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**HEAT AND CHARGE TRANSPORT AT
“THERMOELECTRIC MATERIAL-METAL”
BOUNDARY**

A physical model of heat and electric charge transport at pthermoelectric material-metal boundary is considered that takes into account phonon reflection at the boundary and the impact of potential barrier on charge carrier motion through the boundary. The methods for calculation of the boundary thermal and electric resistances and thermopower in conformity with said model are described. The values of these resistances and the Seebeck coefficient at the boundary between Bi – Te materials and Cu or Ni metals are evaluated and their temperature dependences are determined. It is shown that the boundary thermal resistance reaches the value of $10^8 K/W \cdot m^2$, the electric resistance is at a level of $5 \cdot 10^{11} \Omega \cdot m^2$, and the boundary thermopower due to emission is about $500 \mu V/K$. Caused by the above resistances, the thermal and electric losses at thermoelectric material-metal boundary can have a considerable impact on the parameters of microminiature power converters.

Key words: boundary thermal resistance, boundary electric resistance, boundary thermopower.

Introduction

Nowadays, widespread application of thermoelectricity is essentially constrained by high power conversion unit costs. The main contributor to the cost of thermocouple converters is thermoelectric material (TEM). The specific feature of thermoelectric power conversion is independence of its maximum efficiency of hermocouple gomety. Therefore, ideally, microminiaturization might achieve cost reduction without performance degradation.

However, a major microminiaturization problem lies in growing impact of thermal and electric losses in the zone of contact between semiconductor material of thermocouple legs and metal interconnect electrodes. These losses are caused by thermal and electric resistances of transient contact layer formed between TEM and metal at interconnection of legs. Under conditions of microminiaturization the thickness of contact layer becomes commensurate with the leg height, and thermal and electric contact resistances – with the respective leg resistances. In so doing, the impact of losses in the contact layer is increased, and the efficiency of thermoelectric converter is reduced [1, 2].

Modern technologies of creating micromodules by embedding, sputtering or chemical deposition of film electrodes with anti-diffusion microlayers on the end surface of leg [3] permit to minimize contact layer thickness and obtain virtually “perfect” (without a transient layer) TEM – metal boundary. However, a drastic difference in the physical properties of semiconductor and metal affects the motion of charge carriers and phonons through TEM – metal boundary, which defines the thermal and electric resistance and thermopower of such a “perfect” boundary [4 – 6].

To determine the values of the boundary resistances and thermopower is a relevant and important task in the design of micromodule thermoelectric power converters [6, 7].

Therefore, *the purpose* of this paper is to estimate the values of thermal and electric resistances and thermopower, to determine their temperature dependences for the boundaries between conventional *Bi – Te* based thermoelectric materials and metal interconnect electrodes for their further use in the design of thermoelectric microconverters of energy.

Physical model of heat and electric charge transport at TEM-metal boundary

Fig.1 illustrates a physical model of heat and electric charge transport by phonons and charge carriers at TEM-metal boundary.

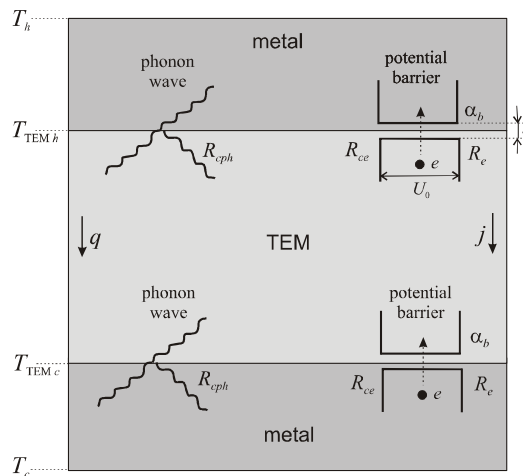


Fig. 1. Physical model of heat and charge carrier transport at TEM-metal boundary.

A perfect boundary is characterized by the mismatch of semiconductor and metal physical properties, which causes thermal and electric resistance to motion of phonons and charge carriers through the boundary.

