the behavior of the effective conductivity σ_e with a change in the concentration of phases occurs when the concentration of the first phase σ_1 is equal to $p_c=1/3$. At the same time, the value of the percolation threshold p_c of real composites can be different depending on their structure, which is related to their fabrication technique.

In [11], a modification of the Bruggeman – Landauer approximation was presented, which allows one to specify any percolation threshold $\tilde{p}_c$ for the problem of calculating the effective conductivity.

The objective of this work is to modify the self-consistent approximation for calculating the effective thermoelectric properties of composites (Bruggeman-Landauer approximation for thermoelectric phenomena) and to show the influence of the assigned percolation threshold on the effective properties of two-phase thermoelectric composites.

The article is organized as follows: in the first section, we consider the Bruggeman-Landauer approximation and its modification based on [11]. In the second section, the system of thermoelectric equations is rewritten in a convenient form for constructing the Bruggeman-Landauer approximation and a modification of the approximation is proposed. From the obtained results it follows that the thermoelectric figure of merit, depending on the percolation threshold, has a maximum, which is an interesting consequence for experimental verification. In the third section, we present the calculation of effective properties in the "anomalous" case, where an unusual behavior of the effective conductivity is observed.

Modification of the Bruggeman-Landauer approximation in the effective conductivity problem

The Bruggeman-Landauer approximation can be written as

$$\frac{\sigma_e - \sigma_1}{2\sigma_e + \sigma_1}p + \frac{\sigma_e - \sigma_2}{2\sigma_e + \sigma_2}(1-p) = 0 \,, \tag{1}$$

where σ_1 is the first phase conductivity, σ_2 is the second phase conductivity.

The effective conductivity obtained from solution (1) describes well the entire concentration range. With greater inhomogeneity, at $\sigma_1/\sigma_2 \gg 1$ close to concentration $p=p_c=1/3$ there is a sharp change in the concentration behavior of σ_e, which qualitatively describes the percolation behaviour (an analogue of the second-order phase transition). Naturally, such a "simple" approximation as (1) cannot completely describe percolation laws. For example, the critical indices σ_e close to p_c obtained from (1) [1, 11]. Their numerical value $t=q=1$ differs from the percolation ($t=2$ and $q=0.73$).

According to modification [11], the Bruggeman-Landauer approximation (1) is replaced by

$$\frac{\dfrac{\sigma_e - \sigma_1}{2\sigma_e + \sigma_1}}{1 + c(p,\tilde{p}_c)\dfrac{\sigma_e - \sigma_1}{2\sigma_e + \sigma_1}}p + \frac{\dfrac{\sigma_e - \sigma_2}{2\sigma_e + \sigma_2}}{1 + c(p,\tilde{p}_c)\dfrac{\sigma_e - \sigma_2}{2\sigma_e + \sigma_2}}(1-p) = 0 \,, \tag{2}$$

where $c(p,\tilde{p}_c)$, the Sarychev-Vinogradov term, is of the form

$$c(p,\tilde{p}_c) = (1 - 3\tilde{p}_c)\left(\frac{p}{\tilde{p}_c}\right)^{\tilde{p}_c}\left(\frac{1-p}{1-\tilde{p}_c}\right)^{1-\tilde{p}_c} \,, \tag{3}$$

and $\tilde{p}_c$ is the preassigned percolation threshold.

According to (2), $\sigma_e(p)$ at $h=\sigma_1/\sigma_2 \to 0$ close to $\tilde{p}_c$ has the same exponential behavior, as in the standard Bruggeman-Landauer approximation (1)

$$\sigma_e(p) \sim \sigma_1\left(p - \tilde{p}_c\right)^t, \ \sigma_2 = 0, \ p > p_c,$$

$$\sigma_e(p) \sim \sigma_2\left(\tilde{p}_c - p\right)^{-q}, \ \sigma_1 = \infty, \ p < p_c, \tag{4}$$

where critical indices $t=1$ and $q=1$.

Modification of the Bruggeman-Landauer approximation for thermoelectric phenomena

In the case of thermoelectric phenomena we write the local relation between the electric current j, heat flux density q, temperature gradient ∇T and electric field strength E in the form [8, 12]

$$\mathbf{j} = \sigma\mathbf{E} + \sigma\alpha(-\nabla T),$$

$$\frac{\mathbf{q}}{T} = \sigma\alpha\mathbf{E} + \kappa\frac{1+ZT}{T}(-\nabla T), \tag{5}$$

where σ, κ are electric conductivity and thermal conductivity, α is differential thermoEMF, and

$$ZT = \frac{\sigma\alpha^2}{\kappa}T, \tag{6}$$

thermoelectric figure of merit (the Ioffe number) Z multiplied by temperature.

