AN EXPERIMENTAL STUDY OF NONSTEADY-STATE THERMOELECTRIC COOLING

E. K. Iordanishvili, B. E.-Sh. Malkovich, and I. I. Khazanovich

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An experimental study is made of nonsteady-state thermoelectric cooling upon passage through a thermoelement of a current of rectilinear pulses. It is shown that the cooling process can be adequately described within the framework of a model which considers only the Peltier and Joule effects (within the volume and at the junction).

The first theoretical and experimental studies of cold thermoelements in a nonsteady-state mode were reported in [1]. It was shown that by application of rectangular pulses to a thermoelement, already investigated in steady-state conditions, additional cooling of the cold junction can be obtained. Results of an experimental investigation of a thermoelement in a nonsteady-state mode were published in [2, 3]. A series of works [1, 2, 4-7] have been devoted to a theoretical analysis of nonsteady-state thermoelectric cooling.

We present below the results obtained upon passage through a thermoelement of a single rectangular pulse (the so-called impulse mode) over a wide range of current density and pulse length. The impulse mode was investigated theoretically earlier in [2, 3], but analysis of the data was conducted without consideration of Joulean heat loss in the contact resistance of the cold junction. Moreover, the pulse length range was limited to times less than a second, which significantly lowers the practical value of the investigation. According to the conclusions of the authors of [2, 3], the impulse mode can achieve temperature differences exceeding ΔT_{opt} (ΔT_{opt} is the greatest temperature difference in the stationary mode). In as much as these results disagree both qualitatively and quantitatively with theory [2, 5, 7-9], experimental investigation of the impulse mode is of decided interest.

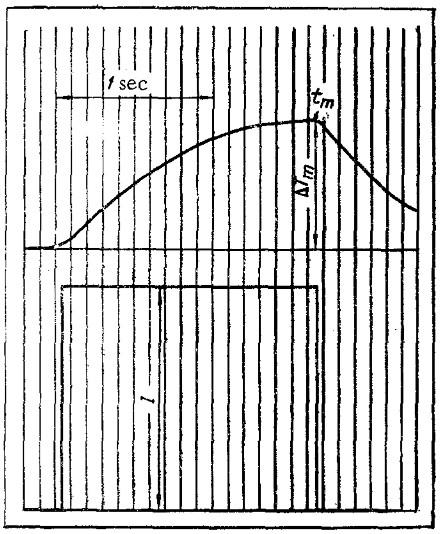

Fig. 1. Typical oscillogram of pulse mode. Upper curve is cold junction temperature; lower, thermoelement current. I = 84 A, ΔT_m = 32°, t_m = 1.6 sec.

EXPERIMENTAL METHOD

The thermoelements were prepared from zone-melted p-type (Bi_2Te_3–Sb_2Te_3) and n-type (Bi_2Te_3–Bi_2Se_3) materials. The mean parameter values of these materials at room temperature were: α = 185-215 μV/deg, σ = 900-1500 $\Omega^{-1}\cdot cm^{-1}$, $\varkappa$ = (1.6-2.0) $\cdot 10^{-2}$ V/cm $\cdot$ deg. The branch length l of a typical thermoelement was 1.6-2.6 cm, with cross section 0.28-0.5 cm^2. To reduce the thermal load at the cold junction the thermoelements were joined without a commutation membrane (butted). All measurements were conducted in a vacuum of ~$5\cdot 10^{-2}$ torr. Current was varied from 19 to 104 A, and pulse length from ~1 sec (at currents of ~100 A) to 30 sec (at currents of ~20 A). Before the start of measurements the temperature was identical

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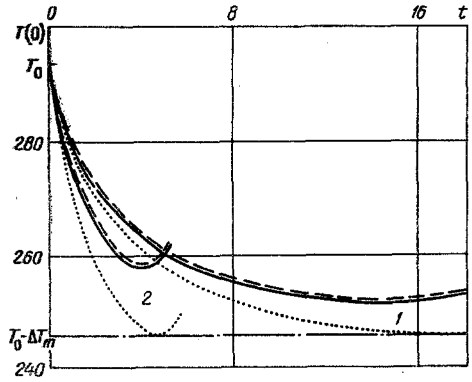

