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ON THE EFFECT OF INTERMETALLICS ON THE ELECTRICAL AND THERMAL CONTACT RESISTANCES THERMOELECTRIC MATERIAL - METAL

It is shown that the electrical and thermal contact resistances thermoelectric material (TEM) - metal in structures with anti-diffusion layers increase significantly, if transient contact layer consists of a sublayer of intermetallic and a sublayer of TEM-intermetallic composite. In a couple of bismuth telluride-nickel, $NiTe_2$ is a dominant intermetallic. With a total thickness of transient layer of bismuth telluride-nickel ditelluride of 40 μm its electrical resistance will vary in the range from $1.28\cdot10^{-6}$ to $3.46\cdot10^{-6}$ Ohm·cm^2, and thermal –in the range from 0.131 to 0.195 K·cm^2/W. Over time, this layer can grow and, for instance, with a total thickness of 200 μm its electrical contact resistance will vary in the same temperature range from $6.40\cdot10^{-6}$ to $1.73\cdot10^{-5}$ Ohm·cm^2, and thermal – in the range from 0.655 to 0.975 K · cm^2 /W. This growth significantly affects not only the consumer characteristics, but also the reliability, life and durability of thermoelectric energy converters. In addition, it is shown that nickel ditelluride - bismuth telluride composite is not a highly efficient thermoelectric material, but the dimensionless thermoelectric figure of merit of the bismuth telluride - high-conductivity metal clusters can become significantly higher. The boundary thermoelectric figure of merit of such a composite was found. Bibl. 9, Fig. 5.

Key words: contact resistance**,** nonstationary diffusion, intermetallic, thermoelectric material, doping, metallized composite, percolation threshold, nanoclusters, optimal composition of composite, boundary dimensionless thermoelectric figure of merit, high-temperature superconductivity.

Introduction

It is known that the electrical and thermal contact resistances TEM - metal have a significant impact on the consumer properties of thermoelectric energy converters, especially under conditions of miniaturization. But if contact structures, and, consequently, transient contact layers, are created by soldering, the possibility of lowering these resistances is significantly limited by the formation of intermetallics, which have significantly lower electrical conductivity and thermal conductivity than "pure" metals. It should be noted that nickel itself diffuses into bismuth telluride and alloys based on it relatively slowly [1]. On the other hand, during the manufacture and aging of contact structures, tellurium intensively diffuses into nickel, forming due to thermally activated reactions at the interface, layers of nickel telluride, among which nickel ditelluride $NiTe_2$ dominates [2]. The thermoelectric properties of nickel ditelluride formed in this way have not been studied. However, the thermoelectric properties of specially prepared alloys of the *Ni-Te* system of various compositions were studied

[3, 4]. Therefore, in [3], in particular, it is noted that $NiTe_2$ at 300 K has an electrical conductivity of 7500 S/cm and a thermoEMF of about 10 μV/K. Thus, to a certain extent, its properties are similar to those of a metal.

It is the formation of this intermetallic that primarily causes a significant increase in the electrical and thermal contact resistances of transient contact layers. Moreover, intermetallic layers are able to grow under the influence of temperature, and, consequently, electric current. This leads to an increase in the electrical and thermal contact resistances. In addition, intermetallics formed as a result of interfacial reactions have significantly lower mechanical strength than alloys of the same composition. Thus, the formation of intermetallics has a negative effect not only on consumer characteristics, but also on the reliability, life and durability of thermoelectric energy converters. However, the purpose of this paper is mainly to study the temperature dependences of the electrical and thermal contact resistances of transient layers formed by intermetallics.

Calculation of contact layer structure

The distribution of intermetallic molecular clusters in TEM will be found from the equation of nonstationary diffusion which in the case of zero intensity of the source is given by:

$$\frac{\partial n}{\partial t} - D\frac{\partial^2 n}{\partial x^2} = 0\,, \tag{1}$$

where $n -$ concentration of intermetallic clusters at depth x from TEM-metal interface, $t -$ time, $D -$ diffusion coefficient which we consider independent of the coordinate, time and concentration of intermetallic clusters. For the unambiguous solution of Eq. (1) we make the following model assumptions: 1) at the beginning of the process, a thin layer of cluster atoms of thickness δ with concentration n_0 is created; 2) near the interface, concentration n_0 is maintained all the time. Under assumption 1, Eq.(1) has the following solution:

$$n(x,t) = n_0\left[\operatorname{erf}\left(\frac{x}{2\sqrt{Dt}}\right) - \operatorname{erf}\left(\frac{x-\delta}{2\sqrt{Dt}}\right)\right], \tag{2}$$

where $\operatorname{erf}(...) -$ the so-called error integral.

