above equation assumes an instantaneous response of the measuring instruments. For measuring instruments with finite response time, Eq. (II-8) can be modified to:

$$I(t) = gz + \tau_I g \frac{dz}{dt}$$  \hspace{1cm} \text{(II-8a)}

where $\tau_I$ is the time constant of the measuring instrument. Then from Eqs. (II-6, II-7, and II-8a) the output, $z$, produced by the measuring instrument can be given by:
This equation can be used to calculate the output at any time, $t$, for known rates of heat liberation on the surface of the thermopile.
C. Simplified Relations Between the Heat Liberated on the Surface of a Thermopile and the Output of Measuring Instrument

Equation (II-9) can be considerably simplified under certain conditions.

1) Constant rate of heat release on the surface ($a_1 = 0$, $a_2 = \infty$).

Equation (II-9) gives:

\[
z = \frac{\lambda}{g} \left[ \frac{aL}{k} \left(1 - e^{-t/\tau_I}\right) - \sum_{n=0}^{\infty} 2a(-1)^n L \frac{\sin\left(\frac{2n+1}{2} \frac{L-\delta}{L}\right) - \sin\left(\frac{2n+1}{2} \frac{\pi n}{L}\right)}{k \left(\frac{2n+1}{2} \cdot \pi\right)^2} \right] \]

For time $t$, much greater than the time constant $\tau_I$, ($t/\tau_I \gg 1$), one gets

\[
z = \frac{\lambda}{g} \left[ \frac{aL}{k} \left(1 - e^{-t/\tau_I}\right) - \sum_{n=0}^{\infty} 2a(-1)^n \frac{\sin\left(\frac{2n+1}{2} \frac{L-\delta}{L}\right) - \sin\left(\frac{2n+1}{2} \frac{\pi n}{L}\right)}{k \left(\frac{2n+1}{2} \cdot \pi\right)^2} \right] \]

At steady state ($t \to \infty$) this gives:

\[
z = \frac{\lambda}{g} \cdot \frac{aL}{k} \]

2) Slow heat release on the surface.

Under the condition:
and assuming that the heat liberated on the surface can be represented by Eq. (II-4), the Eq. (II-9) reduces to

\[
z = \frac{\lambda}{g} \frac{aL}{k} \left[ \frac{\sin(\sqrt{\omega_1/\alpha} (L-8)) - \sin(\sqrt{\omega_1/\alpha} 8)}{\sqrt{\omega_1/\alpha} L \cdot \cos(\sqrt{\omega_1/\alpha} L)} \left( e^{-t/\tau_I} - e^{-\omega_1 t} \right) \right] \]

\[
- \frac{\sin(\sqrt{\omega_2/\alpha} (L-8)) - \sin(\sqrt{\omega_2/\alpha} 8)}{\sqrt{\omega_2/\alpha} L \cdot \cos(\sqrt{\omega_2/\alpha} L)} \left( e^{-t/\tau_I} - e^{-\omega_2 t} \right) \right] \]

(II-13)

Usually, the value of \(1/\omega_1\) is very high as compared to the time constant of the instrument, \(\tau_I\). This fact, together with Eq. (II-12) further simplifies the Eq. (II-13) to give

\[
z = \frac{\lambda}{g} \frac{aL}{k} \left[ e^{-\omega_1 t} - e^{-\omega_2 t} \right]
\]
or

\[
z = \frac{\lambda L}{gk} q(t)
\]

(II-13a)

This indicates that in such a case, at any time \(t\), the output from the measuring instrument is proportional to the rate of heat released at the surface.

For a typical epoxy resin-embedded thermopile with length of 1 cm and thermal diffusivity of about \(10^{-3}\) cm²/sec., Eq. (II-12) gives:

\[
\omega_1 \ll 2.5 \times 10^{-3} \text{ sec}^{-1}
\]
This value corresponds to the characteristic time constant of the heat release on the surface of the order of a few hours. Therefore, for the cases, where heat is released on the surface over a period of one hour or more, the rate of heat release on the surface as a function of time can be obtained from Eq. (II-13a) by multiplying the output, \( z \), at any time by a factor \((g \cdot k/\lambda I)\), where \( g \) is the sensitivity of the measuring instrument given by Eq. (II-8), \( k \) is the average conductivity of the thermopile, \( \lambda \) is the current produced in the circuit by \( 1^\circ C \) difference of temperature between the opposite set of junctions as defined by Eq. (II-7), and \( L \) is the length of the thermopile.

3) **Fast heat release on the surface of the thermopile.**

Under the condition:

\[
\frac{\omega_1 L^2}{(\pi^2/\alpha)} \gg 1
\]

(II-14)

and assuming that the heat liberated on the surface can be represented by Eq. (II-4), Eq. (II-9) reduces to

\[
z = \sum_{n=0}^{\infty} \frac{\lambda}{kL} 2a(-1)^n \alpha \left\{ \sin\left(\frac{2n+1}{2} \cdot \frac{\pi \cdot L}{L} \right) - \sin\left(\frac{2n+1}{2} \cdot \frac{\pi L}{L} \right) \right\}
\]

\[
\left( \alpha \left( \frac{2n+1}{2} \cdot \frac{\pi^2}{L^2} \right) \right) \tau_1 - 1
\]

\[
\left[ e^{-t/\tau_1} - e^{-\alpha \left( \frac{2n+1}{2} \cdot \frac{\pi L}{L} \right)^2 t} \right] \left[ \frac{1}{\omega_1} - \frac{1}{\omega_2} \right]
\]

(II-15)

Defining \( f(t) \), which is a function of time as:
\[ f(t) = \sum_{n=0}^{\infty} \frac{\lambda}{\pi n g} \frac{2(-1)^n \alpha}{kL} \left[ \sin\left(\frac{2n+1}{2} \cdot \frac{\pi L}{L - 5}\right) - \sin\left(\frac{2n+1}{2} \cdot \frac{\pi h}{L}\right)\right] \left[ e^{\frac{-t}{\tau e}} - e^{\frac{-\alpha(2n+1)}{L} \cdot \frac{\pi^2}{2} t} \right] \]

(II-16)

and noting that the total heat liberated on the surface \( Q \), obtained by the integration of Eq. (II-4), is given by:

\[ Q = \int a(e^{-\omega_1 t} - e^{-\omega_2 t}) dt = a(1/\omega_1 - 1/\omega_2), \]

Eq. (II-15) can be rewritten as:

\[ Q = \frac{z}{f(t)} \]

(II-17)

It is evident from Eq. (II-16) that \( f(t) \), which is a function of time, is dependent only on the properties of the thermopile and the measuring instrument and since the total heat liberated, \( Q \), is not a function of time, Eq. (II-17) indicates that the output, \( z \), which is a function of time should be proportional to \( f(t) \) as long as Eq. (II-14) is satisfied. Then the total heat liberated, \( Q \), on the surface can be given by the ratio \( z/f(t) \) at any time.

For a typical epoxy resin embedded thermopile having a length of 1 cm and thermal diffusivity of \( 10^{-3} \text{ cm}^2/\text{sec} \), Eq. (II-14) gives:

\[ \omega_1 \gg 2.5 \times 10^{-3} \text{ sec}^{-1} \]

This condition indicates that the characteristic time constant of the heat release on the surface should be less than about 10 seconds for the above simplification to hold. Therefore, for the cases where
most of the heat is released on the surface in a period of about 10 seconds or less, the total amount of heat released can be obtained from the ratio of the function $f(t)$, defined by Eq. (II-16) and the output, $z$, at any time.

It should be noted from Eq. (II-17) that for the relatively fast rates of heat release, it becomes impossible to compute the time dependence of the heat release from the time dependence of the output.

D. Optimum Design and the Sensitivity of the Thermopile

The sensitivity and optimum design of the thermopile could be obtained from Eq. (II-9). However, it is clear that the final design will depend strongly on the way in which the heat is liberated on the surface and will be optimum only for that particular manner of heat release. In order to provide some guidelines for the design of the thermopile, the case is studied here in which the heat is being evolved uniformly and a steady state temperature distribution has been established. This case is treated here because of the relatively simple mathematics involved.

In such a case one can write:

$$qA = pT_s + \frac{N^2 \beta c_0}{NL(r_1/A_1 + r_2/A_2) + R_f} T_s$$

where $q$ is the heat liberated on the surface per unit area per unit time, $A$ is the total cross-sectional area and $T_s$ is the temperature of the surface on which heat is being liberated relative to that of the heat sink.

Under steady state conditions, the thermal conductance, $\rho$, can be given by:

$$\rho = \frac{N}{L} (k_1A_1 + k_2A_2) + \frac{k_3A_3}{L}$$

(II-19)
where \( k_1, k_2 \) and \( k_3 \) are the thermal conductivities of the two thermocouple materials and the material in which thermocouples are embedded, respectively. The factors \( NA_1, NA_2 \) and \( A_3 \) are the cross-sectional areas of the three materials, respectively.

If \( z \) is the output produced by the measuring instrument on passing current \( I \) through the external circuit, then:

\[
I = \frac{\varepsilon_0 N T_s}{(NL(r_1/A_1 + r_2/A_2) + R_f)} = g z \quad (\text{II-20})
\]

where \( g \) is the current in the external circuit to produce a unit output.

From Eqs. (II-18, II-19, and II-20), one gets:

\[
As = \frac{gA}{z} = \left\{ \frac{N(k_1A_1 + k_2A_2)}{L} + \frac{N^2 \beta \varepsilon_0}{((r_1/A_1 + r_2/A_2)NL + R_f)} \right\} \frac{g[L(r_1/A_1 + r_2/A_2) + R_f]}{\varepsilon_0 N}
\]

where \( s \) is a measure of sensitivity of the thermopile. A higher sensitivity corresponds to a smaller value of \( s \).