Phonon wave carrying thermal flux partially passes through the boundary and partially is reflected. Phonon reflection is the reason for phonon component of the boundary thermal resistance R_{cph} [8, 9].

Motion of charge carriers through the boundary is prevented by potential barrier formed due to the difference in the band structures of semiconductor and metal [5]. This barrier is the reason for resistance to heat transport by carriers moving through the boundary, i.e. the electron component of thermal resistance R_{ce} .

R_{cph} and R_{ce} are the main components of the boundary thermal impedance R_c defined as follows:

$$\frac{1}{R_c} = \frac{1}{R_{cph}} + \frac{1}{R_{ce}}. \quad (1)$$

Charge carrier transport through TEM-metal boundary is accompanied by the origination of electrical boundary resistance R_e caused by their passing through potential barrier. The value of R_e depends on the barrier characteristics, i.e. its height U_0 , width d and shape, as well the mechanism (tunneling or emission) of charge carrier passage through the barrier.

Potential barrier also gives rise to origination of the boundary thermopower which is characterized by the boundary coefficient α_b . Note that α_b is not the difference in the Seebeck coefficients of adjacent materials.

Calculation of phonon component of the boundary thermal resistance

Thermoelectric material and metal are essentially different in sound velocities, phonon densities of states, the Debye temperatures and frequencies. A mismatch of these properties creates a resistance to phonon flux through the boundary. To estimate the value of phonon component of the boundary thermal resistance R_{cph} , two models are employed [10]. The first one is called acoustic mismatch model (AMM) [11]. The main approximation of AMM is that the boundary is considered to be "absolutely flat" and phonons are reflected from it in a mirror-like fashion, i.e. there is no phonon scattering at the boundary. The second, the so-called diffuse mismatch model (DMM) [9, 10] takes into account diffuse phonon scattering on the irregularities and defects of the boundary.

The assumption that phonons are not scattered is valid only on condition of rather low temperatures whereby phonon wavelength λ is much in excess of dimension b characterizing the roughness of the boundary or its deviation from the ideal flatness. Thus, for the AMM model $\lambda/b \ll 1$, and for the DMM model $\lambda/b \geq 1$ [10].

The wavelength λ can be approximately estimated by the formula [12]

$$\lambda \approx \frac{\theta_D}{T} a, \quad (2)$$

where θ_D is the Debye temperature, a is the averaged value of the size of crystal lattice unit cell.

For thermoelectric materials based on $Bi - Te$ $\theta_D \sim 160$ K, $a \sim 20 \text{ \AA}$ [13, 14]. Hence, for standard operating temperature ranges of thermoelements $T = 200 \div 300$ K the acoustic wavelength $\lambda = 1 \div 2$ nm. Thus, for the applicability of the AMM model the roughness of the boundary should be much lower than $1 - 2$ nm, which is scarcely probable. Therefore, to estimate the phonon component of thermal resistance R_{cph} of TEM – metal boundary, it is generally reasonable to use the DMM model.

A method of calculation of R_{cph} in conformity with DMM is detailed in [10]. According to the results of this work it can be written:

$$R_{cph} = \frac{\Delta T}{q}, \quad (3)$$

where $\Delta T = T_{TEM} - T_m$, T_{TEM} , T_m are the temperatures of TEM and metal, respectively. Heat flux q passing through TEM-metal boundary is related to velocities u_j of the j – th mode phonons and density-of-states function $g(\omega)$ of phonons with frequency ω by the following relationship [10, 6]:

$$q = \frac{\hbar}{4} \tau \sum_j u_{TEM,j} \int_0^\infty g(\omega) \omega \left[\frac{1}{\exp(\frac{\hbar\omega}{kT_{TEM}}) - 1} - \frac{1}{\exp(\frac{\hbar\omega}{kT_m}) - 1} \right] d\omega, \quad (4)$$

where k is the Boltzmann constant. Phonon transmission coefficient τ is determined as follows:

$$\tau = \frac{\sum_j u_{m,j}^{-2}}{\sum_j u_{TEM,j}^{-2} + \sum_j u_{m,j}^{-2}}. \quad (5)$$

Formula (5) was obtained in [10] in the approximation of $\Delta T \rightarrow 0$. In [10] it was shown that there are practically no deviations in the values of R_{cph} determined at different ΔT by formula (3), on condition of $\Delta T < 10$ K. Therefore, for calculations, R_{cph} are restricted by $\Delta T = 1$ K.