Fig. 2

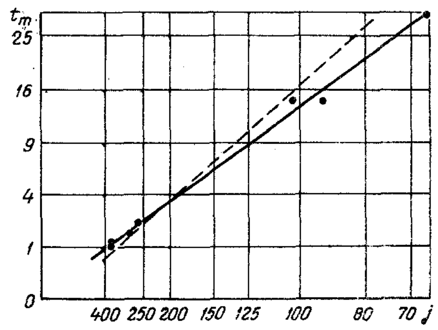

Fig. 3

Fig. 2. Thermocouple cold junction temperature as a function of time for typical thermocouple: solid curves, experiment; dashed curves, calculated, taking account of temperature dependence of Peltier coefficient [7]; dotted curves, case in which Joulean heat loss in junction is absent ($R_c = 0$); horizontal dash-dot line, minimum cold junction temperature for $R_c = 0$. 1) 29 A; 2) 54.5 A. Thermocouple cross section $S = 0.0283\ cm^2$; T, °K; t, sec.

Fig. 3. Time for attainment of maximum cooling (sec) as a function of current density (A/cm^2) (in coordinates $\sqrt{t_m}$ and 1/j) for typical thermoelement. The dashed curve is theoretical, constructed for mean values of the parameters and taking into account the temperature dependence of the Peltier effect [6, 7].

over the entire thermocouple volume. The temperature of the hot junctions was maintained constant over the course of the experiment by passage of a cooling liquid of fixed temperature through the current leads. All experiments were conducted at two hot junction temperatures: +20°C or −30°C. In the latter case, a mixture of ethanol with dry ice was used as the cooling liquid.

Cold and hot junction temperatures were measured by copper–constantan thermocouples ∅ 0.14. The thermocouple emf and current value were recorded simultaneously on an NOO4 loop oscillograph with galvanometer sensitivity of 80 μV/mm. The cold junction thermocouple signal was amplified by a factor of 3-4 before application to the galvanometer. Displacement of the galvanometer light spot by 1 mm corresponded to a cold junction temperature change of approximately 0.7°.

RESULTS AND DISCUSSION

Comparison of curves obtained at the two hot junction temperatures (+20°C and −30°C) showed that no qualitative difference in thermocouple behavior could be observed. Therefore we will present results of the study of a typical thermoelement only at a hot junction temperature of +20°C.

A typical oscillogram of the pulse mode is presented in Fig. 1, which shows the character of cold junction temperature change over time T(0, t).

As experiment showed, a minimum in the curve T(0, t) was observed only with currents of a certain magnitude (approximately twice the optimal current), and was not observed for lower currents.

It can be said that such behavior in cold junction temperature follows from an analysis of the expression for T(0, t) obtained from the thermal conductivity equilibrium equation

$$\frac{\partial T}{\partial t} = a\frac{\partial^2 T}{\partial x^2} + \frac{a}{\sigma\varkappa} j^2$$

with boundary conditions

$$\varkappa \frac{\partial T}{\partial x}\bigg|_{x=0} = \Pi j, \quad T(l, t) = T_0, \quad T(x, 0) = T_0.$$

The expression for T(0, t) has the form

$$T(0, t) = T_0 + \frac{jl^2}{\varkappa\sigma}\left(\frac{j}{2} - j_{opt}\right) + \frac{16}{\pi^3}\cdot\frac{jl^2}{\varkappa\sigma}$$

$$\times \sum_{n=0}^{\infty} \left[\frac{\pi}{2} j_{\mathrm{opt}} - \frac{(-1)^n}{2n+1} j \right] \frac{\exp\left[-(2n+1)^2 \frac{\pi^2}{4} \cdot \frac{at}{l^2}\right]}{(2n+1)^2}. \tag{1}$$

An analogous expression was obtained in [5]. Here $j_{opt} = \Pi\sigma/l$ is the optimal current density (Πj is the Peltier heat, absorbed in each branch; Π = const).

We will examine two cases: $j < (\pi/2) j_{opt}$ and $j > (\pi/2) j_{opt}$.