Local kinetic coefficients σ, κ, α are coordinate-dependent and in the case of a two-phase composite take on the values σ_1, κ_1, α_1 in the first phase and σ_2, κ_2, α_2 - in the second.

The properties of composite as a whole are assigned by the effective kinetic coefficients relating by definition the volume average "currents" – electric $\mathbf{j}$ and thermal $\mathbf{q}$ to "forces" – electric field $\mathbf{E}$ and temperature gradient ∇T

$$\langle\mathbf{j}\rangle = \sigma_e\langle\mathbf{E}\rangle + \sigma_e\alpha_e\langle-\nabla T\rangle,$$

$$\frac{\langle\mathbf{q}\rangle}{T} = \sigma_e\alpha_e\langle\mathbf{E}\rangle + \kappa_e\frac{1+Z_eT}{T}\langle-\nabla T\rangle, \tag{7}$$

where

$$Z_e = \frac{\sigma_e\alpha_e^2}{\kappa_e}.$$

Systems (5) and (7) can be written in matrix form, convenient for further consideration

$$\begin{pmatrix} \mathbf{j} \\ \langle\mathbf{q}\rangle/T \end{pmatrix} = \begin{pmatrix} \sigma & \sigma\alpha \\ \sigma\alpha & \kappa\dfrac{1+ZT}{T} \end{pmatrix}\begin{pmatrix} \langle\mathbf{E}\rangle \\ \langle-\nabla T\rangle \end{pmatrix}. \tag{8}$$

Note that in this record, as it must be according to Onsager's principle [12, 13], the matrix of kinetic coefficients is symmetric. Similarly, for the effective values

$$\begin{pmatrix} \mathbf{j} \\ \dfrac{\langle \mathbf{q} \rangle}{T} \end{pmatrix} = \begin{pmatrix} \sigma_e & \sigma_e \alpha_e \\ \sigma_e \alpha_e & \kappa_e \dfrac{1 + Z_e T}{T} \end{pmatrix} \begin{pmatrix} \langle \mathbf{E} \rangle \\ \langle -\nabla \mathrm{T} \rangle \end{pmatrix}. \tag{9}$$

Introduce a generalized current $\mathbf{i}$ and a generalized force $\mathbf{e}$

$$\mathbf{i} = \begin{pmatrix} \mathbf{j} \\ \langle \mathbf{q} \rangle / T \end{pmatrix}, \quad \mathbf{e} = \begin{pmatrix} \langle \mathbf{E} \rangle \\ \langle -\nabla \mathrm{T} \rangle \end{pmatrix}, \tag{10}$$

which are interrelated by the matrix of local kinetic coefficients $\hat{\Omega}$

$$\mathbf{i} = \hat{\Omega} \mathbf{e}, \qquad \hat{\Omega} = \begin{pmatrix} \sigma & \sigma\alpha \\ \sigma\alpha & \kappa \dfrac{1 + ZT}{T} \end{pmatrix}, \tag{11}$$

and similarly for volume average currents and forces

$$\langle \mathbf{i} \rangle = \hat{\Omega}_e \langle \mathbf{e} \rangle. \tag{12}$$

In matrix notation, a self-consistent approximation of thermoelectric problem (analogue of the Bruggeman-Landauer equation) can be written as [2]

$$\frac{\hat{\Omega}_e - \hat{\Omega}_1}{2\hat{\Omega}_e + \hat{\Omega}_1} p + \frac{\hat{\Omega}_e - \hat{\Omega}_2}{2\hat{\Omega}_e + \hat{\Omega}_2} (1 - p) = 0, \tag{13}$$

where expressions of the type $1/\left(2\hat{\Omega}_e + \hat{\Omega}_1\right)$ are understood as multiplying on the right by the inverse matrix.