The above equation simplifies to:

\[
As = \frac{g}{\varepsilon_0 N} \left[ \frac{N(k_1A_1 + k_2A_2)}{L} \right] \left[ NL(r_1/A_1 + r_2/A_2) + R_f \right] + N\beta g
\]

(II-21)

The optimum dimensions of the thermopile could be obtained by setting the derivatives of \( s \) with respect to the various parameters in Eq. (II-21) equal to zero.
The expression for maximum sensitivity (corresponding to minimum value of $s$) can be obtained by feeding the value of optimum $N$, $A_1$ and $A_2$ in Eq. (II-21) which on rearranging gives:

$$A_s = \frac{2g}{\varepsilon_0} \sqrt{\frac{k_a R}{3L}} \left[ (\sqrt{r_{1k_z}} + \sqrt{r_{2k_z}}) + \sqrt{(\sqrt{r_{1k_z}} + \sqrt{r_{2k_z}})^2 + \beta \varepsilon_0} \right]$$

As $A = 2A_2$, one gets:
The product of specific electrical resistivity and the thermal conductivity of a given metal is related to the Lorentz number, \( L_0 \), which is defined by the equation

\[ \kappa \rho = L_0 T \]  \hspace{1cm} (II-23a)

where \( T \) is the absolute temperature. The Lorentz number for a given metal is constant but may vary from metal to metal.

One can define:

\[ \langle \sqrt{k_{11}} + \sqrt{k_{22}} \rangle^2 = L_0 \text{avg} \ T \]  \hspace{1cm} (II-23b)

where \( L_0 \text{avg} \) is a sort of average Lorentz number. Also the Peltier coefficient, \( \beta \), and the thermoelectric constant, \( \varepsilon_0 \), are related by:

\[ \beta = \varepsilon_0 \ T \nu \]  \hspace{1cm} (II-23c)

where \( \nu \) is a constant to take into account the difference of units.

Therefore, Eq. (II-23) can be modified to give:

\[ s = \frac{\varepsilon_0^T}{\varepsilon_0} \sqrt{k_{2} R \rho / A_2 L} \left[ \sqrt{L_0 \text{avg}} + \sqrt{L_0 \text{avg} + \nu \varepsilon_0^2} \right] \]  \hspace{1cm} (II-24)

This equation can be rewritten as:

\[ s = \nu g T \sqrt{k_{2} R \rho / A_2 L} \left[ \sqrt{L_0 \text{avg} / \varepsilon_0^2} + \sqrt{L_0 \text{avg} / \varepsilon_0^2 \nu + 1} \right] \]  \hspace{1cm} (II-24a)
The above equation indicates that the sensitivity can be increased by increasing the area of cross-section and length of the thermopile, by decreasing the resistance in the external circuit and by having a lower value of the dimensionless quantity \( \frac{L_0 \text{avg}}{\epsilon_0^2} \). Generally, an increase in the value of the thermoelectric coefficient, \( \epsilon_0 \), is associated with an increase in the Lorentz number. This explains why the use of semiconductor thermocouple elements for a thermopile of our requirement is not necessarily beneficial even though a high thermoelectric coefficient, \( \epsilon_0 \), might be achieved.

For a typical thermopile made of copper and constantan thermocouples embedded in epoxy resin and having the following properties:

- Length = 1 cm
- Average thermal conductivity of epoxy resin = \( 4 \times 10^{-4}\) cal/sec cm\(^\circ\)C
- Thermoelectric coefficient = \( 4.0 \times 10^{-5}\) volt/\(^\circ\)C
- Electrical resistivity of copper = \( 1.6 \times 10^{-6}\) ohm cm
- Electrical resistivity of constantan = \( 44.2 \times 10^{-6}\) ohm cm
- Thermal conductivity of copper = \( 0.925\) cal/sec cm\(^\circ\)C
- Thermal conductivity of constantan = \( 0.054\) cal/sec cm\(^\circ\)C

and connected by a load of 10,000 ohms in the external circuit to a measuring instrument of sensitivity of \( 2 \times 10^{-12}\) amp/unit of output, Eq. (II-24) gives:

\[
s = \frac{2.3 \times 10^{-2}}{\sqrt{A_3}} \text{ ergs/cm}^2 \text{ sec}
\]

\[
A_{\text{copper}} = 2.62 \times 10^{-7} \sqrt{A_3} \text{ cm}^2
\]

\[
A_{\text{constantan}} = 5.71 \times 10^{-6} \sqrt{A_3} \text{ cm}^2
\]

and \( N = 717 \sqrt{A_3} \)

For such a thermopile having 2 cm\(^2\) area of the surface, one gets:

\[
s = 2.3 \times 10^{-2} \text{ ergs/cm}^2 \text{ sec}
\]

\[
A_{\text{copper}} = 2.62 \times 10^{-7} \text{ cm}^2 (= 5.75 \times 10^{-4} \text{ cm dia})
\]
A_{\text{constantan}} = 5.71 \times 10^{-6} \text{ cm}^2 \ (=2.69 \times 10^{-3} \text{ cm dia})

and \( N = 717 \)

It was impossible to construct a thermopile with 717 junctions in an area of 2 cm$^2$ using present fabrication techniques. The actual thermopile constructed is described in Section IV-A.

E. Minimum Detectable Heat Release. Based on Noise Consideration

It is important to analyze the electronic and thermal noise which may impose a lower limit to the detectability of a thermopile. The noise encountered in the thermopile and its circuit consists of:

1. Johnson noise due to the resistance in the circuit.
2. Current noise in the circuit due to flow of current.
3. Thermal noise (temperature fluctuations) in the temperature probe.
4. Electrical and magnetic induction in the circuit from stray fields.

The last source of noise can be minimized by properly shielding the circuits. Johnson noise is the most important of the remaining kinds of noise.

The minimum measurable energy, \( \Delta W_t \), can be given by:

\[
\Delta W_t^2 = \Delta W_T^2 + \frac{V_E^2}{\sigma^2}
\]  \hspace{1cm} (II-25)

where \( \Delta W_T \) is the minimum detectable energy due to the thermal noise alone and \( V_E/\sigma \) is the one due to electrical noise alone, \( V_E \) being the minimum detectable voltage and \( \sigma \) the responsivity which is defined as the ratio.
of the voltage produced in open circuit to the power input.

Equation (II-25) can be written in its detailed form as:

\[ \Delta W_t^2 = 4 \frac{\Delta f}{\Delta f} \left( \frac{4 kT R_t}{\sigma} + \frac{4 kT}{\sigma} \Delta f \right) \]

where \( R_t \) is the total circuit resistance, \( p \) is the thermal conductance, \( \Delta f \) is the frequency bandwidth of the measuring instrument, \( k \) is the Boltzmann constant and \( T \) is the absolute temperature.

Since the responsivity, \( \sigma \), depends on the rate at which heat is being released on the surface in addition to the properties of the thermopile, it is difficult to arrive at any definite value for the minimum measurable energy. However, to get an estimate of the minimum detectable energy, it is sufficient to take the case of a constant rate of heat release on the surface of the thermopile. Then, at steady state,

\[ \sigma = \frac{N \epsilon_o T_0}{p T_s} = \frac{N \epsilon_o}{p} \]

where \( N \) is the number of thermocouples in the thermopile and \( \epsilon_o \) is the thermoelectric constant.

\[ \therefore \Delta W_t^2 = 4 kT \Delta f \left( pT + \frac{R_t p^2}{N^2 \epsilon_o^2} \right) \]

The thermal conductance, \( p \), is given by:

\[ p = \frac{N(k_1 A_1 + k_2 A_2) + k_3 A_3}{L} \]

and the resistance of the circuit, \( R_t \), is given by:

\[ R_t = L N (r_1/A_1 + r_2/A_2) + R_l \]
Using the values of $A_1$, $A_2$ and $N$ evaluated in Section III-D for an optimally designed thermopile of copper and constantan with epoxy insulation, connected to an external resistance, $R_f$, of 10,000 ohms and a measuring instrument of bandwidth, $\Delta f$, of one cycle per second, one gets:

$$\Delta w_t^2 = 4.520 \times 10^{-18} \text{ watt}^2$$

or

$$\Delta w_t = 2.13 \times 10^{-2} \text{ erg/sec}$$

$$= 1.06 \times 10^{-2} \text{ erg/cm}^2 \text{ sec.}$$

The value of the minimum detectable power is in the same range as the sensitivity of the optimally designed thermopile calculated without noise consideration in Sec. (II-D).

The noise level in the thermopile built, which contains considerably less junctions, should be less than $10^{-2}$ erg/cm$^2$ sec because the thermal conductance, $p$, and the total circuit resistance, $R$, are smaller than the corresponding values for optimally designed thermopile. Noise observed in the measurements could largely be accounted for by the characteristics of the measuring instrument.
III. PYROELECTRIC DEVICE

Some of the ferroelectric crystals which have no center of symmetry and have a unique polar axis possess a temperature dependent macroscopic electric moment along the polar axis. These crystals are called pyroelectrics. A change in temperature of such a material, which may also be polycrystalline, makes it electrically polarized and as a result electrostatic charge collects on the faces perpendicular to the polar axis. The direction and magnitude of the charge depends on the direction and magnitude of the temperature change.

A. Effect of Change of the Bulk Temperature of the Pyroelectric Crystal on the Pyroelectric Voltage

The amount of charge produced, $\Delta Q$, as a result of polarization can be related to the temperature change, $\Delta T$, by the following equation:

$$\Delta Q = p A \Delta T$$  \hspace{1cm} (III-1)

where $A$ is the area of the electrodes (placed perpendicular to the polar axis or the "direction of poling," $p$ is the pyroelectric coefficient which is a function of temperature, its value being greatest near the Curie point, which is the highest temperature up to which the pyroelectric phenomenon exists for a given material. When the pyroelectric element is connected to an electrical load, the accumulated charge is lost partly by internal leakage through the element and partly by external leakage through the electrical circuit. A pyroelectric element can be regarded as a charge generator with its own impedance in parallel to the impedance of the external circuit. An equivalent electrical diagram is shown in Fig. 3. $C_p$ and $R_p$ are the capacitance and resistance of the
Fig. 3  Equivalent circuit of a pyroelectric element and measuring circuit for uniform temperature change of the element.

\[ \Delta E = \text{voltage produced across the pyroelectric element} \]
\[ \Delta Q = \text{charge produced by the pyroelectric element} \]
\[ C_P, R_P = \text{electrical capacitance and resistance, respectively, of the pyroelectric element} \]
\[ C_I, R_I = \text{electrical capacitance and resistance, respectively, of the external circuit} \]
pyroelectric element and \( C_1 \) and \( R_1 \) are the capacitance and resistance of the external circuit. Then the voltage, \( \Delta E \), measured across the two electrodes of the element can be related to the instantaneous change of the bulk temperature, \( \Delta T \), of the element by the equation:

\[
\Delta E = \frac{P}{C_p} \frac{\Delta T}{e^{-t/R_t C_t}}
\]  

(III-2)

where \( R_t \) and \( C_t \) are the total resistance and capacitance of the circuit and are given by:

\[
\frac{1}{R_t} = \frac{1}{R_p} + \frac{1}{R_1}
\]

and

\[
C_t = C_p + C_1
\]

B. Pyroelectric Voltage Resulting from a Non-Uniform Temperature of the Pyroelectric Element Due to Heat Release on One of its Surfaces

In order to find a relationship between the pyroelectric voltage and the heat released on the surface, it is necessary to find the change of temperature with time everywhere in the pyroelectric element. This rate of change of temperature at any point in the element could then be related to the pyroelectric voltage produced by the element.