But in order for assumption 2 to be satisfied, a layer of thickness δ must change with time according to the law $\delta = 6\sqrt{Dt}$. It is this change that actually happens in many cases. In this case, this is a layer of intermetallic. It follows from formula (2) that the actual contact structure consists of two parts: a layer of intermetallic of thickness $6\sqrt{Dt}$ and a layer of TEM of thickness $6\sqrt{Dt}$ with interspersed clusters of intermetallic, the concentration of which decreases to zero. This coordinate distribution in the structure of total thickness $12\sqrt{Dt}$ is schematically shown in Fig.1.

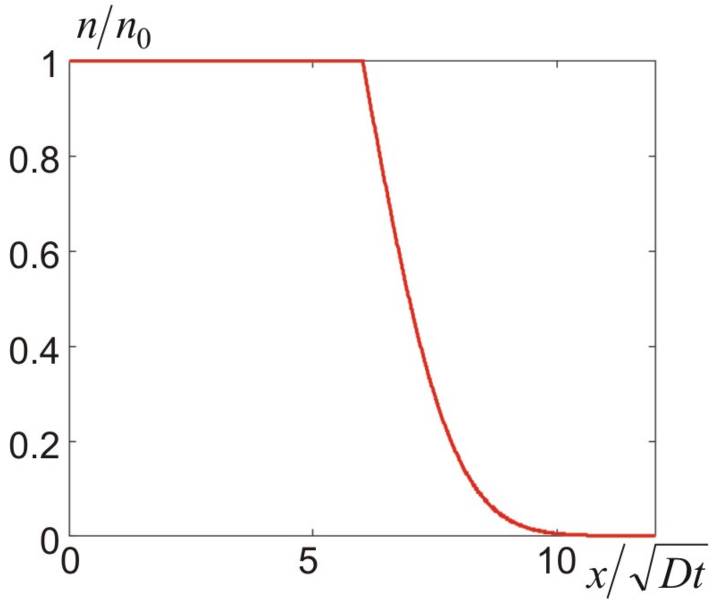

Fig.1.Distribution of intermetallic clusters in transient layer

This distribution is matched by distribution of the volumetric fraction of intermetallic in transient layer normalized to transient layer thickness:

$$\nu_i(y) = \frac{(A_i/\gamma_i)[1-\operatorname{erf}(3y)]}{(A_i/\gamma_i)[1-\operatorname{erf}(3y)]+(A_s/\gamma_s)\operatorname{erf}(3y)}, \tag{3}$$

where A_i, A_s, γ_i, γ_s – molecular masses and densities of intermetallic and TEM, respectively, $\operatorname{erf}(z)$ – the so-called error integral. Assuming by convention a cluster of intermetallic to be spherical, we define its minimum diameter. It is equal to:

$$d_{\min} = \sqrt[3]{\frac{6A_i N_{\min}}{\pi\gamma_i N_A}}, \tag{4}$$

where N_A – Avogadro's number, $N_{\min}$ – the minimum number of atoms in a cluster required to attribute to it the macroscopic properties of intermetallic as a whole. If a cluster comprises, for instance, 10^6 atoms, then such macroscopic properties can be attributed to it with an accuracy of 0.1%. Assuming $A_i = 312$, $\gamma_i = 7750\,\text{kg/m}^3$, we obtain $d_{\min} = 50.3\,\text{nm}$. Hereinafter, we will assume that the clusters have a diameter of 100 nm.

Calculation of the electrical and thermal contact resistances and discussion of the results

Taking into account that in the context of the above approach a transient layer is considered as a composite where the percolation effect occurs, its electrical conductivity σ_c and thermal conductivity κ_c will be determined as [5, 6]:

$$\sigma_c = 0.25\left\{\sigma_s(2-3\nu_i)+\sigma_i(3\nu_m-1)+\sqrt{[\sigma_s(2-3\nu_i)+\sigma_i(3\nu_i-1)]^2+8\sigma_i\sigma_s}\right\}, \tag{5}$$

$$\kappa_c = 0.25\left\{\kappa_i(2-3\nu_i)+\kappa_i(3\nu_i-1)+\sqrt{[\kappa_s(2-3\nu_i)+\kappa_i(3\nu_i-1)]^2+8\kappa_i\kappa_s}\right\}. \tag{6}$$

