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THERMOELECTRIC COMPOSITES WITH DIFFERENT PERCOLATION THRESHOLDS

A modification of the mean-field approximation is considered for describing the behaviour of effective kinetic coefficients, including for thermoelectric composites. The proposed modification makes it possible to describe randomly heterogeneous media with different percolation thresholds at arbitrary values of local kinetic coefficients. Bibl. 16, Fig. 6.

Key words: thermoelectric composites, kinetic coefficients.

Introduction

The widely used mean-field approximation (Bruggeman-Landauer approximation, self-consistent approximation) [1 – 6] has a drawback. It does not allow describing media with different thresholds. In [7], a term (Sarychev-Vinogradov term, SV-term, SVt) was introduced in the Bruggeman-Landauer approximation, which allows obtaining concentration dependences of effective galvanomagnetic coefficients for media with a predetermined percolation threshold. In [8 – 10], this approach was used to describe magneto-elastomers, and the concept of moving percolation threshold was introduced. In [11], SVt was used to describe thermoelectric phenomena in randomly heterogeneous media, but the case of "normal" and "abnormal" local kinetic coefficients [12] had to be considered separately.

This paper proposes a generalization of the SVt term for the description of kinetic phenomena in randomly heterogeneous media with any percolation threshold and for any («normal» and abnormal») values of local kinetic coefficients.

The problem of percolation threshold in the mean-field approximation in the single-flow case (by the example of effective conductivity)

For a single-flow case, for example, the case of conductivity, when there is one thermodynamic flow – electric current density $\mathbf{j}$, one thermodynamic force – electric field intensity $\mathbf{E}$, which are related by the Ohm law

$$\mathbf{j} = \sigma \mathbf{E} . \tag{1}$$

Effective conductivity σ_e is determined as

$$\langle \mathbf{j} \rangle = \sigma_e \langle \mathbf{E} \rangle , \tag{2}$$

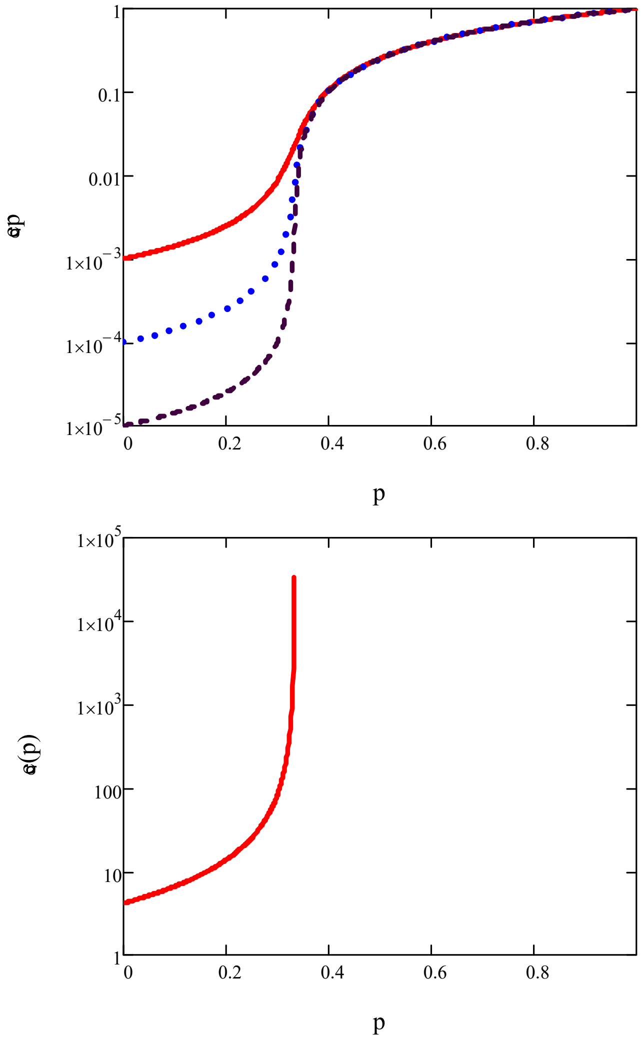
where $\langle ... \rangle = 1 / V \int ... dV$ is volume average and in the case of a two-phase medium

$$\sigma(\mathbf{r}) = \begin{cases} \sigma_1, \mathbf{r} \in O_1 \\ \sigma_2, \mathbf{r} \in O_2 \end{cases} , \tag{3}$$