Consider a pyroelectric element of cross-sectional area, \( A \), (perpendicular to the direction of poling) and the length, \( L \), with its one face in contact with a constant temperature heat sink, as shown in Fig. 2.

Let the temperature of the element and the sink be uniform at time, \( t = 0 \). Let \( q(t) \) be the rate of heat evolved per unit area of the surface at time \( t \) and let all of it be conducted away through the element with no loss by conduction and convection through the surrounding medium.
The release of heat on the surface causes a temperature gradient in the element. However, if the element is divided into a large number of very thin slices of cross-sectional area A and thickness $\delta x_i$, the temperature of each slice can be assumed to be uniform at any instant but changing with time. As a result, each slice acts as a small charge generator and the equivalent electrical circuit of the pyroelectric element when connected to an external load is shown in Fig. 4. $Q_i$ is the charge produced by the slice, $i$, (between points $i$ and $i + 1$) which has an electrical resistance $R_i$ and thickness $\delta x_i$. $C_p$ is the total electrical capacitance of the element between the electrodes. $R_f$ and $C_f$ are the resistance and electrical capacitance of the external load. Let $I(t)$ be the current flowing at any time, $t$, as shown in Fig. 4. Then by Kirchoff's rule, the current, $I(t)$, can be given by the sum of the current flowing through the condensers of capacitance $C_f$ and $C_p$ and the current flowing through resistor, $R_f$, or

$$I(t) = \frac{dQ_f}{dt} + \frac{dQ_p}{dt} + \frac{E(t)}{R_f} \quad \text{(III-3)}$$

where $dQ_f/dt$ and $dQ_p/dt$ are the rates of charging the condensers of capacitance $C_f$ and $C_p$ respectively and $E(t)$ is the voltage developed across the resistance $R_f$.

Using the relation:

$$\frac{dQ}{dt} = C \frac{dE}{dt}$$

Eq. (III-3) can be written as:

$$I(t) = (C_p + C_f) \frac{dE(t)}{dt} + \frac{E(t)}{R_f} \quad \text{(III-4)}$$
Fig. 4 Equivalent circuit of a pyroelectric element with non-uniform temperature change in the element.

- $Q_i$ = charge generated by a slice, $i$, of the pyroelectric element
- $C_p, C_f$ = electrical capacitance of pyroelectric element and external circuit, respectively.
- $R_i$ = electrical resistance of a slice, $i$, of the element
- $R_f$ = electrical resistance of external circuit
- $E(t)$ = voltage produced across resistance, $R_f$, at any time, $t$
- $I(t)$ = pyroelectric current at any time, $t$. 
Consider point "1" in Fig. 4, Kirchoff's rule gives:

\[ I(t) = \frac{dQ_1}{dt} - \frac{E_1 - E_2}{R_1} \]  

(III-5)

where \( E_1 \) and \( E_2 \) are the values of electrical potential at points 1 and 2 respectively. The charge, \( Q_1 \), is related to the temperature change of the slice between points 1 and 2 by:

\[ \frac{dQ_1}{dt} = pA \frac{dT}{dt} \bigg|_{1,2} \]  

(III-6)

Here \( T \) is the temperature of the slice between points 1 and 2 relative to the heat sink temperature. Therefore, from Eqs. (III-5) and (III-6),

\[ I(t) = pA \frac{dT}{dt} \bigg|_{1,2} - \frac{E_1 - E_2}{R_1} \]  

(III-7)

Using the relation \( R_1 = \rho \frac{x_1}{A} \), in Eq. (III-7) and re-arranging, one gets:

\[ E_1 - E_2 = \rho r \frac{dT}{dt} \bigg|_{1,2} \frac{x_1}{A} - I(t) \frac{r}{A} x_1 \]  

where \( r \) is the specific electrical resistivity of the material of the element. Other slices can be treated in a similar way and for any slice of the element between points \( i \) and \( i+1 \), the general equation is:

\[ E_i - E_{i+1} = \rho r \frac{dT}{dt} \bigg|_{i,i+1} \frac{x_i}{A} - I(t) \frac{r}{A} x_i \]  

(III-8)

Summing the Eq. (III-8) for \( i = 1, 2, \ldots, n \), one gets:
\[ E(t) = \sum_{i=1}^{n} (E_i - E_{i+1}) = \frac{pr}{A} \sum_{i=1}^{n} \frac{dt}{dx} \delta x_i - I(t) \frac{r}{A} \sum_{i=1}^{n} \delta x_i \]  \hspace{1cm} (III-9)

As \( n \to \infty \), and \( \delta x_i \to 0 \), Eq. (III-9) is modified to:

\[ E(t) = \frac{pr}{A} \int_{0}^{L} \left[ \frac{dt}{dx} \right] dx - I(t) \frac{r}{A} \int_{0}^{L} dx \]  \hspace{1cm} (III-10)

Then, from Eqs. (III-10) and (III-4),

\[ E(t) = \frac{pr}{A} \int_{0}^{L} \left[ \frac{dt}{dx} \right] dx - \frac{r}{A} L \left[ (C_p + C_f) \frac{dE(t)}{dt} + \frac{E(t)}{R_f} \right] \]

where \( \int_{0}^{L} dx = L \)

or,

\[ \int_{0}^{L} \left[ \frac{dt}{dx} \right] dx = \frac{L}{Ap} \left[ C_t \frac{dE(t)}{dt} + \frac{E(t)}{R_t} \right] \]  \hspace{1cm} (III-11)

where \( R_t \) is the total electrical resistance given by:

\[ \frac{1}{R_t} = \frac{1}{R_f} + \frac{1}{R_p} \]

\( C_t \) is the total electrical capacitance given by:

\[ C_t = C_p + C_f \]

and \( R_p \) is the resistance of the pyroelectric element,

\[ R_p = \frac{Mr}{A} \]
Since the rate of heat release, \( q(t) \), can be expressed as:

\[
q(t) = \sum_{i} a_i e^{-\omega_i t}
\]  

the integral in Eq. (III-11) can be evaluated from Eq. (II-5), which gives,

\[
\int \frac{dT}{dt} dx = \sum_{i} \frac{a_i \alpha}{k} \frac{\cos(\sqrt{\omega_i}x)}{\cos(\sqrt{\omega_i}L)} e^{-\omega_i t}
\]

- \[\sum_{i} \sum_{n=0}^{\infty} \frac{2(a_i) (-1)^n \alpha \cos \left( \frac{2n+1}{2} \frac{\pi x}{L} \right)}{k \left( \frac{2n+1}{2} \pi \right)^2} \left( 1 - \frac{\omega_i L^2}{(\frac{2n+1}{2} \pi)^2} \right) e^{-\alpha \left( \frac{2n+1}{2} \pi \right)^2 t}
\]

+ a constant of integration

(III-12)

In order to take the finite surface thermal capacity into account, let the thickness of the insulation at each of the two opposite faces of the pyroelectric element be \( \delta \) and let \( L \) be the total distance between these two faces.
The two electrodes then lie at a distance 5 and L-5 from the heat sink.

Evaluation of the integral in Eq. (III-12) between the limits 5 and (L-5) and substitution of into Eq. (III-11), results in,

\[
\frac{E(t)}{R_t} + C_t \frac{dE(t)}{dt} = \left( \frac{pA}{L-28} \right) \sum_i \frac{a_i \alpha}{k} \left[ \cos \frac{\alpha_1}{\alpha} (L-5) - \cos \frac{\alpha_1}{\alpha} 5 \right] e^{-\alpha_1 t}
\]

\[
- \sum_{i}^{n} \frac{2a_i (-1)^n}{k \frac{2n+1}{2} \pi} \left[ \cos \left( \frac{2n+1}{2} \pi \frac{L-5}{L} - \cos \left( \frac{2n+1}{2} \pi \frac{5}{L} \right) \right] e^{\left( \frac{2n+1}{2} \pi \right)^2 t}
\]

Solution of this first order differential equation gives:

\[
E(t) = \sum_i \frac{pA}{L-28} \frac{1}{C_t} \frac{a_i \alpha}{k} \left[ \cos \frac{\alpha_1}{\alpha} (L-5) - \cos \frac{\alpha_1}{\alpha} 5 \right] \left( e^{-t/R_t C_t} - e^{-\alpha_1 t} \right)
\]

\[
- \sum_{n=0}^{\infty} \frac{2(-1)^n}{\pi \left( \frac{2n+1}{2} \right)^2} \left[ \cos \left( \frac{2n+1}{2} \pi \frac{L-5}{L} - \cos \left( \frac{2n+1}{2} \pi \frac{5}{L} \right) \right] \left( e^{-t/R_t C_t} - e^{-\alpha_1 t} \right) \right]
\]

(III-14)
The above equation gives the time dependence of the pyroelectric voltage, $E(t)$, on time for known rates of heat releases. Equation (III-14) does not consider the response time of the measuring instrument. If $\tau_I$ is the time constant of the instrument, then taking it into account in Eq. (III-14) in the same way as given in Section II-B, one gets:

$$E(t) = \sum_i \frac{PA_i}{(L-2h)C_t} \frac{a_i \alpha}{\tau_I} \left[ \frac{[\cos(\sqrt{\omega_1} \alpha L/8) - \cos(\omega_1 \alpha 8)]}{\cos (\sqrt{\omega_1} \alpha L)} (\omega_1 - 1/R_t C_t) \right] e^{-t/\tau_I} - t/R_tC_t \frac{1/R_tC_t - 1/\tau_I}{\omega_1 - 1/\tau_I}$$

$$- \frac{e^{-t/\tau_I} - e^{\omega_1 t}}{\omega_1 - 1/\tau_I} - \sum_n \left( \frac{2(-1)^n}{\pi(2n+1)^2} \right) \left[ \frac{\cos\left(\frac{(2n+1)\pi L}{2L}\right) - \cos\left(\frac{(2n+1)\pi}{2L}\right)}{1 - \left(\frac{2n+1}{2L}\right)^2} \right]$$

$$\cdot \left[ \frac{e^{-t/\tau_I} - e^{\omega_1 t}}{\omega_1 - 1/\tau_I} - \frac{e^{-t/\tau_I} - e^{\omega_1 t}}{\alpha L^2 \frac{\pi^2}{L^2} - 1/\tau_I} \right]$$

(III-15)

This equation can be used for the calculation of the output voltage as a function of time for known rates of heat release on a surface.