The thermoEMF of transient contact layer will be determined as:

$$\alpha_c = \frac{\int\limits_0^1 \{(\alpha_i/\kappa_i)v_i(y)+(\alpha_s/\kappa_s)[1-v_i(y)]\}dy}{\int\limits_0^1 \{\kappa_i^{-1}v_i(y)+\kappa_s^{-1}[1-v_i(y)]\}dy} \ . \tag{7}$$

The electrical and thermal contact resistances will be determined for the formulae:

$$r_{ce} = d\int\limits_0^1 \frac{dy}{\sigma_c}, \tag{8}$$

$$r_{te} = d\int\limits_0^1 \frac{dy}{\kappa_c}. \tag{9}$$

In addition, to these values we add the electrical and thermal contact resistances of intermetallic layers proper.

Approximation of the necessary temperature dependences of the kinetic coefficients of TEM and intermetallic on the basis of known general relations [7,8] according to the model assumptions presented in [6,9] will be carried out in the following order.

According to known thermoEMF value α_{s0} of TEM at a certain temperature T_0 from equation:

$$\alpha_{s0} = \frac{k}{e}\left[\frac{2F_1(\eta_0)}{F_0(\eta_0)}-\eta_0\right] \tag{10}$$

we determine the value η_0 of reduced chemical potential of carrier gas at this temperature.

Using the condition of constant charge carrier concentration, from the equation

$$\frac{T^{1.5}F_{0.5}(\eta)}{T_0^{1.5}F_{0.5}(\eta_0)} = 1 \tag{11}$$

we determine the temperature dependence of reduced chemical potential η in given temperature range.

From the relation

$$L_s(\eta) = \left(\frac{k}{e}\right)^2\left[\frac{3F_2(\eta)}{F_0(\eta)}-\frac{4F_1^2(\eta)}{F_0^2(\eta)}\right] \tag{12}$$

we determine the temperature dependence of TEM Lorentz number.

The temperature dependence of TEM electrical conductivity is determined as

$$\sigma_s = \sigma_{s0}\left(\frac{T_0}{T}\right)^{1.5}\frac{F_0(\eta)F_{0.5}(\eta_0)}{F_{0.5}(\eta)F_0(\eta_0)}\,. \tag{13}$$

The temperature dependence of thermal conductivity is determined as:

$$\kappa_s = \sigma_s L_s(\eta)T + \left[\kappa_{s0} - \sigma_{s0}L_s(\eta_0)T_0\right]\frac{T_0}{T}\,. \tag{14}$$

In formulae (8) – (11), $F_m(\eta)$ denote the Fermi integrals that are determined by the following relation:

$$F_m(\eta) = \int\limits_0^\infty x^m\left[\exp(x-\eta)+1\right]^{-1}dx\,. \tag{15}$$

In conformity with [3], the $NiTe_2$ intermetallic will be considered as metal, and its electrical conductivity will be assumed to be inversely proportional to temperature:

$$\sigma_i = \sigma_{i0}\left(T_0/T\right), \tag{16}$$

and its thermal conductivity, in accordance with the Wiedemann-Franz law, will be considered mainly due to free charge carriers, independent of temperature and equal to:

$$\kappa_i = \left(\pi^2k^2/3e^2\right)\sigma_{i0}T_0\,. \tag{17}$$

Moreover, taking into account the scattering of charge carriers in the intermetallic at the cluster boundaries, the values of electrical and thermal conductivity of the intermetallic "bulk sample" will be multiplied by the following correction factor:

$$K_c = 1.5\int\limits_0^1\int\limits_{-1}^1\frac{\left(d_cT/l_0T_0\right)\sqrt{1+x^2-2xy}}{\left(d_cT/l_0T_0\right)\sqrt{1+x^2-2xy}+2}x^2dydx\,. \tag{18}$$

In this formula, d_c – cluster diameter, l_0 – mean free path of electron in the intermetallic at a temperature of T_0.

Note that we use this correction factor exclusively for transient layer with clusters. We consider the intermetallic layer to be solid, and, therefore, for it we assume the correction factor equal to 1.

Results of calculations of the temperature dependences of the electrical and thermal resistances of bismuth telluride-nickel in the presence of intermetallics in transient layer are given in Figs. 2 and 3.