Here, we (similarly to [11]) propose the following modification of a self-consistent approximation (13) for thermoelectric problem

$$\frac{\dfrac{\hat{\Omega}_e - \hat{\Omega}_1}{2\hat{\Omega}_e + \hat{\Omega}_1}}{1 + c(p, \tilde{p}_c) \dfrac{\hat{\Omega}_e - \hat{\Omega}_1}{2\hat{\Omega}_e + \hat{\Omega}_1}} p + \frac{\dfrac{\hat{\Omega}_e - \hat{\Omega}_2}{2\hat{\Omega}_e + \hat{\Omega}_2}}{1 + c(p, \tilde{p}_c) \dfrac{\hat{\Omega}_e - \hat{\Omega}_2}{2\hat{\Omega}_e + \hat{\Omega}_2}} (1 - p) = 0. \tag{14}$$

Note at once that at $\tilde{p}_c = 1/3$ Eq.(14) goes over to standard approximation (13).

Fig.1 shows the concentration dependences of σ_e, κ_e, α_e, $Z_e T$ for various $\tilde{p}_c$.
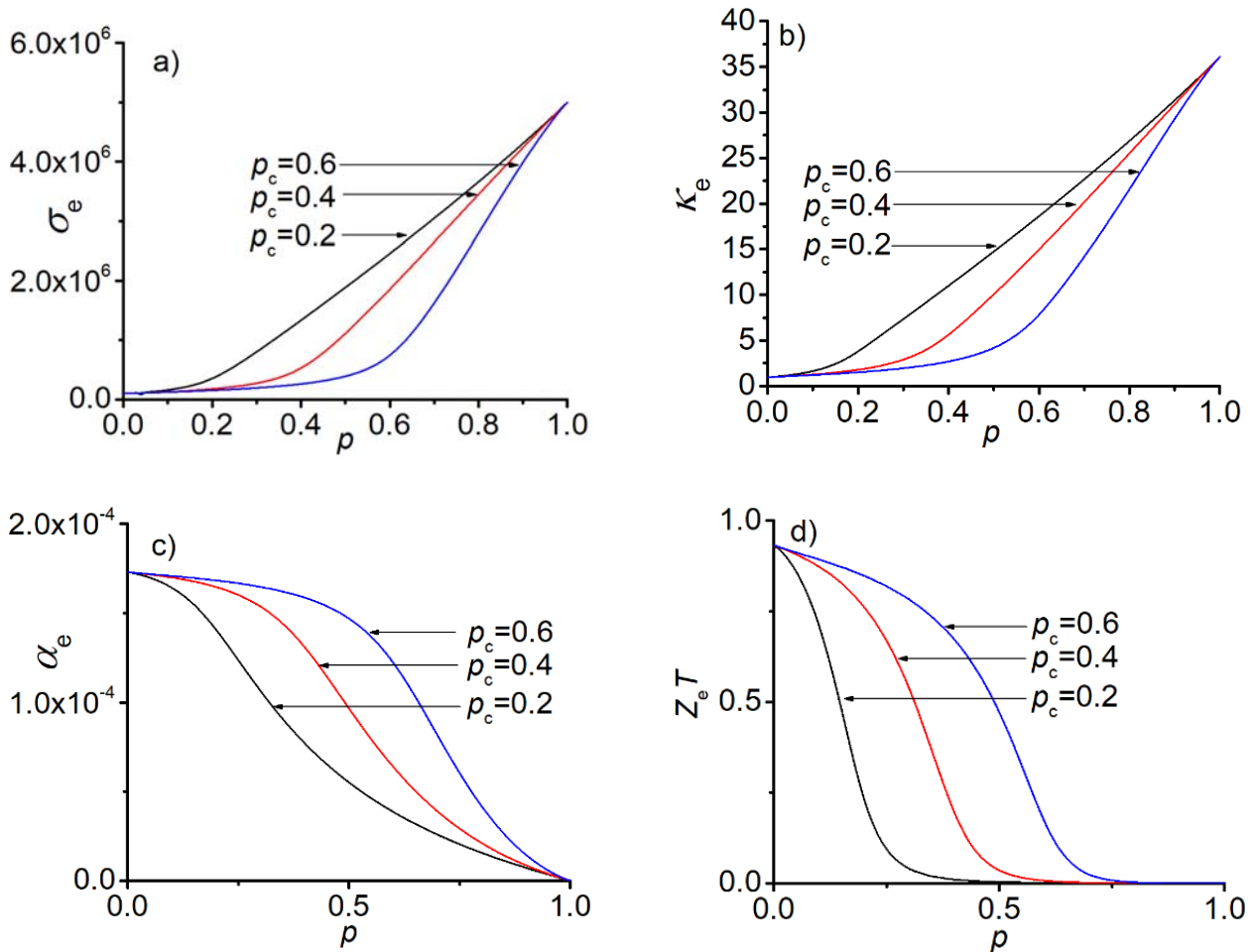
*Fig. 1 Concentration dependences of effective conductivity, thermal conductivity, thermoEMF
and figure of merit. The values of coefficients in the phases were selected [15]:
for the first phase – $\sigma_1=5\cdot10^6$ Ohm^{-1}m^{-1}, κ_1=36.1 W/(m·K), α_1=0 V/K
for the second phase – $\sigma_2=10^5$ Ohm^{-1}m^{-1}, κ_2=0.963 W/(m·K), α_2=173 ·10^6*