The next approximation is related to determination of phonon velocity in thermoelectric material. Due to the absence of information on the velocities of longitudinal and two transverse phonon modes in *Bi – Te* based materials, an assumption is made that these two velocities are equal [6], i.e.:

$$\sum_j u_{TEM,j}^{-2} = \frac{3}{u_{TEM}^2}. \quad (6)$$

In so doing, u_{TEM} is determined by the formula relating phonon velocity to the Debye temperature:

$$\frac{kT_D}{\hbar} = (6\pi^2 u_{TEM}^3 n)^{1/3}, \quad (7)$$

where n is the number of crystal lattice unit cells per unit volume.

For the calculation of q by formula (4) one can use the experimental density-of-states function $g(\omega)$, determined for a series of *Bi – Te* based compounds [15] or take it in the Debye approximation, i.e. in the form:

$$g_D(\omega) = \frac{\omega^2}{2\pi^2 u_{TEM}^3}. \quad (8)$$

Note that in the latter case integration in (4) is done to maximum possible value of phonon frequency, i.e. to the Debye frequency $\omega_D = kT_D/\hbar$. In [6] it is shown that the values of phonon component of resistance at TEM-metal boundary determined with the use of experimental density-of-states function and the Debye density-of-states function actually coincide.

Hence, using the ratios (3) – (8), one can determine pretty exactly the value of phonon component of TEM-metal boundary thermal resistance.

The electron component of the boundary thermal resistance, electric resistance and thermopower

As mentioned above, potential barrier accounts for the electron component of the boundary thermal resistance R_{ce} and electric boundary resistance R_e , as well as the boundary thermopower α_b . The method for calculation of these boundary impedances is proposed in [4, 5, 16]. It is based on the analogy of description of heat and electric charge transport through potential barrier and in the bulk of TEM itself.

Potential barrier at the boundary is considered to be a rectangle of height U_0 and width d . Charge carriers can overcome it by tunneling or electron emission. Maximum value of barrier width d whereby tunneling is possible is set by inequality

$$\Lambda kT \leq 1, \quad (9)$$

where $\Lambda = 1/\sqrt{E_d U_0}$, $E_d = \hbar^2/2md^2$, m is electron mass in TEM. For wider barriers the mechanism of emission is involved.

The ratios for boundary impedances in case of tunnelling were obtained in [4, 5, 16]. The electric boundary resistance R_e is determined by the formula

$$\frac{1}{R_e} = \frac{e^2 m P}{2\pi^2 \Lambda \hbar^3}, \quad (10)$$

where P is probability of tunneling through the barrier, calculated as follows [17]:

$$P = \left\{ 1 + \frac{U_0^2 \sin^2 h^2 [2m(U_0 - E)d^2/\hbar^2]^{1/2}}{4E(U_0 - E)} \right\}^{-1}, \quad (11)$$

where E is the energy of carriers and $E < U_0$. As a rule, one selects $E = U_0/2$.

The electron component of thermal boundary resistance R_{ce} is related to the electric resistance R_e by the Wiedemann-Franz law [16]:

$$\frac{1}{R_{ce}} = \frac{\pi^2}{3} \frac{T}{R_e} \left(\frac{k}{e} \right)^2. \quad (12)$$

The boundary Seebeck coefficient is equal to:

$$\alpha_b = \left(\frac{k}{e} \right) \frac{\pi^2}{3} \Lambda k T. \quad (13)$$

In case of emission, the impedances are determined using expressions [5, 18].

$$\frac{1}{R_e} = \frac{e}{k} A T \exp(-U_0/kT), \quad (14)$$

$$\frac{1}{R_{ce}} = 2 \frac{T}{R_e} \left(\frac{k}{e} \right)^2, \quad (15)$$

$$\alpha_b = \frac{1}{eT} (U_0 + 2kT), \quad (16)$$

where $A = \frac{emk^2}{2\pi^2 \hbar^3}$ is the Richardson constant for TEM.