C. Simplified Relations Between Heat Liberated and Voltage Across the Electrodes

The integral in Eq. (III-11) can be easily evaluated to give considerably simplified equations for cases where the time in which the heat is released on the surface is small compared to the time required by the heat to diffuse through the pyroelectric element. Mathematically, this means

$$L \gg (\alpha \tau)^{1/2}$$
where $\alpha$ is the thermal diffusivity of the material and $\tau$ is the characteristic time constant of the heat generation. In such a case, it could be assumed that no heat is lost from the pyroelectric element to the heat sink. Therefore,

$$q(t) = \rho c \int_0^L \left[ \frac{dT}{dt} \right]_x \, dx$$

where $\rho$ is the density and $c$ is the specific heat of the material. Then Eq. (III-11) gives:

$$q(t) = C_t \frac{L \rho c}{Ap} \left[ \frac{dE(t)}{dt} + \frac{E(t)}{R_t C_t} \right] \quad (III-16)$$

Generally the rate of heat evolution, $q(t)$, can be represented by Eq. (II-4) in which case the solution of the differential Eq. (III-16) gives:

$$E(t) = \frac{Ap \alpha}{C_t \rho c L} \left[ \frac{e^{-\omega_1 t} - e^{-t/R_t C_t}}{1/R_t C_t - \omega_1} - \frac{e^{-\omega_2 t} - e^{-t/R_t C_t}}{1/R_t C_t - \omega_2} \right] \quad (III-17)$$

This equation can be further simplified under the following conditions.

1) **Constant heat liberation** ($\omega_1 = 0$, $\omega_2 = \infty$)

Equation (III-17) reduces to

$$E(t) = \frac{Ap R_t}{\rho c L} \left( 1 - e^{-t/R_t C_t} \right)$$

For barium titanate ceramic of 1 cm length and 1 cm$^2$ cross-section area, attached to a vibrating reed electrometer which has a resolution of $2 \times 10^{-5}$ volts,

$$R_t = 10^{10} \text{ ohms}$$
$$C_t = 10^{-10} \text{ farad}$$
\[
\alpha = 4 \times 10^{-3} \text{ cm}^2/\text{sec} \\
p = 2 \times 10^{-8} \text{ coulombs/cm}^2\text{C at 25°C} \\
c = 0.12 \text{ cal/gm°C} \\
\text{and} \quad \rho = 5.3 \text{ gm/cm}^3
\]

Using these numerical values in the above equation, one gets:

\[
E(t) = 0.752 \times 10^{-5} \alpha (1-e^{-t}) \text{ volts}
\]

The units of \( \alpha \) being ergs/cm\(^2\) sec.

For a value of \( \alpha = 100 \) ergs/cm\(^2\) sec,

\[
E(t) = 75.2 \times 10^{-5} (1 - e^{-t}) \text{ volts}
\]

90\% of the maximum value of \( E(t) \) will be reached in about 2.3 seconds.

It should therefore be possible to measure, with an accuracy of about 10\%,

a total of 230 ergs/cm\(^2\) of heat energy released over a period of 2.3 seconds

at a uniform rate.

The assumption that

\[
L >> \sqrt{\alpha \tau}
\]

is also satisfied because

\[
L = 1 \text{ cm and } \sqrt{\alpha \tau} = 0.1 \text{ cm}.
\]

2) \textbf{Instantaneous heat release on the surface} \quad (\text{large values of } \omega_1 \text{ and } \omega_2 \text{ as compared to } 1/RtC_t).

In such a case, the Equation (III-17) simplifies to:

\[
E(t) = \frac{Ap}{C_t \rho CL} e^{-t/RtC_t} \left[ a \left\{ \left( \frac{e^{-\omega_2 t}}{\omega_2} - \frac{e^{-\omega_1 t}}{\omega_1} \right) + \left( \frac{1}{\omega_1} - \frac{1}{\omega_2} \right) \right\} \right]
\]
or
\[ E(t) = \left[ \frac{Ap}{Ct \rho cL} e^{-t/RtCt} \right] \left[ \text{Heat produced up to time, } t \right] \] (III-18)

This indicates that the voltage across the two electrodes at any time is proportional to the total heat liberated on the surface up to that time. The conditions to be satisfied are:

\[ \tau \ll \frac{L^2}{\alpha} \]

and

\[ \tau \ll \frac{RtCt}{\rho cL} \]

where, \( \tau \) is the characteristic time constant for heat liberation on the surface. For barium titanate ceramic mentioned in case 1) Eq. (III-17) is valid for \( \tau \ll 1 \) sec.

3) Slow heat release on the surface (small values of \( \omega_1 \) and \( \omega_2 \) as compared to \( 1/RtCt \)).

In this case, Eq. (III-17) simplifies to:

\[ E(t) = \frac{Ap Rt_c}{\rho cL} q(t) \] (III-19)

This indicates that the voltage across the two electrodes is proportional to the rate of heat liberation on the surface. In order to satisfy the assumption made, that is

\[ \frac{RtCt}{\rho cL} \ll \tau \ll \frac{L^2}{\alpha} \]

the heat should not be released for more than 10 to 15 seconds and \( Rt \) should be less than \( 10^9 \) ohms.
D. Effect of Electrical and Thermal Noise

It is important to consider the noise which imposes lower limits to the sensitivity of the pyroelectric device. Cooper\(^8\) has estimated the minimum detectable power of the pyroelectric crystal after taking into account thermal and electrical noise. The value reported for barium titanate ceramic is \(4.8 \times 10^{-3}\) erg/sec at 300\(^\circ\)K for a probe having 1 mm\(^2\) area. This minimum detectable power based on the noise consideration is less than the value of 100 ergs/cm\(^2\) sec obtained without noise consideration in Section III.C.
IV. THERMOSTAT SYSTEM

It is necessary to maintain the temperature of the heat sink constant within a very narrow limit such that the temperature fluctuations of the heat sink are much smaller than the temperature fluctuations being measured by either a pyroelectric device or a thermopile. Since the temperature difference arising due to liberation of heat on the surface is of the order of $10^{-5} \degree C$, the temperature of the heat sink should not fluctuate by more than about $10^{-8} \degree C$ during the period of observation, which may last for about five minutes.

Control of temperature within such a narrow limit can be obtained by the use of a composite concentric cylinder thermostat, which consists of alternate cylinders of thermally insulating and thermally conducting materials. The temperature wave which originates at the outer surface of the thermostat dampens out as it propagates towards the inside. Thus any temperature fluctuation inside the thermostat will show a greatly reduced amplitude compared to those on the outside.

In order to provide guidelines for the design of such a thermostat, a model composite cylinder thermostat of infinite length, shown in Fig. 5, has been analyzed theoretically. Three concentric cylinders of infinite length, are considered; the outermost and the innermost cylinders are composed of conducting material and the central one is composed of insulating material.
Fig. 5 Cross-section of the thermostat model for theoretical calculations.
A. Mathematical Analysis of the Characteristics of the Model Thermostat System

Consider a section of the thermostat shown in Fig. 4. It is assumed that the temperature of the outer surface varies sinusoidally with an amplitude $T_3$, and a frequency $n$ cycle/min. The differential equation which describes such a heat propagation for cylindrical bodies is given by:

$$\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} - \frac{j\omega T}{\alpha} = 0$$  \hspace{1cm} (IV-1)

where $T$ is the temperature (all temperatures and fluxes obtained are to be multiplied by $e^{j\omega t}$ to give the time dependent temperature and flux), $\alpha$ is the thermal diffusivity, $r$ is the radial distance from the axis and

$$\omega = 2\pi n$$

$$j = \sqrt{-1}$$

The solution of the differential Eq. (IV-1) is:

$$T = P^r I_0(\beta r^{1/2}) + Q^r K_0(\beta r^{1/2}).$$  \hspace{1cm} (IV-2)

where $\beta = \sqrt{\omega/\alpha}$, $I_0(z)$ and $K_0(z)$ are modified Bessel functions of zero order with imaginary argument and $P^r$ and $Q^r$ are constants of integration. The radial flux, $f$, is given by:

$$f = -k \frac{\partial T}{\partial r},$$

where $k$ is the thermal conductivity of the material of the cylinder.

With Eq. (IV-2), one obtains

$$f = -P^r \beta k r^{1/2} I_1(\beta r^{1/2}) + Q^r \beta k r^{1/2} K_1(\beta r^{1/2}).$$  \hspace{1cm} (IV-3)

where $I_1(z)$ and $K_1(z)$ are modified Bessel functions of first order with imaginary argument.
Equations (IV-2) and (IV-3) can be solved step by step numerically to get the attenuation factor which is defined as the ratio of amplitudes of the temperature fluctuation on the outer surface to that on the inner surface of the thermostat. The general procedure consists in solving Eqs. (IV-2) and (IV-3) for \( P' \) and \( Q' \) for the inner side of the innermost cylinder in terms of \( f \) and \( T \). These values of \( P' \) and \( Q' \) are then used to calculate new values of \( f \) and \( T \) for the outer side of the innermost cylinder. The new values of \( f \) and \( T \) are then used to calculate \( P' \) and \( Q' \) for the inner side of the next cylinder. The procedure is repeated until the value of \( f \) and \( T \) for the outermost surface of the thermostat has been calculated. The details of the calculations are given below:

Let

\[
A_{l,m} = I_0(\beta_f r_m^{1/2})
\]

(IV-3a)

\[
B_{l,m} = K_0(\beta_f r_m^{1/2})
\]

(IV-3b)

\[
C_{l,m} = -k_f \beta_f r_m^{1/2} I_1(\beta_f r_m^{1/2})
\]

(IV-3c)

and

\[
D_{l,m} = k_f \beta_f r_m^{1/2} K_1(\beta_f r_m^{1/2})
\]

(IV-3d)

Subscript \( l \) and \( m \) denote the cylinder number and are given in Fig. 4.