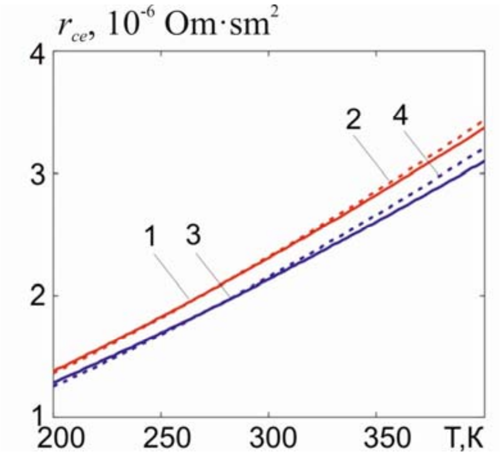

Fig.2. Temperature dependences of electrical contact resistance of transient contact layer 40 μm thick: 1,2 – prior to levelling the concentration of intermetallic clusters excluding and taking into account charge carrier scattering at the boundaries of clusters, 3,4 – after levelling the concentration of intermetallic clusters excluding and taking into account charge carrier scattering at the boundaries of clusters

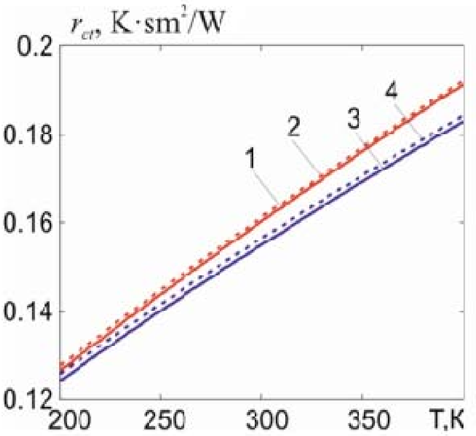

Fig.2. Temperature dependences of thermal contact resistance of transient contact layer 40 μm thick: 1,2 – prior to levelling the concentration of intermetallic clusters excluding and taking into account charge carrier scattering at the boundaries of clusters, 3,4 – after levelling the concentration of intermetallic clusters excluding and taking into account charge carrier scattering at the boundaries of clusters

When plotting, it was assumed that T_0=300 K, α_{s0}=200 μV/K, σ_{s0}=800 S/cm, κ_{s0}=1.4 W/(m·K), σ_{i0}=7500 S/cm, l_0=5 nm.

The plots show that for the total thickness of bismuth telluride - nickel ditelluride transient layer of 40 μm its electrical contact resistance will vary in the range from $1.28 \cdot 10^{-6}$ to $3.46 \cdot 10^{-6}$ Ohm·cm^2, and thermal - in the range from 0.131 to 0.195 K · cm^2 / W. Over time, this layer can grow, and, for example, with a total thickness of 200 μm, its electrical contact resistance will vary in the same temperature range from $6.40 \cdot 10^{-6}$ to $1.73 \cdot 10^{-5}$ Ohm·cm^2, and thermal - in the range from 0.655 to 0.975 K·cm^2/W. This growth has a significant impact not only on consumer characteristics, but also on the reliability, life and durability of thermoelectric energy converters. Note also that after levelling the concentration of clusters in transient layer, the electrical and thermal contact resistances somewhat decrease in comparison with the non-uniform distribution, which is dictated by the processes of nonstationary diffusion; therefore, this levelling can be considered to be a positive factor. The scattering of charge carriers at the boundaries of clusters as a whole increases the electrical and thermal contact resistances, although only slightly.

During the calculations, we had to assume that the intermetallic in the contact layer has the same properties as the alloy of the corresponding composition. But this is a very significant approximation, because the conditions of formation of this intermetallic due to interfacial reactions in the contact layer are radically different from the conditions that occur during the direct fusion of nickel with tellurium. This difference can only lead to an increase in the electrical and thermal contact resistances compared to their calculated values.

Can nickel ditelluride-bismuth telluride composites serve as thermoelectric materials?

To answer this question, the dependence of the dimensionless thermoelectric figure of merit of the aforementioned composites at a temperature of 400 K on the volumetric content of nickel ditelluride in them was calculated, which is shown in Fig. 4.