In order to determine whether tunneling or emission take place in a specific case, one should know barrier characteristics, namely height U_0 and width d . In [19], the estimate for metal-semiconductor barrier heights $U_0 \leq 0.1$ eV was obtained. For impedance calculation it is reasonable to use maximum value of barrier height $U_0 = 0.1$ eV, as it is accepted in [6]. Barrier width is estimated from the formula [17]:

$$d = \frac{1}{2} \left(\frac{2\varepsilon\varepsilon_0 U_0}{eN} \right)^{1/2}, \quad (17)$$

where ε is dielectric permittivity of TEM, ε_0 is dielectric constant, N is charge carrier concentration in TEM.

Thus, the ratios (10) – (17) are used to estimate the values of boundary impedances characterizing charge carrier motion through potential barrier.

Results of calculation of the resistances and thermopower at TEM – metal boundary

The values of thermal and electric resistances and thermopower were estimated for the boundaries between *n*-type $Bi_2Te_{2.7}Se_{0.3}$ and *p*-type $Bi_{0.5}Sb_{0.5}Te_3$ conventional thermoelectric materials and metal connecting electrodes of *Cu* or *Ni*. Parameters of TEM and metals necessary for the calculations are listed in Table 1.

Table 1

Parameters of TEM and metals

Parameter	TEM		References
	$Bi_2Te_{2.7}Se_{0.3}$ <i>n</i> -type	$Bi_{0.5}Sb_{0.5}Te_3$ <i>p</i> -type	
Charge carrier concentration N , m^{-3}	$3 \cdot 10^{25}$	$2 \cdot 10^{25}$	[22]
Charge carrier mass m (m_0 electron mass)	$1.25m_0$	$0.6m_0$	[13, 22]
Parameters of crystal lattice hexagonal cell			[14]
a , Å	4.35	4.3	
c , Å	30.2	30.5	
Debye temperature T_D , K	157	159	
Dielectric permittivity ϵ , $K^2/J \cdot m$	98	62	
	Metal		
	<i>Cu</i>	<i>Ni</i>	
Phonon velocities			[23]
κ_{\parallel} , m/s	4760	5630	
u_{\perp} , m/s	2325	2960	

The Debye temperatures (Table 1) of $0.9Bi_2Te_3 + 0.1Bi_2Se_{0.3}Se_3$ and $0.25Bi_2Te_3 + 0.75Sb_2Te_3$ solid solutions were determined by the formula:

$$T_D = \left(\frac{x_1}{T_{D1}^3} + \frac{x_2}{T_{D2}^3} \right)^{-1/3}, \quad (18)$$

where x_i , T_{Di} is molar content and the Debye temperature of solution components (Table 2), respectively.

The dielectric permittivity of solid solutions ϵ was calculated by the formula:

$$\frac{\epsilon - 1}{\epsilon + 2} = \sum_{i=1}^2 x_i \frac{\epsilon_i - 1}{\epsilon_i + 2}, \quad (19)$$

where ϵ_i is the dielectric permittivity of components (Table 2).

Table 2

Parameters of components of *Bi – Te* based solid solutions

Параметр	<i>Bi₂Te₃</i>	<i>Sb₂Te₃</i>	<i>Bi₂Se₃</i>	Ссылки
Debye temperature T_{Di} , K	155.5	160	180	[13]
Dielectric permittivity ϵ_i , K ² /J·m	100	55	80	[6.24]

Formulae (18) and (19) have been derived on the basis of formula for heat capacity of solids at low temperatures and the Clausius-Mossotti formula cited in [20].