As can be seen from Fig. 1, the effective conductivity increases with the addition of a better conductive phase, as it must be. Effective thermal conductivity behaves similarly. Accordingly, the effective thermoEMF decreases when a phase with a lower thermoEMF is added. It can also be seen from Fig. 1 that, for example, for effective conductivity, all lines behave identically, however, the region is shifted in which there is a sharp increase in conductivity, or, in other words, the percolation threshold is shifted. The thermoelectric figure of merit (the Ioffe number) decreases monotonically, as it must be, when the first phase with a lower thermoelectric figure of merit is added.

It can be strictly shown that in the case of $h_\sigma=\sigma_1/\sigma_2\rightarrow0$ and $h_\kappa=\kappa_1/\kappa_1\rightarrow0$, σ_e and κ_e either become equal to zero at $\sigma_2=0$ and $\kappa_2=0$ and $p\rightarrow\tilde{p}_c$, or diverge at $\sigma_1=\infty$ and $\kappa_1=\infty$ and $p\rightarrow\tilde{p}_c$.

As follows from (14), the values of effective coefficients at a given concentration p of the first phase depend on the percolation threshold of composite $\tilde{p}_c$. The closer concentration p to $\tilde{p}_c$, the more significant is this dependence, but even at lower concentrations, the difference occurs.

Fig. 2 shows the dependences of effective coefficients σ_e, κ_e, α_e, Z_eT on the value of $\tilde{p}_c$ for different values of p-concentration of the first phase.
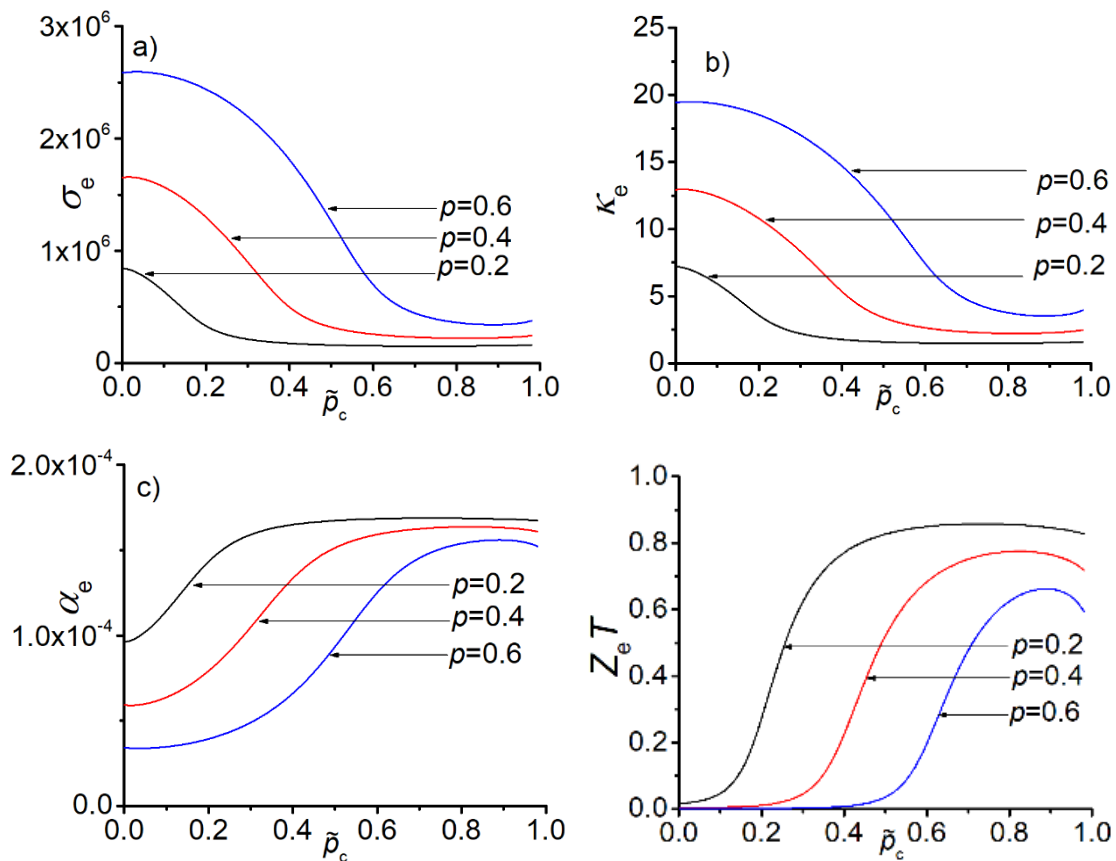
Fig. 2 Dependences of effective kinetic coefficients on the percolation threshold $\tilde{p}_c$
at a given concentration p of the first phase. The values of coefficients in the
phases were selected the same as in Fig. 1.