Then Eqs. (IV-2) and (IV-3) reduce to:

\[
T_m = P^l A_{l,m} + Q^l \beta_{l,m}
\]

(IV-4)

and

\[
f_m = P^l C_{l,m} + Q^l D_{l,m}
\]

(IV-5)

At the axis (point \( Y \)), as \( r \to 0 \), \( b_{0,0} \to \infty \), which from Eq. (IV-4) yields:

\[
Q^l = 0
\]

Therefore, at point \( X \), on the right hand side,
and which gives:

\[ f_0 = \frac{c_{1,0}}{A_{1,0}} T_0 \]  \hspace{1cm} (IV-6)

Using these values of \( T_0 \) and \( f_0 \) for the left hand side at point \( X \), the value of new \( p^t \) and \( q^t \) are obtained from Eqs. (IV-4) and (IV-5) as:

\[ T_0 = p^t A_{1,0} + q^t B_{1,0} \]
\[ f_0 = p^t C_{1,0} + q^t D_{1,0} \]

Writing in matrix form, gives:

\[
\begin{bmatrix}
T_0 \\
f_0
\end{bmatrix} =
\begin{bmatrix}
A_{1,0} & B_{1,0} \\
C_{1,0} & D_{1,0}
\end{bmatrix}
\begin{bmatrix}
p^t \\
q^t
\end{bmatrix}
\]

which can be rearranged to give:

\[
\begin{bmatrix}
p^t \\
q^t
\end{bmatrix} = \frac{1}{M_{1,0}}
\begin{bmatrix}
D_{1,0} & -B_{1,0} \\
-C_{1,0} & A_{1,0}
\end{bmatrix}
\begin{bmatrix}
T_0 \\
f_0
\end{bmatrix}
\]

where \( M_{l,m} = A_{l,m} D_{l,m} - B_{l,m} C_{l,m} \) is the determinant of the matrix.

These values of \( p^t \) and \( q^t \) can be used to evaluate \( T_1 \) and \( f_1 \) at point \( W \) with the help of Eqs. (IV-4) and (IV-5) as:

\[
\begin{bmatrix}
T_1 \\
f_1
\end{bmatrix} =
\begin{bmatrix}
A_{1,1} & B_{1,1} \\
C_{1,1} & D_{1,1}
\end{bmatrix}
\begin{bmatrix}
p^t \\
q^t
\end{bmatrix}
\]
or, rearranging,

\[
\begin{bmatrix}
T_1 \\
f_1
\end{bmatrix} = \frac{1}{M_{1,0}} \begin{bmatrix}
A_{1,1} & B_{1,1} \\
C_{1,1} & D_{1,1}
\end{bmatrix} \begin{bmatrix}
D_{1,0} & -B_{1,0} \\
-C_{1,0} & A_{1,0}
\end{bmatrix} \begin{bmatrix}
T_0 \\
f_0
\end{bmatrix}
\]

Proceeding in the same way, one gets:

\[
\begin{bmatrix}
T_3 \\
f_3
\end{bmatrix} = \frac{1}{M_{1,0} M_{2,1} M_{3,2}} \begin{bmatrix}
A_{3,3} & B_{3,3} \\
C_{3,3} & D_{3,3}
\end{bmatrix} \begin{bmatrix}
D_{3,2} & -B_{3,2} \\
-C_{3,2} & A_{3,2}
\end{bmatrix} \begin{bmatrix}
A_{2,2} & B_{2,2} \\
C_{2,2} & D_{2,2}
\end{bmatrix} \begin{bmatrix}
D_{2,1} & -B_{2,1} \\
-C_{2,1} & A_{2,1}
\end{bmatrix} \begin{bmatrix}
T_0 \\
f_0
\end{bmatrix}
\]

(IV-7)

This equation, along with Eq. (IV-6), can be used to evaluate the attenuation factor, \(|T_3| / |T_0|\).

A typical composite cylinder thermostat whose dimensions are shown in Fig. 5, and which has outer and inner cylinders of copper and a middle cylinder of cork, was used to calculate the effect of frequency of an artificially created temperature oscillation (on the outer surface) on the attenuation factor. The following physical properties of copper and insulator (cork) were used:

<table>
<thead>
<tr>
<th>Material</th>
<th>Thermal conductivity (\text{cal/cm sec}^\circ\text{C})</th>
<th>Thermal diffusivity (\text{cm}^2/\text{sec})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Copper</td>
<td>0.925</td>
<td>1.12</td>
</tr>
<tr>
<td>Cork</td>
<td>(1.03 \times 10^{-4})</td>
<td>(1.29 \times 10^{-3})</td>
</tr>
</tbody>
</table>
The Bessel functions needed were taken either from tables or from expressions given in ref. 10.

Figure 6 shows the strong dependence of the calculated attenuation factor on the frequency of the temperature oscillation on the outer surface. From a value of $3 \times 10^{16}$ for temperature fluctuations of one cycle per minute the attenuation factor drops to almost 10 for 1 cycle per 1000 minutes. Since the fast temperature fluctuations have a short penetration distance, while the slow fluctuations penetrate deeper into the thermostat, in order to get a high degree of attenuation, it is important to apply drift-free fast temperature oscillation to the outer surface. To reduce the effect of possible low frequency components, it is desirable to use a larger thickness of insulation.

Based on these considerations, the total thickness of the insulation used in the construction of the thermostat was 12.5 cm as compared to 5 cm used for the above model. Circulating water was used to control the temperature of the outside cylinder. The residual temperature fluctuations between heating and cooling of the water thermostat were about 0.2°C and had a frequency of about one cycle per minute. Details of the thermostat constructed are given in Section V-C.
Fig. 6  Effect of the frequency of temperature cycling on attenuation factor (ratio of amplitude of temp. wave outside to that of inside) of thermostat.
V. EXPERIMENTAL

A. Construction of Thermopile Devices

Three thermopiles, named A, B, and S were built for experimental studies.

Thermopile, A, was made by taking a 1 cm$^3$ block of epoxy resin and drilling thirty-two 0.025 cm diameter holes at small angles between two opposite faces of the cube so as to form four rows of the zigzag pattern shown in Fig. 1. 0.025 cm diameter chromel and alumel wires were forced into adjacent holes and were connected in such a way as to form 16 thermocouples connected in series. The junctions of the thermocouples were formed by welding them with a small acetylene torch. Two copper leads of 0.025 cm diameter were welded to the remaining two of the set of thermocouples. Additional epoxy resin was applied to the two faces of the cube containing the junctions. These two faces were then ground until about 0.025 cm of epoxy resin insulation was left over the junctions.

The thermopile was then mounted on a short copper bar by glueing the face of the thermopile containing the leads to it. The copper bar becomes a part of the heat sink when screwed to the top of the innermost copper vessel of the thermostat.

In order to increase the sensitivity of the thermopile device, a new thermopile, named B, was made using smaller diameter thermocouple wires and larger physical dimensions. A new technique was developed to make the thermopile, as drilling accurately placed small holes (less than 0.025 cm diameter) in epoxy resin blocks and welding the junctions was found to be impossible. Chromel and constantan wires of 0.0077 cm diameter were placed on either side of an epoxy resin tape of 0.01 cm thickness and
1.7 cm with at regular spacing and angles and then sandwiched between two pieces of similar epoxy resin tape, as shown in Fig. 7A. The junctions were soft soldered to give a total of twenty-seven thermocouples connected in series. Copper connecting wires were attached to the remaining two junctions.

The epoxy resin tape containing the thermocouples was loosely wound around and glued by epoxy resin to six 1.7 cm broad, 3 mm thick and about 1.5 cm long pieces of solid epoxy resin, as shown in Fig. 7B, to roughly form a rectangular block. After curing of the resin, additional epoxy resin was used to fill up the gaps so as to form a rectangular thermopile of 1.74 cm length and 3.48 cm² surface area of each of the faces containing the thermopile junctions. In order to make the surface containing the thermopile junctions smooth and to electrically insulate the junctions from the surroundings, a 0.01 cm thick epoxy tape was attached to the two faces using epoxy resin.

The face of the thermopile containing the leads was fixed to a short 2.54 cm diameter copper cylinder which forms a part of the heat sink. The completed thermopile is shown in Fig. 8.

Since not much could be done about: 1) reducing the size of the thermocouple wires, 2) increasing the physical dimensions of the thermopile, or 3) increasing the number of junctions per unit area, the only easy way to improve the sensitivity appeared to be to use a construction material with lower thermal conductivity than the epoxy resin. For this purpose, styrofoam insulation was selected. Because of its softness and heat sensitivity a new technique was used to fabricate a thermopile named, S.
Details of construction of thermopile "B"
Fig. 8 Thermopile, B.
A Surface on which heat is released
B Thermopile
C Part of heat sink
Six, 1.9 cm long, 1.7 cm broad and about 3 mm thick strips of styrofoam were taken. Chromel and constantan wires of 0.0077 cm diameter were embedded inside each strip by making eleven fine cuts about 1.5 mm deep and spaced 0.16 cm (1/16") along the breadth of the strip as shown in Fig. 9. The thermocouple junctions were formed by soldering the ends of adjacent constantan and chromel wires so as to give a total of 10 junctions in series on each strip. All the strips were then brought together to form a rectangular block and free chromel and constantan wires of the adjacent strips were also soldered so as to give a total of 33 thermocouples connected in series. In order to hold the strips together, to make the surface containing the junctions smooth and to electrically insulate the junctions, 0.01 cm thick epoxy resin tape was glued to both the faces. The dimensions of the thermopile are:

- length = 1.86 cm
- Area of the faces containing junctions = 3.27 cm$^2$ each.

The face of the thermopile containing the leads was fixed to a short 2.54 cm diameter copper piece.

In order to calibrate and study the performance of the thermopiles, it is necessary to liberate heat on the surface containing the junctions in a known and reproducible manner. Electrical heating was chosen for this purpose. A nichrome film of about 200Å thickness, vacuum deposited on the surface between two thin copper electrodes (about 0.0077 cm diameter) glued to the opposite edges of the face served as the resistor. The evaporation of nichrome was carried out at about 0.1 micron pressure from a 0.075 cm diameter tungsten filament through which a current of about 20 amps was passed. The distance between the filament and the substrate was
Fig. 9  Details of construction of thermopile, "S"  
(one of the six elements shown)
about 12 cm. Deposition was carried out until a resistance of about 500 ohms was measured between the copper electrodes. This required about five to ten seconds.