The calculated properties of TEM necessary for estimation of thermal and electric boundary resistances are given in Table 3.

Table 3

Calculated TEM properties

Parameter	<i>Bi₂Te_{2.7}Se_{0.3}</i> <i>n</i> -type	<i>Bi_{0.5}Sb_{0.5}Te₃</i> <i>p</i> -type
Debye frequency ω_D , rad/s	$2.06 \cdot 10^{13}$	$2.08 \cdot 10^{13}$
Number of unit cells per unit volume n , m ⁻³	$6.1 \cdot 10^{27}$	$6.14 \cdot 10^{27}$
Phonon velocity u_{TEM} , m/s	$2.9 \cdot 10^3$	$2.91 \cdot 10^3$
Phonon transmission coefficient		
$\tau_{TEM \rightarrow Cu}$	0.537	0.539
$\tau_{TEM \rightarrow Ni}$	0.421	0.424
Potential barrier width d , nm	3.0	2.93
Λ , J ⁻¹	$3.39 \cdot 10^{20}$	$2.29 \cdot 10^{20}$
$\Lambda \cdot kT$	1.4	0.95

From the data of Table 3 it follows that tunneling condition (9) for charge carriers is not met. Therefore, ultimate impedances were determined for the emission model of potential barrier. The results of calculation of phonon and electron components and thermal boundary impedance at $T = 300K$ are presented in Table 4.

Table 4

Design values of thermal resistance of TEM – metal boundaries at $T = 300K$

Boundary	<i>n-Bi₂Te_{2.7}Se_{0.3}/Cu</i>	<i>n-Bi₂Te_{2.7}Se_{0.3}/Ni</i>	<i>p-Bi_{0.5}Sb_{0.5}Te₃/Cu</i>	<i>p-Bi_{0.5}Sb_{0.5}Te₃/Ni</i>
Phonon component $R_{c_{ph}}$, K·m ² /W	$1.04 \cdot 10^{-8}$	$1.32 \cdot 10^{-8}$	$1.01 \cdot 10^{-8}$	$1.29 \cdot 10^{-8}$
Electron component R_{c_e} , K·m ² /W	$0.21 \cdot 10^{-5}$	$0.21 \cdot 10^{-5}$	$0.43 \cdot 10^{-5}$	$0.43 \cdot 10^{-5}$
Thermal impedance R_c , K·m ² /W	$1.03 \cdot 10^{-8}$	$1.31 \cdot 10^{-8}$	$1.01 \cdot 10^{-8}$	$1.29 \cdot 10^{-8}$

The temperature dependences of these resistances are shown in Figs. 2 – 5.

From the obtained data it follows that electron component which is essentially temperature dependent (Fig. 3) has little effect on thermal resistance value. As a result, thermal boundary resistance is practically independent of temperature (Fig. 4). The value of thermal resistance at TEM – copper boundary reaches the values close to 10^{-8} K·m²/W, and at nickel boundary – $1.3 \cdot 10^{-8}$ K·m²/W.

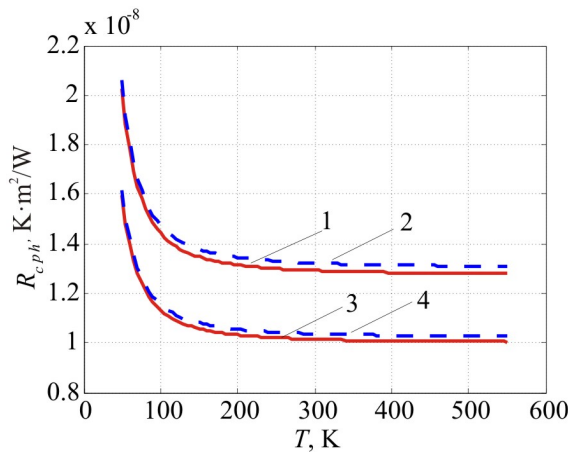


Fig. 2. Temperature dependences of the phonon component of thermal resistance at the boundaries: 1 – $p\text{-Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3/\text{Ni}$, 2 – $n\text{-Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}/\text{Ni}$, 3 – $p\text{-Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3/\text{Cu}$, 4 – $n\text{-Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}/\text{Cu}$.