The Bruggeman-Landauer approximation has the form

$$\frac{\sigma_e - \sigma_1}{2\sigma_e + \sigma_1} p + \frac{\sigma_e - \sigma_2}{2\sigma_e + \sigma_2}(1 - p) = 0 , \tag{4}$$

The concentration dependence according to (4) is given in Fig.1



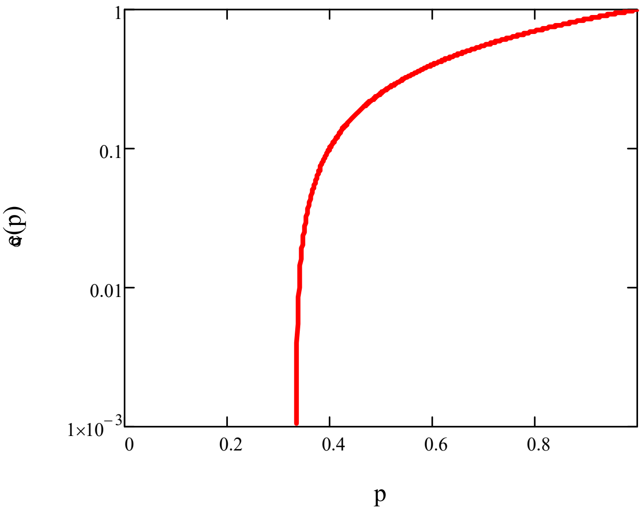


Fig. 1. Concentration behaviour of effective conductivity
a – final conductivity ratio,
b,c – percolation

Due to great heterogeneity $\sigma_1 / \sigma_2 \to \infty$, the so-called percolation concentration range $|p - p_c| \ll 1$ stands out, for which the percolation dependences are valid [1,2,4], see Fig. 1

$$\sigma_e = \begin{cases} \sigma_1 (p_c - p)^{-t}, p < p_c \\ (\sigma_1^q \sigma_2^t)^{1/t+q}, |p - p_c| \ll \Delta, \\ \sigma_2 (p - p_c)^q, p > p_c \end{cases} \tag{5}$$

where $\Delta = (\sigma_2 / \sigma_1)^{1/t+q}$.

It should be noted that percolation regularities are valid only for very large heterogeneity and in a very narrow concentration ($|p - p_c| \ll 1$) range. However, the numerical value of the percolation threshold is a characteristic of the entire concentration range and thus a characteristic of the behaviour of the effective kinetic coefficients in the entire concentration range and at any heterogeneity.

The Bruggeman-Landauer approximation is based on the calculation of fields in a solitary inclusion; it is surprising that this approximation describes the limiting behaviour of the effective kinetic coefficients quite well. In particular, at $p = p_c$ the concentration dependence of the effective conductivity $\sigma_e(p)$ has a kink. The sharper, the greater the heterogeneity. Thus, the Bruggeman-Landauer approximation describes well the concentration behaviour $\sigma_e(p)$ and can be used to describe experimental data.

There is a drawback of this approximation, the sharp transition of the effective conductivity $\sigma_e(p)$ (at $\sigma_1 / \sigma_2 \to \infty$), at the same time, the percolation threshold is always equal to $p_c = 1/3$. However, in real media [7], the percolation threshold may take on different values depending on the method of creating the composite. Despite the logic of the derivation and the simplicity of the resulting expression, the Bruggeman-Landauer approximation needs modification.