B. Construction of Pyroelectric Device

The pyroelectric element used was a 1.27 cm diameter and 0.63 cm (1/4") long barium titanate ceramic.* Lead wires were taken from the two flat surfaces. The surfaces had been made electrically conducting by means of silver paint. Epoxy resin tape, 0.01 cm thick, then was used to electrically insulate both faces. One of the faces was then glued to a short, 2.54 cm diameter copper bar. For calibration and performance studies, a nichrome film was vacuum deposited on the other face between two thin copper electrodes to serve as a resistor for electrical heating.

C. Experimental Arrangement of the Thermostat System

Based on the results in Section IV.A, a composite cylinder thermostat made of alternate cylinders of copper and styrofoam, respectively, was constructed. Details of the thermostat are given in Fig. 10. Four concentric cylinders of copper and three of styrofoam were used instead of the two copper cylinders separated by one insulating layer considered in the theoretical analysis. This design further reduces the effect of any slow drifts in the outside surface temperature and uneven heat penetration.

*Gulton Industries, Inc.
Fig. 10 Details of the experimental thermostat.
The innermost copper vessel acts as a constant temperature heat sink for the probe. To facilitate the testing of different temperature probes, the probe was fixed to a short copper bar of 2.5 cm in diameter screwed to the top of the innermost cylindrical copper vessel as shown in Fig. 10.

The temperature of the outermost copper cylinder was controlled by circulating water at a controlled temperature through 6.2 mm diameter copper tube coil soldered to the external surface of the thermostat. In order to avoid excessive heating and cooling loads of the circulating water and also, to reduce the effects of the temperature fluctuations of the room, the thermostat was enclosed in a rectangular wooden box packed with asbestos.

The circulating water of controlled temperature was pumped at the rate of 0.5 g.p.m. from a water bath equipped with refrigeration and heating units.* Under normal operation the water bath gave a temperature cycling with an amplitude of about 0.2°C and a frequency of about one cycle per minute.

The equipment is shown in Fig. 11.

D. Experimental Arrangement and Procedure for Calibration of the Thermopile and Pyroelectric Devices

The test probe was fixed in position in the thermostat, as shown in Fig. 10. Two leads for passing current through the nichrome film heater and two for transmitting the output from the probe to the measuring instrument were provided. Teflon coated wires were used for this purpose.

* Model 2095, Forma Scientific Inc.
Fig. 11. Experimental set-up for measuring small heat release on the surface of the probes.

A  Timers for frequency control
B  Water bath
C  Pump
D  Thermostat
E  Vibrating reed electrometer
F  Microvolt-ammeter
to increase the insulation and thus reduce the current leakage to a minimum. The connection between the two wires carrying the output from the probe and the input leads of the measuring instrument were made in a small metal box, stuffed with cotton to minimize temperature fluctuations and hence to minimize generation of spurious thermoelectric voltage at the connections. To reduce the noise due to stray electrical and magnetic fields, all the wires were shielded and the shields were connected to a common ground. A microvoltammeter* was used to measure the output from the thermopile and a vibrating-reed electrometer** was used for the pyroelectric devices. The output from the meters was recorded by a potentiometric recorder.***

A constant heat release at the surface was achieved by a constant voltage supply.**** Exponential decay type of heat release was achieved by discharging a condenser through the surface heater. The time constant of the exponentially decaying heat release was controlled by changing either the capacitance of the condenser or the additional resistor connected in series with the heater resistor. Resistors up to a value of $10^5$ ohms and capacitors of 11.22 μf and 583 μf were used in the circuit.

A period of about 72 hours was allowed to bring the thermostat to equilibrium before taking readings.

E. Experimental Procedure for Testing the Thermostat

The thermostat was tested by finding the attenuation factor for large temperature oscillation at low frequencies applied to the outermost cylinder.

---

* Model 150AR, Keithley Instruments Inc.
** Cary 31, Applied Physics Corp.
*** Model MR, E. H. Sargent and Co.
**** Model 605, Power Design Inc.
The temperature of the innermost copper vessel was measured with a single copper-constantan thermocouple, whose reference junction was kept at 0°C. In order to reduce the fluctuations of the reference junction, it was enclosed in a thin glass tube filled with white oil and suspended in the stirred ice bath contained in a dewar. The EMF generated in the thermocouple was measured by a microvolt-ammeter whose output was recorded by a potentiometric recorder. Precautions mentioned in Section V.D were taken to prevent spurious electrical noise.

The frequency and the amplitude of the heating and cooling cycle of the circulating water was varied by using two temperature controller elements in the water bath. Both were connected by means of a timer system to the rest of the controller circuit in such a way that at any time, only one of the controller elements was effective. The desired frequency (one cycle in one to three hours) and amplitude (10°C) of the temperature cycle was obtained by adjusting the timers and the temperature settings of the controller elements.
VI. RESULTS AND DISCUSSION

A. Thermopile Device

The experimentally determined output curves obtained for a heat pulse with an exponential decay are shown in Figs. 12 through 19 for thermopile, "B" and in Figs. 20 through 25 for thermopile, "S". The time constants of the decay were between 0.005 seconds and 30 seconds. Both thermopiles were calibrated by the procedure given in Appendix B. The calculated output curves which best fit the experimental data are also shown in Figs. 10 through 19. The resulting calculated values of the thickness of the insulation over the junctions, average thermal conductivity and average thermal diffusivity are given in Tables 1 and 2 for the two thermopiles, along with those calculated from the dimensions and the properties of the materials of the thermopile. In the latter case the observed thickness of the insulation is the thickness over the junctions while the average thermal diffusivity and thermal conductivity are those obtained from the weighted average of the thermal conductivity and the thermal capacity of the different materials of thermopile. Good agreement is observed between the two. Since the thickness calculated for the best fit is the thickness of the insulation which has the same thermal properties as the bulk of the thermopile, the difference between this thickness and the observed thickness of the epoxy resin insulation for thermopile S could be due to the difference in the thermal properties of the two materials. It can be seen from Tables 1 and 2 that the values of the calibration parameters for a given thermopile are independent of the rate of heat release.
Total heat released = 285 ergs/cm²
Time constant of heat released = 0.0064 sec.

Fig. 12 Output-time relation for thermopile B.
Time constant of heat release = 0.0064 sec.

Total heat released = 573 ergs/cm²

Fig. 13 Output-time relation for thermopile, B.
Fig. 14  Output-time relation for thermopile, B.

Time constant of heat release = 0.055 sec.
Total heat released = 366 ergs/cm$^2$
Time constant of heat released = 0.055 sec
Total heat released = 748 ergs/cm²

Fig. 15  Output-time relation for thermopile, B.
Time constant of heat release: 0.33 sec.
Total heat released = 364 ergs/cm$^2$

Fig. 16 Output-time relation for thermopile, B.
Time constant of heat release: 0.33 sec.
Total heat released: 750 ergs/cm²

Fig. 17  Output-time relation for thermopile, B.
Time constant of heat release: 4.41 sec.
Total heat released: 364 ergs/cm²

Fig. 18 Output-time relation for thermopile, B.
Time constant of heat release: 29.2 sec.
Total heat released: 639 ergs/cm²

Fig. 19 Output-time relation for thermopile, B.
Time constant of heat release: 0.0065 sec.
Total heat released: 997 ergs/cm²

Fig. 20 Output-time relation for thermopile, S.
Time constant of heat release: 0.064 sec.
Total heat released: 1350 ergs/cm$^2$

Fig. 21. Output-time relation for thermopile, S.
Time constant of heat release: 0.34 sec.
Total heat released: 1326 ergs/cm$^2$

Fig. 22 Output-time relation for thermopile, S.
Time constant of heat release: 4.42 sec.
Total heat released: 464 ergs/cm²

Fig. 23  Output-time relation for thermopile, S.
Time constant of heat release: 4.42 sec.
Total heat released: 1209 ergs/cm²

Fig. 24  Output time relation for thermopile, S.
Time constant of heat release: 29.17 sec
Total heat released: 770 ergs/cm²

Fig. 25 Output-time relation for thermopile, S.
Table 1. Thermopile "B"

<table>
<thead>
<tr>
<th>Fig. No.</th>
<th>12</th>
<th>14</th>
<th>16</th>
<th>18</th>
<th>19</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness of the insulation, cm.</td>
<td>0.028</td>
<td>0.028</td>
<td>0.028</td>
<td>0.028</td>
<td>0.028</td>
</tr>
<tr>
<td>Average thermal diffusivity, cm²/sec</td>
<td>0.001</td>
<td>0.001</td>
<td>0.001</td>
<td>0.001</td>
<td>0.001</td>
</tr>
<tr>
<td>Average thermal conductivity, cal/cm·sec°C ×10⁴</td>
<td>4.27</td>
<td>4.29</td>
<td>4.36</td>
<td>4.13</td>
<td>3.82</td>
</tr>
</tbody>
</table>

Thickness of the insulation over the junctions (calculated from best fit) = 0.028 cm.

Thickness of the insulation over the junctions (observed) = 0.015 cm.

Average thermal diffusivity of the thermopile (calculated from best fit) = 10⁻³ cm²/sec.

Average thermal diffusivity of the thermopile (calculated from physical properties) ≈ 1.1×10⁻³ cm²/sec.

Average thermal conductivity (calculated from best fit) = 4.23×10⁻⁴ cal/cm·sec°C.

Average thermal conductivity of the thermopile (calculated from physical properties) ≈ 4.4×10⁻⁴ cal/cm·sec°C.
Table 2. Thermopile "S".

Length = 1.86 cm.
Area of the surface on which heat is released = 3.27 cm².
Number of thermocouples = 33, chromel and constantan.
Diameter of the wires = 0.0077 cm.
Thermal insulation used = styrofoam.
Thermoelectric coefficient of the thermocouple = 60 µV/°C (experimental value)
Electrical resistivity (chromel) = 8.3 × 10⁻⁵ ohm cm.
Electrical resistivity (constantan) = 4.9 × 10⁻⁵ ohm cm.
Input resistance of measuring instrument = 3 ohms.
Time constant of the instrument = 0.5 second.