For $j < (\pi/2) j_{opt}$ all factors in square brackets under the summation sign are positive. In fact, if n is an odd number, all factors are positive for any j, while if n is even, all factors are positive if $j < (\pi/2) j_{opt}$. Consequently, for all t, $(\partial/\partial t) T(0, t) < 0$, i.e., the curve $T(0, t)$ approaches the straight line $T(0, \infty)$ from above (from the high temperature side) and has no minimum.

For the case $j > (\pi/2) j_{opt}$ for large t ($t \gg l^2/a$), when it is possible to consider only the first term of the summation in Eq. (1), $T(0, t) < T(0, \infty)$ and $(\partial/\partial t) T(0, t) > 0$, i.e., the curve $T(0, t)$ approaches the line $T(0, \infty)$ from below. Consequently, for $j > (\pi/2) j_{opt}$ the curve $T(0, t)$ must have a minimum.

As follows from experiment (Figs. 2 and 3), as the current is increased the time t_m at which maximum cooling is obtained decreases.

For an explanation of this result, we turn again to Eq. (1), presenting it in the form (see [9], p. 103)

$$T(0, t) = T_0 + \frac{a}{\varkappa\sigma} j^2 t - \frac{8a}{\varkappa\sigma} j^2 t \sum_{m=0}^{\infty} (-1)^m i^2 \operatorname{erfc}\left(m + \frac{1}{2}\right) \frac{l}{\sqrt{at}}$$

$$- \frac{\Pi j}{\varkappa} 2\sqrt{at} \sum_{m=0}^{\infty} (-1)^m \left[i \operatorname{erfc} m \frac{l}{\sqrt{at}} - i \operatorname{erfc}(m+1) \frac{l}{\sqrt{at}} \right]. \tag{1a}$$

We will examine the case of small t ($t \ll l^2/a$), when we may limit ourselves to terms with $m = 0$. From Eq. (1a) it follows that for such t*

$$T(0, t) = T_0 + \frac{a}{\varkappa\sigma} (j\sqrt{t})^2 - \frac{2\Pi}{\varkappa} \sqrt{\frac{a}{\pi}}\, j\sqrt{t}. \tag{2}$$

Equation (2) was obtained earlier by solution of the thermal conductivity equation for a semispace [2]. It is not difficult to show, as was done in [2], that for $j\sqrt{t} = \Pi\sigma/\sqrt{\pi a}$ on the curve $T(0, t)$ there is observed a minimum, the position of which is approximately $t_m \sim 1/j^2$. Thus, the time for attainment of minimum temperature must be inversely proportional to the square of the current density.

As is evident from Eq. (2), the value of the maximum cooling ΔT_m is independent of j and t_m, and is determined exclusively by the thermoelement parameters: $\Delta T_m = \Pi^2\sigma/\pi\varkappa$ [2, 5]. Meanwhile, the experimental data indicate that as current increases, ΔT_m decreases.

To explain this contradiction, it is necessary to take into account a supplementary factor not considered earlier in the theory – the Joulean heat dissipation in the contact resistance of the cold junction [8].†

Subtracting from the Peltier heat Πj one half the Joulean contact heat $(1/2) j^2 R_c S$ (R_c is the contact resistance of the cold thermoelement junction), we find the heat absorbed in each of the thermoelement branches: $\Pi(1-(R_c S/2\Pi) j) j$. Thus it is easy to see that in the presence of contact resistance the value of Π in all the above formulas must be replaced by $\Pi(1-(R_c S/2\Pi) j)$. As a result we have

$$\sqrt{t_m} = \frac{\Pi\sigma}{\sqrt{\pi a}} \left(\frac{1}{j} - \frac{R_c S}{2\Pi} \right) \tag{3}$$

* In the present study, the condition $t \leq 0.1\, l^2/a$ was always maintained and hence comparison of experimental data with theoretical conclusions is completely valid.

† Joulean heat dissipation in the cold thermoelement junction was first considered in [7], where the temperature dependence of Π was also partially considered (the relationship $\Pi = \alpha T$ was employed, but the thermo-emf coefficient α was considered constant).