In [7], a modification was considered that allows one to set the value of percolation threshold for the case $\sigma_1 > \sigma_2$. This modification was used in the model of magnetoelastic composites with the introduction of the method of moving percolation threshold [8 – 10].

$$\frac{\dfrac{\sigma_e-\sigma_1}{2\sigma_e+\sigma_1}}{1+c(p,\tilde{p}_c)\dfrac{\sigma_e-\sigma_1}{2\sigma_e+\sigma_1}}p+\frac{\dfrac{\sigma_e-\sigma_2}{2\sigma_e+\sigma_2}}{1+c(p,\tilde{p}_c)\dfrac{\sigma_e-\sigma_2}{2\sigma_e+\sigma_2}}(1-p)=0\,, \tag{6}$$

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

$$c(p,\tilde{p}_c)=(1-3\tilde{p}_c)\left(\frac{p}{\tilde{p}_c}\right)^{\tilde{p}_c}\left(\frac{1-p}{1-\tilde{p}_c}\right)^{1-\tilde{p}_c} \tag{7}$$

A more complicated situation is observed in the mean-field approximation in the descriptions of the effective elastic properties of composites [10]. The mean-field approximation for elasticity is the Budiansky approximation [13, 14], with a large heterogeneity of elastic properties for the three-dimensional case, it gives the percolation threshold 1/2, and for the two-dimensional case – 2 /3, which contradicts the geometric considerations of the percolation structure.

Fig.2 shows the concentration dependence of the effective conductivity with regard to the Sarychev-Vinogradov term (6, 7). As can be seen from Fig.3, for a large heterogeneity ($\sigma_1 \gg \sigma_2$), the percolation threshold coincides, as it should be, with the one specified in the term $\tilde{p}_c$.

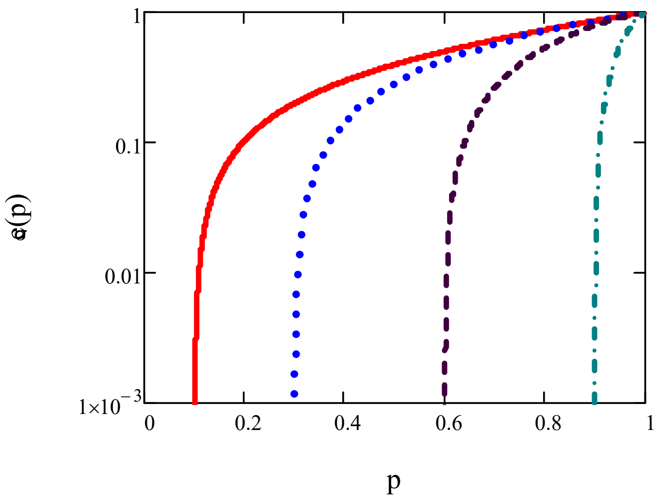


Fig. 2 Concentration behaviour of the effective conductivity according to (6, 7)

The situation is more complicated with a given value of the ratio σ_1/σ_2. The resulting threshold deviates from the one set in Svt $\tilde{p}_c$. To determine the type and magnitude of the deviation of p_c from $\tilde{p}_c$, we write down the solution of Eq.(6) in an explicit form.

$$\sigma_e\left(\sigma_1,\sigma_2,p,\tilde{p}_c\right)=\frac{1}{4}\left[2\frac{3p-1+c}{2+c}\sigma_1+2\frac{2-3p}{2+c}\sigma_2+\right.$$
$$\left.+\sqrt{4\left(\frac{3p-1+c}{2+c}\sigma_1+\frac{2-3p}{2+c}\sigma_2\right)^2+16\frac{1-c}{2+c}\sigma_1\sigma_2}\;\right] \tag{8}$$

(for ease of notation $c(p,\tilde{p}_c)$ is written as c), and from the equation that determines the inflection point,

$$\frac{d^3}{dp^3}\sigma_e\left(\sigma_1,\sigma_2,p,\tilde{p}_c\right)=0 \tag{9}$$

we find the "percolation" threshold (in which the value of p is fulfilled (9)) for a given ratio σ_1/σ_2 – Fig. 3. As can be seen, the deviation values are not very large.