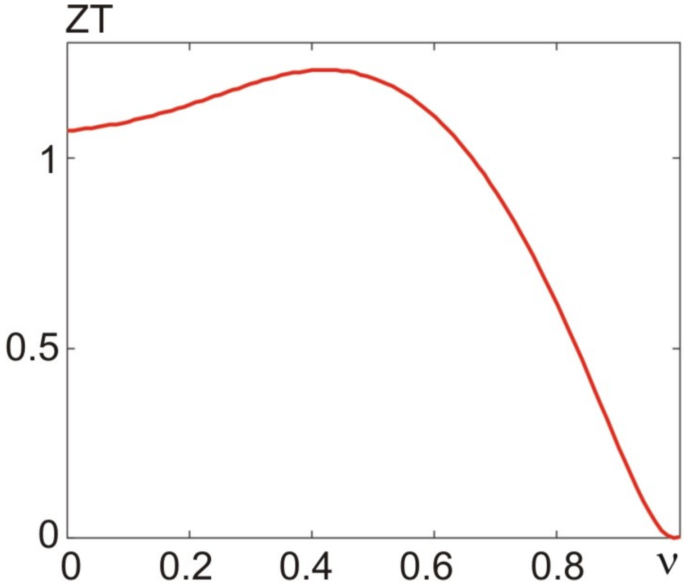

Fig.4. Dependence of the dimensionless thermoelectric figure of merit of nickel ditelluride-bismuth telluride composites at a temperature of 400 K on the volumetric content of nickel ditelluride in them

When plotting, it was assumed that the thermoEMF of nickel ditelluride is equal to 10 μV/K and does not depend on temperature. The plot shows that when the content of nickel ditelluride is approximately 50 vol.%, the thermoelectric figure of merit increases by approximately 20% compared to pure bismuth telluride. Then it rapidly decreases to the small value inherent in pure nickel ditelluride. But this is true only when our hypothetical material is really a composite and the nickel ditelluride in it has the properties of an alloy of appropriate composition. Only in this case, you can consider the feasibility of manufacturing and using such material, also taking into account the other aspects mentioned above.

On the boundary opportunities of "metallized" thermoelectric composites

Note that even at first glance a slight increase in thermoelectric figure of merit of the above hypothetical composite compared to pure TEM, shown in Fig.4, is possible mainly because the addition of highly conductive, and hence highly thermally conductive, impurity to the original TEM at a fairly large interval of the volumetric content of the impurity has relatively little effect on the thermoEMF of the composite as a whole. Therefore, assuming that the thermoEMF of an impurity is small or even equal to zero in absolute value, it makes some sense to investigate the effect of this impurity on the thermoelectric figure of merit of the composite with increasing electrical conductivity. The results of this study are shown in Fig. 5.

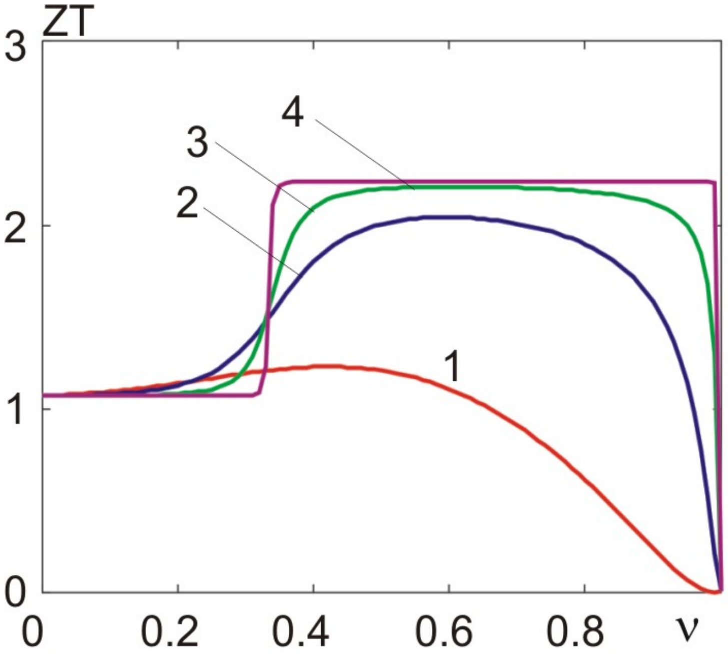

Fig.5. Dependence of thermoelectric figure of merit of metal impurity-bismuth telluride composites on the volumetric fraction of impurity at a temperature of 400 K and the value of impurity electric conductivity at 300 K in S/cm: 1 – 7500, 2 – 75000, 3 – $5 \cdot 10^5$, 4 – $5 \cdot 10^7$.