<table>
<thead>
<tr>
<th>Fig. No.</th>
<th>21</th>
<th>22</th>
<th>23</th>
<th>25</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness of the insulation, cm.</td>
<td>0.15</td>
<td>0.15</td>
<td>0.15</td>
<td>0.15</td>
</tr>
<tr>
<td>Average thermal diffusivity, cm²/°C/sec</td>
<td>10⁻²</td>
<td>10⁻²</td>
<td>10⁻²</td>
<td>10⁻²</td>
</tr>
<tr>
<td>Average thermal conductivity, cal/cm.°C. × 10⁻⁴</td>
<td>3.65</td>
<td>3.38</td>
<td>3.38</td>
<td>3.13</td>
</tr>
</tbody>
</table>

Thickness of the insulation over the junctions (calculated from best fit) = 0.15 cm.
Thickness of the insulation over the junctions (observed) = 0.015 cm.
Average thermal diffusivity of the thermopile (calculated from best fit) = 10⁻² cm²/sec.
Average thermal diffusivity of the thermopile (calculated from physical properties) = 9.6 × 10⁻³ cm²/sec.
Average thermal conductivity of the thermopile (calculated from best fit) = 3.39 × 10⁻⁴ cal/cm. sec°C.
Average thermal conductivity of the thermopile (calculated from physical properties) = 1.3 × 10⁻⁴ cal/cm. sec°C.
Comparison of experimental curves obtained for the same time constants but different total amounts of heat released on the surface is made in Table 3. It shows that for the same time constant of the heat release the experimental output of the thermopile is proportional to the amount of heat released on the surface.

Table 4 gives the ratio of the heat liberated at the surface with respect to the current at the peak for cases where the heat release is fast. The constancy of the ratio for a given thermopile verifies Eq. (II-17) which indicates that for fast heat release, the total heat released on the surface can be given by the ratio of the output of the thermopile at any time to the value of function, \( f(t) \) at the same time, where \( f(t) \) is a function of time completely defined for a given thermopile and measuring instrument.

Inspection of the experimental curves indicates that the magnitude of the random fluctuations in the measuring instrument due to electrical noise is about \( 3 \times 10^{-12} \) amp at \( 10^4 \) ohms input resistance. The instantaneous release of heats of the order of 80 ergs/cm\(^2\) and 35 ergs/cm\(^2\) on the surface of the thermopiles, B and S, respectively, with air as the surrounding medium produces a peak current of about \( 1.25 \times 10^{-11} \) amp. The heat released in these cases could therefore be measured with an accuracy of 20 to 25%. The accuracy of measurement is improved for larger amounts of heat release on the surface.
Table 3

<table>
<thead>
<tr>
<th>Fig. No.</th>
<th>Thermopile</th>
<th>ratio of total heat evolved</th>
<th>ratio of the peak current</th>
</tr>
</thead>
<tbody>
<tr>
<td>12 and 13</td>
<td>B</td>
<td>2.01</td>
<td>2.03</td>
</tr>
<tr>
<td>14 and 15</td>
<td>B</td>
<td>2.04</td>
<td>2.00</td>
</tr>
<tr>
<td>16 and 17</td>
<td>B</td>
<td>2.06</td>
<td>2.09</td>
</tr>
<tr>
<td>23 and 24</td>
<td>S</td>
<td>2.60</td>
<td>2.63</td>
</tr>
</tbody>
</table>

Table 4

<table>
<thead>
<tr>
<th>Fig. No.</th>
<th>Thermopile</th>
<th>time constant of heat released on the surface, sec</th>
<th>ratio of total heat released to the peak current</th>
</tr>
</thead>
<tbody>
<tr>
<td>12</td>
<td>B</td>
<td>0.0064</td>
<td>$8.15 \times 10^{12}$</td>
</tr>
<tr>
<td>14</td>
<td>B</td>
<td>0.0550</td>
<td>$7.82 \times 10^{12}$</td>
</tr>
<tr>
<td>16</td>
<td>B</td>
<td>0.3320</td>
<td>$8.96 \times 10^{12}$</td>
</tr>
<tr>
<td>20</td>
<td>S</td>
<td>0.0065</td>
<td>$2.85 \times 10^{12}$</td>
</tr>
<tr>
<td>21</td>
<td>S</td>
<td>0.0640</td>
<td>$2.77 \times 10^{12}$</td>
</tr>
<tr>
<td>22</td>
<td>S</td>
<td>0.3400</td>
<td>$2.83 \times 10^{12}$</td>
</tr>
</tbody>
</table>
B. Pyroelectric Device

Attempts to measure the output of the pyroelectric element for known rates of heat release on one of its surfaces, were unsuccessful. A spurious output, which was larger than the expected output was obtained from the electrometer when current was passed through the nichrome film heater. As a result, it was not possible to get a reliable experimental output.

C. Thermostat

Table 5 gives the experimentally observed attenuation factor given by the ratio of the amplitude of the temperature fluctuation on the outermost copper cylinder and the innermost copper cylinder of the thermostat for different frequencies of the temperature oscillations in the outermost copper cylinder of the thermostat.

Experiments at lower cycle time could not be carried out due to the difficulties in measuring very small temperature changes with time in the innermost copper cylinder. Table 5 shows that a decrease in the cycle time results in an increase in the attenuation factor. This observation is in qualitative agreement with the numerical results given in Fig. 6 for the model thermostat.

Extrapolation of the curve passing through the experimental points shown in Fig. 6, based on the curve obtained by numerical calculations for the model thermostat, gives an attenuation factor greater than $10^{17}$ for temperature cycles of one minute at the outer surface of the thermostat. Since, during normal operation, the temperature fluctuation at the outer surface of the thermostat occurs at about one cycle per minute with an
amplitude of 0.2°C, the extrapolation of the experimental data implies that the temperature in the innermost copper cylinder of the thermostat should not fluctuate by more than $2 \times 10^{-18} \degree C$ (if such a temperature is defined).

<table>
<thead>
<tr>
<th>Run</th>
<th>Cycle time (min/cycle)</th>
<th>Amplitude of the temp. wave in the outermost cylinder, $T_0$</th>
<th>Amplitude of the temp. wave in the innermost cylinder, $T_1$</th>
<th>Attenuation factor ($T_0/T_1$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>63</td>
<td>10°C</td>
<td>$10^{-3}$°C</td>
<td>$10^4$</td>
</tr>
<tr>
<td>2</td>
<td>117</td>
<td>10°C</td>
<td>$8 \times 10^{-3}$°C</td>
<td>1250</td>
</tr>
<tr>
<td>3</td>
<td>175</td>
<td>10°C</td>
<td>$2.25 \times 10^{-2}$°C</td>
<td>445</td>
</tr>
</tbody>
</table>
VII. CONCLUSIONS

In the presence of air as the surrounding medium, total instantaneous heats of the order of 80 ergs/cm² and 35 ergs/cm² released on the surface of the thermopiles B and S, respectively, could be measured with an accuracy of about 20%. However, better accuracy can be obtained for larger amounts of heat. The sensitivity of the thermopile decreases as the rate of heat release on the surface is decreased (which correspond to higher time constants of heat release).

The values of calibration parameters - thickness of the insulation over the junctions, average thermal diffusivity and thermal conductivity of the thermopile are found to be independent of the rate of heat release.

The sensitivity of the thermopile is affected by the thermal properties of the fluid medium in contact with the surface. In general, the higher the thermal conductivity and the heat capacity of the fluid, the lower will be the sensitivity of the thermopile. In order to take the effect of thermal properties of the fluid into account, it is essential to recalibrate the thermopile every time the fluid medium is changed. The change of the fluid properties is reflected by the change in the value of the average thermal conductivity, k, in the Eq. (B-1a) which is used for calibration of the thermopile.

Throughout this work, it has been assumed that the heat is transferred from the surface by conduction alone. This assumption is valid for the thermopile surface in contact with a stagnant fluid because in such a case, the heat lost from the surface by natural convection and radiation is negligible due to the very small temperature differences involved.
The calculations performed indicate that pyroelectric devices should be able to measure with an accuracy of 10%, a total heat release of a few hundred ergs/cm$^2$. The pyroelectric device appears to be specially attractive for the measurement of short thermal pulses or high rates of heat release.
I am grateful to Dr. R. H. Muller for encouragement and guidance during this investigation.

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APPENDIX A

Solution of One Dimensional Heat Conduction Equation for Time Dependent Heat Release on the Surface of the Probe

The one dimensional heat conduction equation:

\[ \frac{\partial^2 T}{\partial x^2} = \frac{1}{\alpha} \cdot \frac{\partial T}{\partial t} \]  \hspace{1cm} (A-1)

can be solved to find temperature as a function of time at any point in the probe, shown in Fig. 2, by using the following initial and boundary conditions.

at \( t = 0 \), \( T = 0 \) for all \( x \)

at \( x = 0 \), \( T = 0 \) for all \( t \)

and \( \frac{\partial T}{\partial x} \bigg|_{x=L} = \frac{q(t)}{k} \)

where, \( T \) is the temperature relative to the heat sink at any point, at a distance \( x \) from the heat sink; \( \alpha \) and \( k \) are the thermal diffusivity and the thermal conductivity respectively, of the probe. The value \( q(t) \) is the rate of heat release per unit surface area of the probe at time \( t \) and is represented by:

\[ q(t) = \sum_{i} a_i e^{-\omega_i t} \]

where \( a_i \) and \( \omega_i \) are constants.

Taking the Laplace transform of Eq. (A-1), and using the initial condition, one gets:

\[ \alpha \frac{\partial^2 T}{\partial x^2} = \mathcal{L}[T] \]  \hspace{1cm} (A-2)
where, $\mathcal{T}$ is the Laplace transform of $T$, given by

$$
\mathcal{T} = \int_0^\infty e^{-st} T(t) \, dt
$$

Equation (A-2) can be solved to give:

$$
\mathcal{T} = A e^{\sqrt{s/\alpha} x} + B e^{-\sqrt{s/\alpha} x}
$$

(A-3)

where, $A$ and $B$ are constants of integration. The transformed boundary conditions are:

at $x = 0$, $\mathcal{T} = 0$

at $x = L$, $\frac{\partial \mathcal{T}}{\partial x} = \sum_i \frac{a_i}{k(s + \omega_i)}$

The constants of integration, $A$ and $B$ in Eq. (A-3), can be evaluated by the use of transformed boundary conditions to give:

$$
\mathcal{T} = \sum_i \frac{a_i \sqrt{\alpha}}{k \sqrt{s}(s + \omega_i)} \left[ -j \sin(j \sqrt{s/\alpha} x) \cos(j \sqrt{s/\alpha} L) \right]
$$

(A-4)

where $j = \sqrt{-1}$.