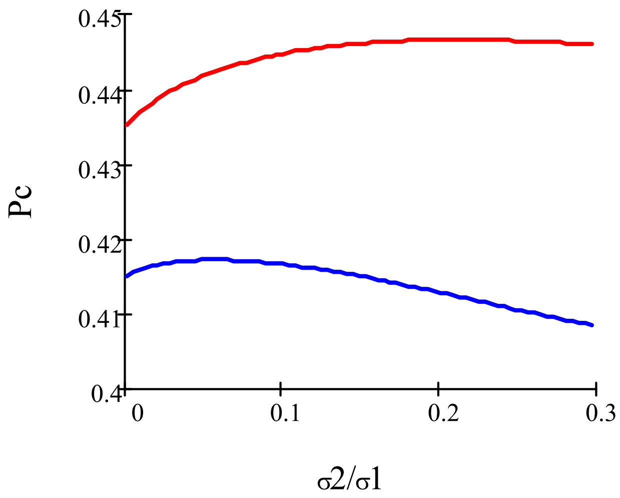
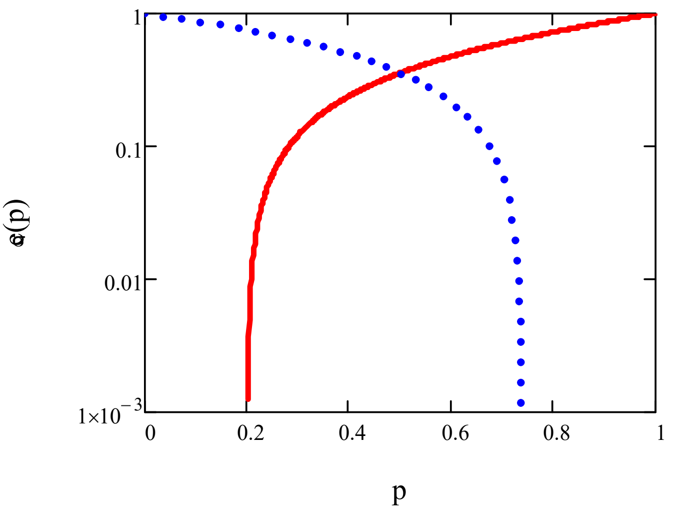


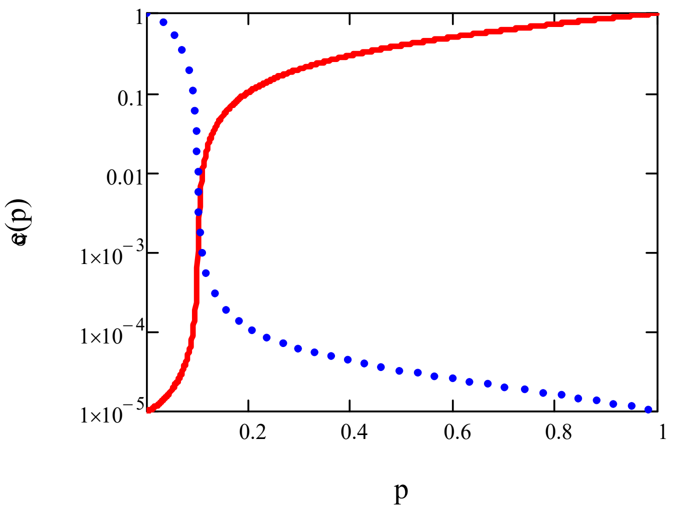
Fig.3 Percolation threshold at a given phase conductivity ratio

Term modification

A s is obvious from the direct solution of the self-consistency equation (the Bruggeman-Landauer approximation) the term works only at $\sigma_1>\sigma_2$. Thus, for instance, see Fig. 4, on setting $\tilde{p}_c=0.2$ at $\sigma_1>\sigma_2$ the obtained percolation threshold is really equal to 0.2, however, at $\sigma_1<\sigma_2$ (in both cases we have strongly inhomogeneous composite) the threshold is not 0.2, but 0.737.



a)



b)

Fig.4 Percolation thresholds
4a – incorrect case, 4б – correct case

It is necessary to modify the term so that it gives correct values of percolation threshold both at $\sigma_1 > \sigma_2$ and at $\sigma_1 < \sigma_2$.