Fig. 2 shows that the value of effective conductivity, at a given concentration, decreases with increasing the threshold $\tilde{p}_c$. The effective thermal conductivity behaves similarly. It is also noteworthy that, at a given concentration, with increasing the threshold $\tilde{p}_c$, the thermoelectric figure of merit (the Ioffe number) also increases, but at large $\tilde{p}_c$ the maximum is noticeable and the growth changes to a small drop. It would be interesting to verify the existence of such a maximum experimentally and to determine whether this maximum is an "artifact" of our modification.

Modification of the Bruggeman-Landauer approximation for thermoelectric phenomena in the "anomalous" case

Correction (3) introduced in the Bruggeman-Landauer approximation (2) suggests that the conductivity of the first phase is greater than the conductivity of the second phase $\sigma_1 > \sigma_2$. In this case, at $\tilde{p}_c$ given in (3), the percolation threshold that can be found when solving (2) will be equal to the preassigned $p_c = \tilde{p}_c$. Subject to the inverse inequality $\sigma_2 > \sigma_1$, correction (3) should be changed.

At $\alpha_1 = \alpha_{e2} = 0$, i.e. in the absence of thermoelectric phenomena, the effective medium approximations (13) or (14) are divided into two independent – one for electric conductivity, the other – for thermal conductivity. In the case when $\sigma_1 > \sigma_2$ and $\kappa_1 > \kappa_2$ these independent equations includes the same correction $c(p, \tilde{p}_c)$. However, in the opposite case, when $\sigma_1 > \sigma_2$, but $\kappa_1 > \kappa_2$, corrections for σ_e and κ_e

should be different. In the general case, when $\alpha_1 \neq \alpha_2$, this difference remains and modification (13) (14) assumes a more complex form.

$$\frac{\dfrac{\hat{\Omega}_e - \hat{\Omega}_1}{2\hat{\Omega}_e + \hat{\Omega}_1}}{1 + \hat{C}(p,\tilde{p}_c)\dfrac{\hat{\Omega}_e - \hat{\Omega}_1}{2\hat{\Omega}_e + \hat{\Omega}_1}}\,p + \frac{\dfrac{\hat{\Omega}_e - \hat{\Omega}_2}{2\hat{\Omega}_e + \hat{\Omega}_2}}{1 + \hat{C}(p,\tilde{p}_c)\dfrac{\hat{\Omega}_e - \hat{\Omega}_2}{2\hat{\Omega}_e + \hat{\Omega}_2}}\,(1-p) = 0 ,\tag{15}$$

where

$$\hat{C}(p,\tilde{p}_c) = \begin{pmatrix} c_\sigma(p,\tilde{p}_c) & 0 \\ 0 & c_\kappa(p,\tilde{p}_c) \end{pmatrix},\tag{16}$$

and corrections $c_\sigma(p,\tilde{p}_c)$ and $c_\kappa(p,\tilde{p}_c)$ in (16), depending on the ratios σ_1/σ_2 и κ_1/κ_2 are of different form. In the case when $\sigma_1 > \sigma_2$ and $\kappa_1 < \kappa_2$ the correction $c_\sigma(p,\tilde{p}_c)$ remains the same − (3), and $c_\kappa(p,\tilde{p}_c)$ is given by

$$c_\kappa(p,\tilde{p}_c) = \left[1 - 3(1 - \tilde{p}_c)\right]\left(\frac{p}{\tilde{p}_c}\right)^{\tilde{p}_c}\left(\frac{1-p}{1-\tilde{p}_c}\right)^{1-\tilde{p}_c}.\tag{17}$$