The function $\mathcal{T}$ has a branch point at $s = 0$ and poles at $s = -\omega_i$ and at $s = -(2n+1)\pi/2L)^2/\alpha$, where $n$ is an integer. Equation (A-4) can be inverted by the usual contour integration to give the relative temperature, $T$, as a function of time, $t$, as:

$$
T = \sum_i \frac{a_i \sin(\sqrt{\omega_i/\alpha} x) e^{-\omega_i t}}{k \sqrt{\omega_i/\alpha} \cos(\sqrt{\omega_i/\alpha} L)} - \sum_{n=0}^{\infty} \frac{2a_i (-1)^n}{k(2n+1)^2} \left( 1 - \frac{\omega_i L^2}{(2n+1)^2 \pi^2} \right)^{\alpha \pi^2} e^{-\alpha(2n+1/2 \pi/L)^2 t}
$$
APPENDIX B

Procedure for Calibration of the Thermopile and the Calculation of Heat Released at the Surface from the Experimental Output of the Thermopile

Calibration of the thermopile basically consists in experimentally determining the thickness of the insulation over the junction \( \delta \), average thermal diffusivity \( \alpha \), and average thermal conductivity \( k \) of the thermopile such that Eq. (II-9) fits the experimental output of the thermopile.

In the processes of interest, the heat released on the surface can be represented by the first two terms of Eq. (II-3),

\[
q(t) = a \left( e^{-\alpha_1 t} - e^{-\alpha_2 t} \right)
\]

(II-4)

where \( q(t) \) is the rate of heat release per unit area of the surface at time \( t \), and \( a, \alpha_1 \) and \( \alpha_2 \) are constants. For these cases, Eq. (II-9) can be rearranged to give:

\[
U = \sum_{i=1}^{\infty} \left[ \sin(M_1(1-y)) - \sin(M_1y) \right] \left[ e^{-t/\tau_1} - e^{-\alpha_1 t} \right] (-1)^{i+1}
\]

(II-9)

\[
2(-1)^{n+1} \sum_{i=1}^{\infty} \sum_{n=0}^{\infty} \left[ \frac{\sin(\frac{2n+1}{2} \pi (1-y)) - \sin(\frac{2n+1}{2} \pi y)}{(\frac{2n+1}{2} \pi)^2 - M_1^2} \right] \left[ e^{-t/\tau_1} - e^{-\frac{(2n+1)^2 \pi^2}{M_1^2} \omega_1 t} \right]
\]

(B-1)
where,

\[ U = \frac{zgk}{a\lambda L} \]

\[ M_1 = \sqrt{\frac{\omega_1}{\alpha L}} \]

and

\[ y = \frac{8}{L} \]

\( z \) is the output of the measuring instrument with time constant \( \tau_I \) at time \( t \), \( \lambda \), defined by Eq. (II-7) is the current in the circuit due to 1°C difference of temperature between the two sets of thermocouple junctions, \( \alpha \) and \( k \) are average thermal diffusivity and thermal conductivity of the thermopile of length \( L \), \( \delta \) is the thickness of the insulation over the junctions, and \( g \) is the sensitivity of the measuring instrument.

For the calibration experiments, the rate of heat release decayed exponentially with time. This is given by the first term of the expansion (II-3). In such a case, Eq. (B-1) becomes:

\[
U = \frac{\sin(M_1(1-y)) - \sin(M_1y)}{M_1 \cos M_1 (\omega_1 \tau_I - 1)} \left[ e^{-t/\tau_I} - e^{-\omega_1 t} \right]
\]

\[
\sum_{n=0}^{\infty} \frac{2(-1)^n}{2^n \pi} \left[ \sin \left( \frac{2n+1}{2} \pi (1-y) \right) - \sin \left( \frac{2n+1}{2} \pi y \right) \right] \left( e^{-t/\tau_I} - e^{-\left( \frac{2n+1}{2} \frac{\pi}{M_1} \right)^2 \omega_1 t} \right) \left[ \left( \frac{2n+1}{2} \pi \right)^2 - M_1^2 \right] \left[ \omega_1 \tau_I \left( \frac{2n+1}{2} \frac{\pi}{M_1} \right)^2 - 1 \right]
\]

\[ \text{(B-1a)} \]

To calibrate the thermopile, \( U \) is plotted as a function of time for an array of combinations of values of \( M_1 \) and \( y \). Each curve shows a peak at a time, \( t \).
The curves which have the peak at the same time \( t \), as the experimental output curve, are redrawn by changing the scale of \( U \) in such a way that the height of the peak is equal to that of the experimental output curve. The values of \( M_1 \) and \( y \) for the best fitting curve are used to calculate thermal diffusivity \( \alpha \) and the insulation thickness, \( \delta \). Thermal conductivity \( k \), is then given by:

\[
k = \left( \frac{s \lambda L}{g} \right) \left( \frac{U}{z} \right)
\]

where \( U \) is the height of the peak of the best fitting calculated curve given by Eq. (B-1a) and \( z \) is the height of the peak of the experimental curve.

In order to get meaningful results, it is necessary that the time constant \( 1/\omega_1 \) of the heat released on the surface be greater than the time constant of the measuring instrument.

The heat released on the surface as a function of time can be calculated by the same procedure as used for calibration of the thermopile. The only difference is that Eq. (B-1) has to be used instead of Eq. (B-1a) and the unknowns are \( a, \omega_1 \) and \( \omega_2 \) instead of \( k, \alpha \) and \( \delta \).

The above procedure fails for "instantaneous" heat release on the surface (time constant of the heat release very small as compared to that of the measuring instrument). In such cases, Eq. (II-17) indicates that the height of the peak of the experimental curve is proportional to the total heat released on the surface. Hence, the unknown heat release can be determined by comparing the height of the peak of experimental output curve with the one obtained for a known instantaneous heat release on the same surface.
It should be noted that during calibration, the effect of the heat losses from the surface by conduction through the surrounding medium are automatically included in the calculated value of the thermal conductivity, $k$, because the value of "$a$" actually used in Eq. (B-2) to evaluate $k$ corresponds to the heat released on the surface and not to the heat conducted away through the thermopile. So the thermopile should be recalibrated each time the thermal properties of the surrounding medium are changed.
NOTATIONS

$A$  
area of the surface of heat release (cm$^2$)

$A_1, A_2, A_3$  
cross-sectional areas of the two materials of the thermocouple and of the thermal insulation, respectively (cm$^2$)

$A_{l,m}$  
defined by Eq. (IV-3a)

$a_i$  
constant in Eq. (II-3) (cal/cm$^2$sec)

$b_{l,m}$  
defined by Eq. (IV-3b)

$C_p, C_f$  
electrical capacitance of the pyroelectric element and external circuit, respectively (farad)

$C_t$  
total electrical capacitance ($= C_p + C_f$) (farad)

$C_{l,m}$  
defined by Eq. (IV-3c)

c  
average specific heat (cal/gm°C)

$D_{l,m}$  
defined by Eq. (IV-3d)

$E, E(t)$  
E.M.F. Produced (volt)

$E_i$  
pyroelectric voltage at point "i" (volt)

$\Delta E$  
voltage across the pyroelectric element (volt)

$f$  
heat flux (cal/cm$^2$sec)

$f(t)$  
defined by Eq. (II-16)

$\Delta f$  
bandwidth, cycle/sec

$g$  
defined by Eq. (II-3) (amp/unit output of instrument)

$g_1$  
units of output produced by 1°C difference in temperature between two sets of thermocouple junctions (units output/°C)

$I, I(t)$  
current (amp)

$I_0(z)$  
modified Bessel function of zero order with imaginary argument

$I_1(z)$  
modified Bessel function of first order with imaginary argument

$j = \sqrt{-1}$  
imaginary unit
modified Bessel function of zero order with imaginary argument
modified Bessel function of first order with imaginary argument
average thermal conductivity (cal/sec. cm°C)
thermal conductivity of the two materials of the thermocouple and of the thermal insulation, respectively (cal/sec.cm°C)
Boltzmann constant (= 1.38×10⁻²³ joule/°C)
length of the probe (cm)
average Lorentz number
number of thermocouple junction
an integer; frequency in Chap. IV
constant of integration in Eq. (IV-2)
thermal conductance (cal/sec°C); pyroelectric coefficient (coulomb/cm²°C)
total heat released on the surface (cal/cm²)
pyroelectric charge produced by slice "i" (coulomb)
constant of integration in Eq. (IV-2)
charge produced by a pyroelectric element (coulomb)
rate of heat release per unit surface area at time, t
electrical resistance of the slice, i (ohm)
electrical resistance of the external circuit (ohm)
electrical resistance of the pyroelectric element (ohm)
total electrical resistance (= 1/R₁ + 1/Rₚ) (ohm)
specific electrical resistivity of the pyroelectric element (ohm cm); radial distance in Chap. IV (cm)
specific electrical resistivity of the two materials of the thermocouples (ohm cm)
measure of sensitivity (cal/sec cm² unit output)

absolute temperature (°K); temperature at any point in the thermopile relative to the temperature of heat sink (°C)

temperature difference between two sets of junctions (°C)

bulk temperature change of pyroelectric element (°C)

time (sec)

minimum detectable voltage due to electrical noise (volt)

minimum detectable energy due to thermal noise (watt)

minimum measurable energy due to noise (watt)

distance along x-axis from heat sink (cm)

thickness of the slice, i (cm)

distance along y-axis from origin (cm)

output of the measuring instrument

average thermal diffusivity, (m²/sec)

Peltier coefficient (cal/sec amp per junction); \( \beta = 1/\sqrt{2m\alpha} \)
in Chap. IV

effective thermal capacitance defined in Eq. (II-1) (cal/°C)

thermoelectric constant (volt/°C)

average density (gm/cm³)

constant in Eq. (II-3) (l/sec)

thickness of the insulation over the junctions (cm)

defined by Eq. (II-7) (amp/°C)

characteristic time constant of heat release (sec)

time constant of the measuring instrument (sec)

constant in Eq. (II-23c) (4.184)

responsivity of the device (volt/watt)
REFERENCES


6. ibid; Chapter V.


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