From the plots we see that with an increase in the electrical conductivity of a metal impurity, firstly, the percolation threshold becomes sharply expressed, and secondly, the maximum thermoelectric figure of merit of the “metallized” composite rather quickly tends to the maximum attainable value. Obviously, the boundary value of thermoelectric figure of merit of a "metallized" thermoelectric composite at a certain temperature T is equal to:

$$ZT_{\max} = \frac{\alpha_s^2(T)\sigma_m T}{\left(\pi^2/3\right)\left(k/e\right)^2 \sigma_m T} = \frac{3e^2\alpha_s^2(T)}{(\pi k)^2}. \tag{19}$$

It is important that this formula includes the thermoEMF of *semiconductor* TEM at a given temperature, but the electrical conductivity, thermal conductivity, hence the Lorentz number, of *metal*. It is clear that this approach correctly reflects physical situation if and only if the semiconductor and metal, when incorporated into the composite, whatever its composition, retain their inherent macroscopic values of the kinetic coefficients, including their temperature dependences.

Then, substituting, for instance, $\alpha_s(400) = 235$ μV / K, we obtain $ZT_{max} = 2.236$, which more than twice exceeds the thermoelectric figure of merit typical of traditional alloys based on *Bi(Sb) – Te(Se)* system. An even more striking result can be obtained by taking as TEM, for example, zinc antimonide, for which the thermoEMF can be considered equal to, say, 700 μV / K.

Then the maximum value of thermoelectric figure of merit of a "metallized" thermoelectric composite based on it is 19.8, while, as usual, alloys of the Zn-Cd-Sb system have more than 1 - 2 orders of magnitude lower thermoelectric figure of merit. Thus, the question arises about the technological feasibility of manufacturing such *metallized composites*, rather than just *alloyed with metal impurities materials*, which, unlike composites, obviously will not have such advantages, their stability, durability and the possibility of being used for creation of thermoelectric energy converters taking into account the specificity of their operation.

It should be noted that the reactions between metal and TEM components with the formation of intermetallics with relatively high electrical resistivity are a significant obstacle to significantly increase the thermoelectric figure of merit and efficiency of TEM both by doping them with metal impurities and by creating "metallized" thermoelectric composites. Based on the results of the study, we can formulate the following list of requirements for materials suitable for obtaining highly efficient "metallized" two-phase thermoelectric composites:

1) the semiconductor TEM must have the highest possible absolute value of thermoEMF in the operating temperature range;

2) the metal must have the lowest possible electrical resistivity and either not react with the components of the TEM, or form with them intermetallics, the electrical conductivity and thermal conductivity of which would differ as significantly as possible from the electrical and thermal conductivity of the TEM upward;

3) the thermoEMF of a metal or intermetallic under these conditions is not critical can be small in the absolute value.

However, even if these requirements are compatible with each other and technologically feasible, the composite must still have such thermal and mechanical properties and such compatibility with other materials that would allow it to be used to create specific devices.

In connection with all of the above, the reader may ask what will happen if, instead of metal impurities, specially made granules based on high-temperature superconducting ceramics are used. At this stage, the author is not ready to give a well-argued answer to such a question. There are two reasons for this. The first is that superconductivity, for example, at room temperature has not yet been achieved. The second reason is that the traditional concepts of thermoelectric figure of merit and efficiency of TEM, no matter how “good” or “bad” it is, have a certain meaning, not least because the charge transfer in it is described by Ohm's law. And in the superconducting state, Ohm's law is applicable only to the "normal", that is, not the superconducting part of the material. The electrodynamics of the superconducting part is fundamentally different. And, as can be seen from Fig. 5, there is no urgent need for unlimited electrical conductivity of the "metal" part of the composite. On the other hand, superconducting ceramics, as a component of TEM, may be useful in creating high-quality cooling materials and devices, especially miniature ones, for cryogenic temperatures, but this issue requires special thorough research, which is not the subject of this paper.

Conclusions

1. It is shown that the formation of intermetallics in the TEM-metal transient contact layer significantly increases the corresponding thermal and electrical contact resistances.
2. Conditions of high figure of merit for two-phase "metallized" thermoelectric composites are established and requirements to materials from which it is expedient to make them are formulated.
3. In general, the boundary value of the thermoelectric figure of merit is found for “metallized” thermoelectric composites.