Based on the symmetry of the behaviour of $\sigma_e(\sigma_1, \sigma_2, p)$ depending on the concentration and phase conductivity values

$$\sigma_e(\sigma_1, \sigma_2, p) = \sigma_e(\sigma_2, \sigma_1, 1-p), \tag{10}$$

and term (7), one can define the type of term for the case $\sigma_1 < \sigma_2$

$$c(p, \tilde{p}_c) = (3p-2)\left(\frac{p}{\tilde{p}_c}\right)^{\tilde{p}_c}\left(\frac{1-p}{1-\tilde{p}_c}\right)^{1-\tilde{p}_c}. \tag{11}$$

Combining these terms into one, so that at $\sigma_1 > \sigma_2$ term (7) would occur, and at $\sigma_1 < \sigma_2$ term (11) would have the form

$$c\left(p, \tilde{p}_c\right) = -\frac{1}{2} + \frac{3}{2}\left(1 - 2\tilde{p}_c\right)U\left(\sigma_1, \sigma_2\right), \tag{12}$$

where $U\left(\sigma_1, \sigma_2\right)$ function is related to sign function $\operatorname{sgn}\left(x\right)$ and has the form

$$U\left(\sigma_1, \sigma_2\right) = \begin{cases} 1, \sigma_1 > \sigma_2 \\ 0, \sigma_1 = \sigma_2 \\ -1, \sigma_1 < \sigma_2 \end{cases} . \tag{13}$$

The function $U\left(\sigma_1, \sigma_2\right)$ can be selected in the following way

$$U\left(\sigma_1, \sigma_2\right) = \frac{\sigma_1 - \sigma_2}{\left|\sigma_1 - \sigma_2\right|} . \tag{14}$$

If there is a need to differentiate the expression from з $U\left(\sigma_1, \sigma_2\right)$, it can be approximated by a smooth function $\bar{U}\left(\sigma_1, \sigma_2\right)$ that has derivatives at any point

$$\bar{U}\left(\sigma_1, \sigma_2\right) = \frac{e^{-\beta\left(\frac{\sigma_2}{\sigma_1} - 1\right)} - 1}{e^{-\beta\left(\frac{\sigma_2}{\sigma_1} - 1\right)} + 1}, \tag{15}$$

where the larger the parameter β, the closer the function $\bar{U}\left(\sigma_1, \sigma_2\right)$ to a step function.

Fig.4 b shows the dependence of the effective conductivity on the concentration at a given percolation threshold $\tilde{p}_c = 0.1$ at $\sigma_1 = 1, \sigma_2 = 10^{-5}$ (conditional units) and the opposite case, when $\sigma_1 = 10^{-5}, \sigma_2 = 1$

Now, having a term in the form (12,13) or (12,15), it is possible to find various dependences of the effective coefficients for any inequality σ_1, σ_2 in a uniform way.

Effective thermoelectric properties of randomly heterogeneous media

In [11], the thermoelectric properties of composites with different percolation thresholds were considered. A mean-field approximation with a term similar to Svt was used [7]. Here, we use a modified term (12-15), which allows us to consider any cases of inequalities in the local kinetic coefficients of phases in a uniform way.

We choose the following values of the kinetic coefficients of the first and second phases [15], at a temperature T = 300 K:

$$\sigma_1 = 5\cdot 10^6 \text{Ом}^{-1}\text{м}^{-1},\ \kappa_1 = 36.1\ \text{Вт}/(\text{м}\cdot\text{К}),\ \alpha_1 = 0\ \text{В}/\text{К},$$

$$\sigma_2 = 10^5 \text{Ом}^{-1}\text{м}^{-1},\ \kappa_2 = 0.963\ \text{Вт}/(\text{м}\cdot\text{К}),\ \alpha_2 = 173\cdot 10^{-6}\,\text{В}/\text{К}, \tag{16}$$

where κ is thermal conductivity, α is thermoEMF. Thus, in the selected variant $\sigma_1 > \sigma_2$ and $\kappa_1 > \kappa_2$ the thermoelectric figure of merit of phases is equal to

$$Z_1T = 0, \qquad Z_2T = \frac{\sigma_2\alpha_2^2}{\kappa_2}T = 1.2 \tag{17}$$

We introduce parameter λ, which allows us to consider a set of values σ_2 and α_2. The figure of merit Z_2T remains unchanged, but inequality $\sigma_1 > \sigma_2$ is reversed

$$\sigma_2(\lambda) = \sigma_2(1+\lambda), \qquad \alpha_2(\lambda) = \alpha_2(1+\lambda)^{-1/2},\ \lambda\in[0,9] \tag{18}$$

At $\lambda = 0$ there is an initial set of values of local kinetic coefficients, and at $\lambda = 9$, $\sigma_2 = 10^6$ $Ohm^{-1}m^{-1}$ and $\alpha_2 = 63\cdot 10^{-6}$ *V/K*. Now the reverse inequality $\sigma_2 > \sigma_1$ holds for conductivity.