Fig. 3 shows the concentration dependences of σ_e, κ_e, α_e, Z_eT for different values of percolation threshold.
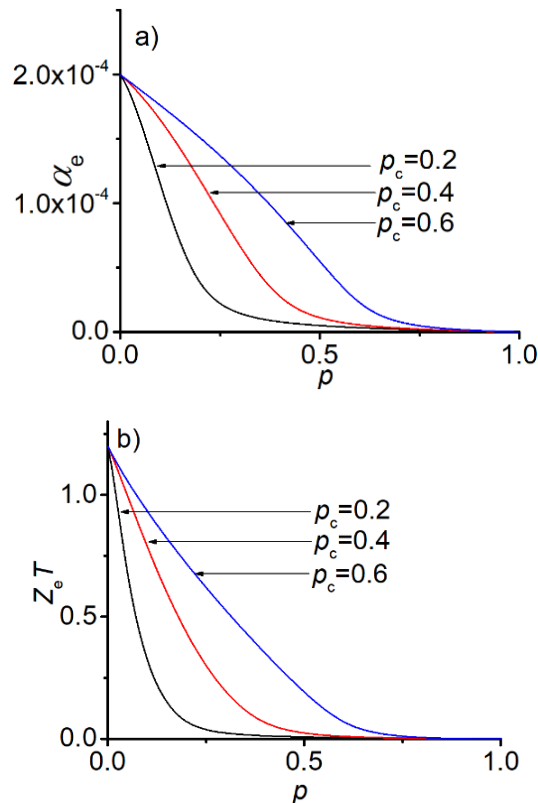

Fig. 3. Dependences of effective thermoEMF and figure of merit on the concentration
p of the first phase at a given percolation threshold $\tilde{p}_c$. The values of
coefficients in the phases were selected the same as in Fig. 1.

One of such materials for which the Wiedemann-Franz law – high conductivity, but low thermal conductivity - is violated significantly, is described in [16].

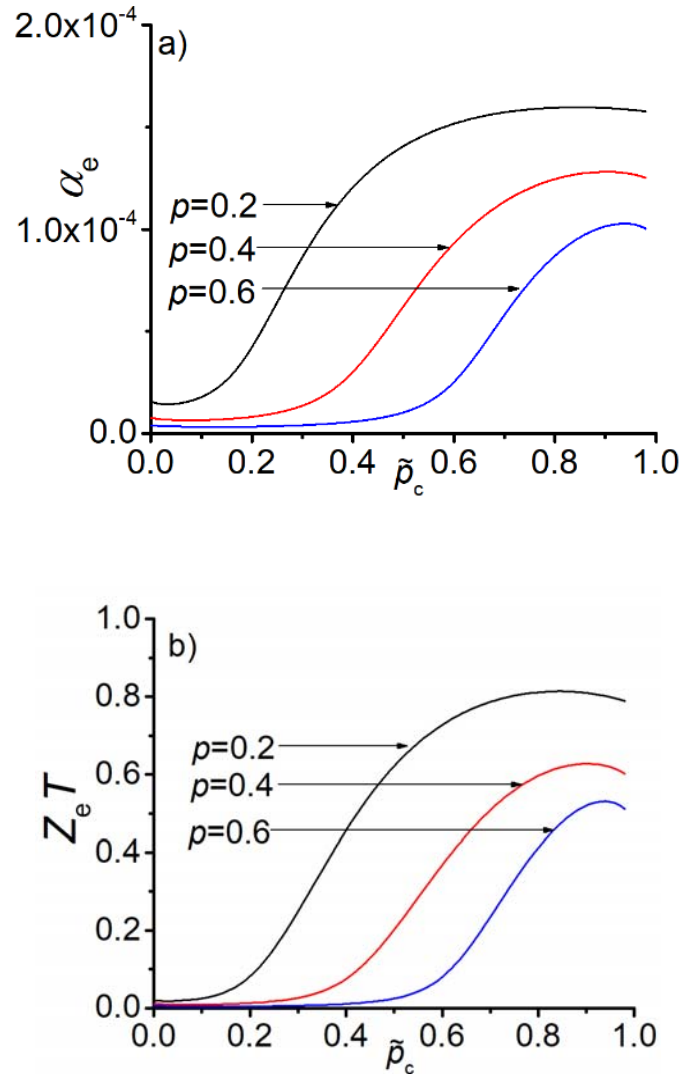Fig.4 shows the dependences of effective properties on the value of percolation threshold.