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ПРО ВПЛИВ ІНТЕРМЕТАЛІДІВ НА ЕЛЕКТРИЧНИЙ ТА ТЕПЛОВИЙ КОНТАКТНІ ОПОРИ ТЕРМОЕЛЕКТРИЧНИЙ МАТЕРІАЛ – МЕТАЛ

Показано, що електричний та тепловий контактні опори термоелектричний матеріал (ТЕМ) метал у структурах з антидифузійними шарами істотно зростають, якщо перехідний контактний шар складається з підшару інтерметаліду та підшару композиту ТЕМ – інтерметалід. У парі телурид вісмуту – нікель таким домінантним інтерметалідам є $NiTe_2$*. За загальної товщини перехідного шару телурид вісмуту-дітелурид нікелю рівної 40 мкм електричний контактний опір змінюється в інтервалі від* $1.28 \cdot 10^{-6}$ *до* $3.46 \cdot 10^{-6}$ *Ом·см*^{2}*, а тепловий – в інтервалі від 0.131 до 0.195 К·см*^{2}*/Вт. З часом цей шар може рости, і, наприклад, за загальної товщини 200 мкм його електричний контактний опір змінюється у тому самому температурному інтервалі від* $6.40 \cdot 10^{-6}$ *до* $1.73 \cdot 10^{-5}$ *Ом·см*^{2}*, а тепловий – в інтервалі від 0.655 до 0.975 К·см*^{2}*/Вт. Це зростання істотно впливає не лише на споживчі характеристики, а й на надійність, довговічність та ресурсну стійкість термоелектричних перетворювачів енергії.*

Поряд з цим показано, що композит дітелурид нікелю – телурид вісмуту не є високоефективним термоелектричним матеріалом, але безрозмірна термоелектрична ефективність композиту телурид вісмуту – висоелектропровідні металеві кластери може стати істотно більшою. Знайдено граничну безрозмірну термоелектричну ефективність такого композиту. Бібл. 9 , рис. 5.

Ключові слова: контактний опір, нестаціонарна дифузія, інтерметалід, термоелектричний матеріал, легування, металізований композит, поріг перколяції, нанокластери, оптимальний склад композиту, гранична безрозмірна термоелектрична ефективність, високотемпературна надпровідність.

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О ВЛИЯНИИ ИНТЕРМЕТАЛЛИДОВ НА ЭЛЕКТРИЧЕСКОЕ И ТЕПЛОВОЕ КОНТАКТНЫЕ СОПРОТИВЛЕНИЯ ТЕРМОЭЛЕКТРИЧЕСКИЙ МАТЕРИАЛ - МЕТАЛЛ

Показано, что электрическое и тепловое контактные сопротивления термоэлектрический материал (ТЭМ) – металл в структурах с антидиффузионными слоями существенно возрастают, если переходный контактный слой состоит из подслоя интерметаллида и подслоя композита ТЭМ - интерметаллид. В паре теллурид висмута - никель таким доминантным интерметаллидом является $NiTe_2$*. При общей толщине переходного слоя теллурид висмута-дителлурид никеля равной 40 мкм в температурном интервале 200 – 400 К его электрическоеое контактное сопротивление изменяется в интервале от* $1.28 \cdot 10^{-6}$ *до* $3.46 \cdot 10^{-6}$ *Ом·см* ^{2}*, а тепловое - в интервале от 0.131 до 0.195 К·см*^{2}*/Вт. Со временем этот слой может расти, и, например, при общей толщине 200 мкм его электрическое контактное сопротивление изменяется в том же температурном* *интервале от 6.40 · 10^{-6} до 1.73 · 10^{-5} Ом·см ^{2}, а тепловой - в интервале от 0.655 до 0.975 К·см^2/Вт. Этот рост существенно влияет не только на потребительские характеристики, но и на надежность термоэлектрических преобразователей энергии. Наряду с этим показано, что композит дителлурид никеля - теллурид висмута не является высокоэффективным термоэлектрическим материалом, но безразмерная термоэлектрическая эффективность композита теллурид висмута - высоэлектропроводные металлические кластеры может стать существенно больше. Найдена предельная безразмерная термоэлектрическая эффективность такого композита. Библ. 9, рис. 5.*

Ключевые слова: контактное сопротивление, нестационарная диффузия, интерметаллиды, термоэлектрический материал, легирование, металлизированный композит, порог перколяции, нанокластеры, оптимальный состав композита, предельная безразмерная термоэлектрическая эффективность, высокотемпературная сверхпроводимость.

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