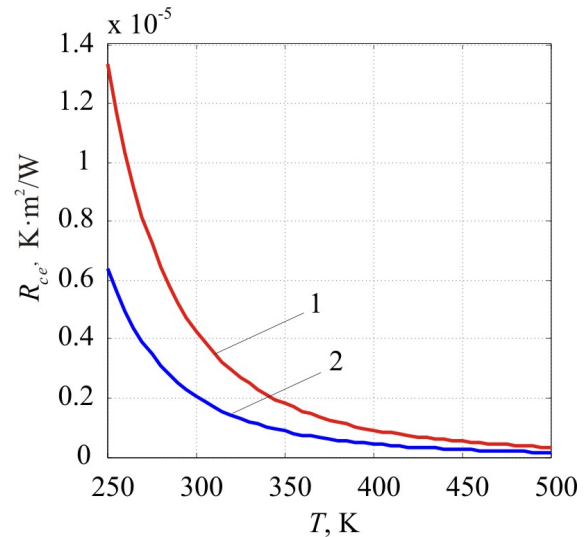


Fig. 3. Temperature dependences of the electron component of thermal resistance at TEM-metal boundaries: 1 – TEM $p\text{-Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$, 2 – TEM $n\text{-Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$.

The calculated temperature dependences of the electric boundary resistance arising due to charge carrier emission are shown in Fig. 5. This resistance does not exceed the value of $5 \cdot 10^{-11}$ $\Omega \cdot \text{m}^2$, which is an order lower than the resistance of contact layer formed in case of imperfect TEM-metal boundary and, as a rule, is about $10^{-9} \div 5 \cdot 10^{-10}$ $\Omega \cdot \text{m}^2$ [1, 2, 21].

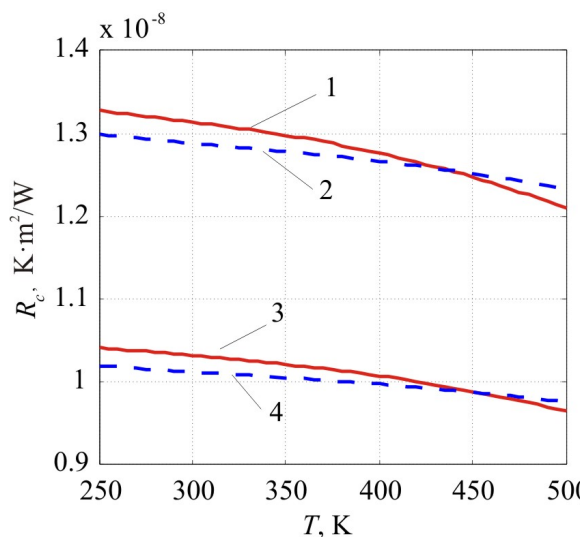


Fig. 4. Temperature dependences of the thermal resistance at the boundaries: 1 – $n\text{-Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}/\text{Ni}$, 2 – $p\text{-Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3/\text{Ni}$, 3 – $n\text{-Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}/\text{Cu}$, 4 – $p\text{-Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3/\text{Cu}$.

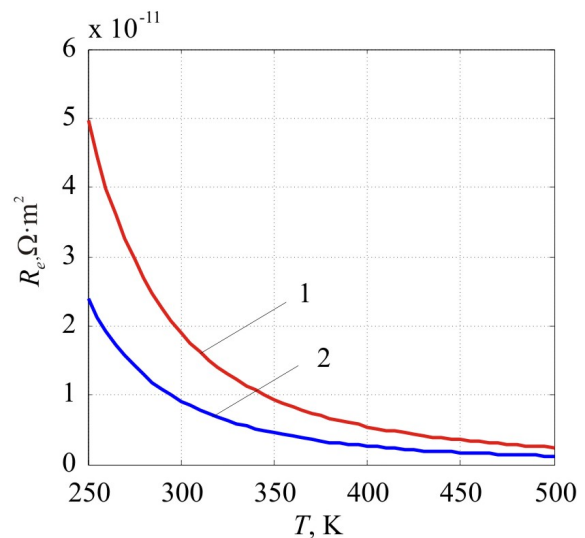


Fig. 5. Temperature dependences of the electric resistance at TEM-metal boundaries: 1 – $p\text{-Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ TEM; 2 – $n\text{-Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ TEM.