Fig. 4. Dependences of effective thermoEMF and figure of merit on the percolation threshold $\tilde{p}_c$ at a given concentration p of the first phase. The values of coefficients in the phases were elected the same as in Fig. 1.

Just as in the usual case, the effective figure of merit has a maximum.

Discussion

The figure below shows the dependences of effective conductivity and thermal conductivity on the concentration and percolation threshold in the "anomalous" case. It should be noted that effective conductivity has an unusual behavior: when a phase with good conductivity is added, effective conductivity first decreases, and then begins to grow. It seems interesting to experimentally verify whether this is a defect in the theory, or whether such conductivity is actually observed in real composites.
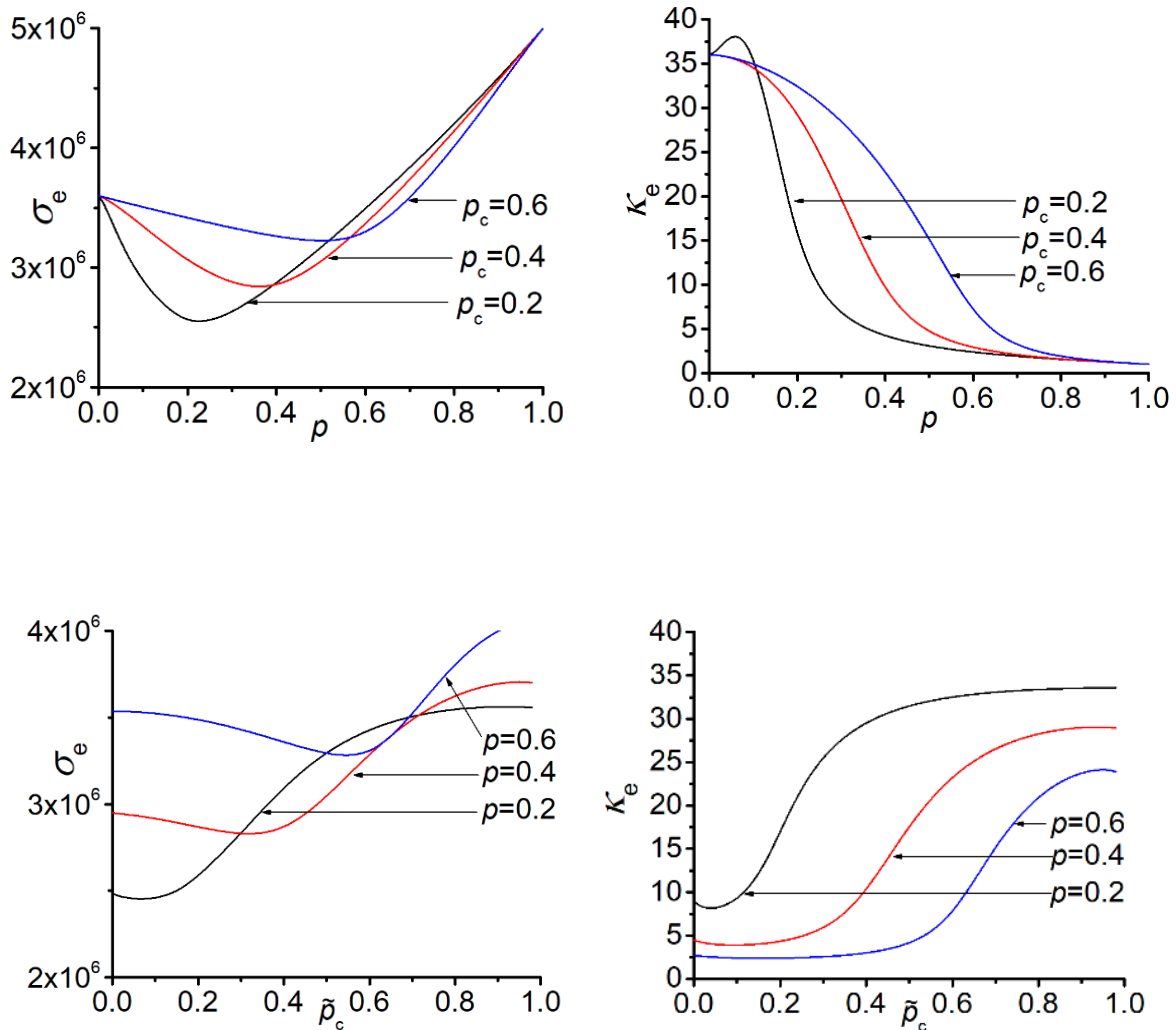
Fig. 5. Dependences of effective thermoEMF and figure of merit on the percolation