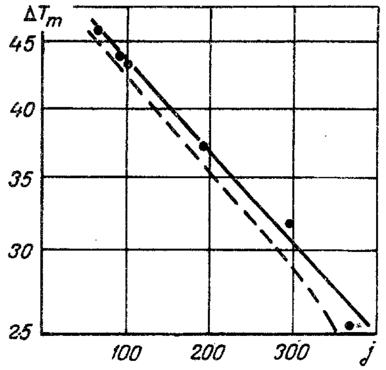

Fig. 4. Maximum cooling (°C) as a function of current density (A/cm^2) for typical thermoelement. The dashed curve is theoretical, calculated from mean parameter values taking into account the Peltier coefficient temperature dependence.

and

$$\Delta T_m = \frac{\Pi^2\sigma}{\pi\varkappa}\left(1 - \frac{R_c S\cdot}{2\Pi} j\right)^2. \tag{4}$$

Comparing Eqs. (3) and (4) with the analogous expressions for the case $R_c = 0$, it may be seen that the presence of contact resistance decreases both the time of the minimum t_m and the maximum cooling ΔT_m by a factor of $(1-(R_cS/2\Pi)j)^2$ times. As follows from Eq. (4), with current growth cooling must decrease, tending to zero as $j \to 2\Pi/R_cS$.

As follows from Eq. (3), the graph of the dependence of $\sqrt{t_m}$ on $1/j$ is a straight line, the slope of which is determined by the parameters Π, σ, and a, and does not depend on the junction contact resistance. The experimental data obtained agree with this relationship (Fig. 3).

Figure 4 also testifies to the satisfactory qualitative agreement between experiment and theory obtained by consideration of Joulean heat dissipation at the contact resistance of the cold junction.

Thus, although in our study of the nonsteady-state cooling process many factors were not studied, in particular, the Thompson effect, the temperature dependence of the parameters Π, $\varkappa$, σ, and a, various parameters of the thermoelement branches, etc., nevertheless, as comparison of experimental and theoretical data shows, the shapes of the curves $T(0, t)$, $t_m(j)$, and $\Delta T_m(j)$ can be satisfactorily described in terms of a simple model, in which only the Peltier and Joule effects are considered (in the volume and at the junction), and the temperature dependence of the thermoelement material parameters is not considered. A strict quantitative analysis of the correspondence between experimental and calculated curves necessitates the consideration of additional factors – the Thompson heat, heat transfer with surrounding medium, temperature dependence of thermoelement parameters, etc.

Figures 2-4 also present curves constructed by the formulas of [6, 7], in which the Peltier coefficient was considered a linear function of temperature: $\Pi = \alpha T$ (α = const). These curves, obtained for mean values of the parameters α, σ, and $\varkappa$, are found to be in satisfactory agreement with experimental data.

From what has been said above it follows that the limiting cooling obtained in the impulse mode ΔT_m is always less than the optimal cooling in the stationary mode ΔT_{opt}, which is in agreement with theory [2, 5-7]. For typical thermoelements studied in our work, $\Delta T_{opt} = 60°$, and the limiting value of ΔT_m (obtained for small currents $I \sim 20a \simeq 2.5\, I_{opt}$) is ~46°. It is important to note that the value of ΔT_m is obtained by a direct method – the independent measurement of the temperatures of hot and cold junctions. In connection with this it may be assumed that the higher values of impulse cooling obtained in [2, 3] are evidently the consequence of systematic error in the determination of the temperature difference between the cold and hot junctions (from the total emf appearing on the thermoelement).

NOTATION

α	is the Seebeck coefficient;
σ	is the specific electrical conductivity;
$\varkappa$	is the thermal conductivity;
Π	is the Peltier coefficient;
a	is the thermal diffusivity;
l	is the thermocouple length;
S	is the thermocouple cross-sectional area;
j	is the current density;
j_{opt}	is the optimum current density;
t	is the time;
t_m	is the time for the attainment of minimum temperature in the nonsteady-state mode;
R_c	is the thermocouple cold junction contact resistance;
T_0	is the initial thermoelement temperature;
ΔT_{opt}	is the maximum temperature difference in steady-state conditions;

ΔT_m is the maximum cooling in the pulse mode;

I is the pulse current.

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