Apparently, such value of electric boundary resistance in thermoelements can affect the efficiency factors of microminiature modules for coolers and generators.

Temperature dependence of the boundary Seebeck coefficient $|\alpha_b|$ is shown in Fig. 6.

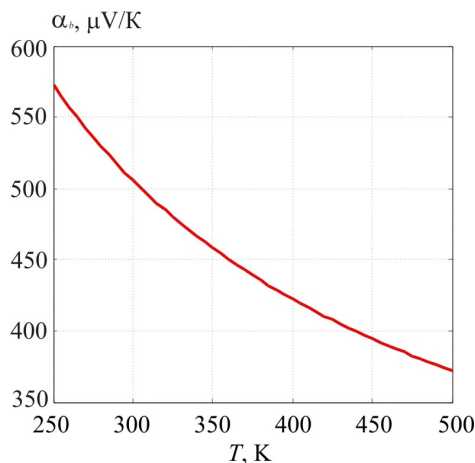


Fig.6. Temperature dependence of electron emission thermopower at TEM-metal boundary.

Note that under conditions of validity of the barrier emission model, the absolute value of the Seebeck coefficient for *n*- or *p*-type TEM – metal boundaries is the same and reaches the value of $|\alpha_b| \approx 500 \mu\text{V/K}$ at $T = 300 \text{ K}$. In [6] it is shown that for *n*-type TEM – metal boundaries α_b should be considered to be positive, and for *p*-type TEM – metal boundaries – negative.

By definition, the Seebeck coefficient of thermoelectric leg (Fig. 1) is determined as follows:

$$\alpha = \frac{\Delta\varphi}{T_h - T_c}, \quad (20)$$

where $\Delta\varphi$ is potential difference between the hot and cold sides of the leg taking into account the Seebeck effect in material of the leg itself and the thermopower of the boundaries, i.e.:

$$\Delta\varphi = \alpha_b(T_h - T_{TEMh}) + \alpha_{TEM}(T_{TEMh} - T_{TEMc}) + \alpha_b(T_{TEMc} - T_c), \quad (21)$$

where α_{TEM} is the Seebeck coefficient of TEM. Then, according to (20), we obtain:

$$\alpha = \alpha_b + (\alpha_{TEM} - \alpha_b) \frac{T_{TEMh} - T_{TEMc}}{T_h - T_c}, \quad (22)$$

From (22) it follows that $\alpha \approx \alpha_{TEM}$, as long as $T_{TEMh} - T_{TEMc} \approx T_h - T_c$. Thus, the thermopower of TEM-metal boundary has no essential impact on the thermoelement efficiency.

Conclusions

1. The methods of calculation of thermal and electric resistances and thermopower arising under heat and charge transport through TEM – metal boundary are considered. The values of thermal and electric resistances and thermopower at the boundaries between *n*-type $Bi_2Te_{2.7}Se_{0.3}$ and *p*-type $Bi_{0.5}Sb_{0.5}Te_3$ materials and *Cu* or *Ni* metals are evaluated.
2. It is established that thermal boundary resistance is about $10^{-8} \text{K/W}\cdot\text{m}^2$ and depends only slightly on temperature.

3. The electric boundary resistance is due to charge carrier emission through the boundary and its values are of the order of $5 \cdot 10^{-11} \Omega \cdot \text{m}^2$.
4. It is shown that the boundary thermopower value due to emission is about $500 \mu\text{V/K}$.

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