threshold $\tilde{p}_c$ at a given concentration p of the first phase.

The values of coefficients in the phases were selected the same in Fig.1.

Conclusions

This paper considers a modification of effective medium theory (self-consistency method) for thermoelectric phenomena in the case of a preassigned percolation threshold. Such a modification was first proposed in [11] for the description of galvanomagnetic phenomena and was used for the description of a series of experimental results [9-11,17]. In [18], the modification was used in the proposed approach of "mobile percolation threshold" to describe the magnetodielectric effect and the peculiarities of magnetic permeability of magnetoelastomers, in [19,20], to describe a gigantic magnetoelastic effect.

A similar modification is proposed here for a system of equations describing thermoelectric phenomena in macroscopically inhomogeneous two-phase composites.

The results obtained can be used for the description of thermoelectric properties of composites with different structures corresponding to different percolation thresholds.

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ТЕОРІЯ СЕРЕДНЬОГО ПОЛЯ ДЛЯ ТЕРМОЕЛЕКТРИЧНИХ ВЛАСТИВОСТЕЙ КОМПОЗИТНИХ МАТЕРІАЛІВ З РІЗНИМИ ПОРОГАМИ ПРОТІКАННЯ

В роботі побудовано модифіковану теорію ефективного середовища для обчислень ефективних кінетичних коефіцієнтів термоелектричних композитів з різними значеннями порогів протікання. Показано, що навіть за концентрацій поза критичною областю величина порогу істотна для визначення ефективних властивостей. Розглянуто два принципово різних випадки набору локальних властивостей композиту. Один з них, коли провідність і теплопровідність однієї з фаз одночасно більша за провідність та теплопровідність другої фази. Другий, аномальний випадок, коли електропровідність першої фази (σ_1) більша від другої, але теплопровідність першої фази менша від другої, показує незвичайну концентраційну поведінку ефективної провідності, тобто при зростанні частки добре провідної фази ефективна провідність σ_e демонструє зменшення, а не зростання як у стандартному випадку,(див. рис.1а), яке при $p \approx \tilde{p}_c$ переходить в зростання. Бібл. 5, рис. 5.

Ключові слова: термоелектрика, теорія протікання, поріг протікання, композити, ефективні властивості.

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ТЕОРИЯ СРЕДНЕГО ПОЛЯ ДЛЯ ТЕРМОЭЛЕКТРИЧЕСКИХ СВОЙСТВ КОМПОЗИТНЫХ МАТЕРИАЛОВ С РАЗЛИЧНЫМИ ПОРОГАМИ ПРОТЕКАНИЯ

В работе построена модифицированная теория эффективной среды для вычислений эффективных свойств термоэлектрических композитов с различными значениями порогов протекания. Показано, что даже при концентрациях вне критической области величина порога существенна для определения эффективных свойств. Рассмотрены два принципиально разных случая набора локальных свойств композита. Один из них, когда проводимость и теплопроводность одной из фаз одновременно больше проводимости и теплопроводности второй фазы. Второй, аномальный случай, когда электропроводность первой фазы (σ_1) больше второй, но теплопроводность первой фазы меньше второй, показывает необычные концентрационное поведение эффективной проводимости, т.е. при возрастании хорошо проводящей фазы эффективная проводимость α_e показывает падение (а не рост как в стандартном случаем, см. рис.1а), которое при $p \approx \tilde{p}_c$ переходит в рост. Библ. 5, рис. 5.

Ключевые слова: термоэлектричество, теория протекания, порог протекания, композиты, эффективные свойства.

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