We write the expression for current densities and heat flux in the form

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

$$\frac{\langle \mathbf{q}\rangle}{T} = \sigma_e\alpha_e\langle \mathbf{E}\rangle + \kappa_e\frac{1+Z_eT}{T}\langle -\nabla T\rangle, \tag{19}$$

where ∇T is temperature gradient.

We introduce the matrix of local and effective kinetic coefficients $\mathbf{\Omega}$

$$\hat{\Omega}_i = \begin{pmatrix} \sigma_i & \sigma_i\alpha_i \\ \sigma_i\alpha_i & \kappa_i\dfrac{1+Z_iT}{T} \end{pmatrix}, \tag{20}$$

where i is phase number.

In such notation, the mean-field approximation for thermoelectric phenomena can be written as (details are given in [11])

$$\mathbf{\Lambda}_1 p + \mathbf{\Lambda}_2(1-p) = 0, \tag{21}$$

where

$$\mathbf{\Lambda}_1 = \frac{\hat{\Omega}_e - \hat{\Omega}_1}{2\hat{\Omega}_e + \hat{\Omega}_1}, \qquad \mathbf{\Lambda}_2 = \frac{\hat{\Omega}_e - \hat{\Omega}_2}{2\hat{\Omega}_e + \hat{\Omega}_2}, \tag{22}$$

where expressions of the type $\dfrac{\hat{\Omega}_e - \hat{\Omega}_1}{2\hat{\Omega}_e + \hat{\Omega}_1}$ are understood as $\left(\hat{\Omega}_e - \hat{\Omega}_1\right)\left(2\hat{\Omega}_e + \hat{\Omega}_1\right)^{-1}$.

The self-consistency equation with a modified term is given by

$$\frac{\Lambda_1}{1+\mathbf{C}\Lambda_1}p + \frac{\Lambda_2}{1+\mathbf{C}\Lambda_2}(1-p) = 0, \tag{23}$$

where now the term is written in the form of a matrix

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

Here, $c_\sigma(p,\tilde{p}_c)$ is taken from (12-15), and $c_\kappa(p,\tilde{p}_c)$ is found therefrom with a corresponding replacement of σ_1 by κ_1 and σ_2 by κ_2.

Let us analyse the resulting solution (23) for the figure of merit. Fig.5 shows the dependence of Z_eT on parameter λ. With a change in parameter λ, the phase figures of merit remain unchanged. Surprisingly enough, the effective figure of merit in this case is a function of parameter λ. Note also the nonlinear dependence of the effective figure of merit on parameter λ.

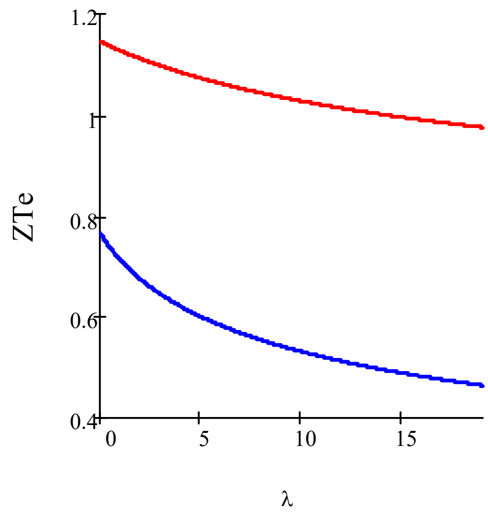


Fig.5 Dependence of Z_eT on parameter λ.
The upper curve at a concentration of 0.1, the other – at 0.3

Fig.6 shows the dependence of the effective figure of merit Z_eT on the concentration of the first phase p for different values of parameter λ. Note that these dependences change the ranking by the values of parameter λ (the larger the parameter, the larger the maximum) when passing through the percolation threshold.

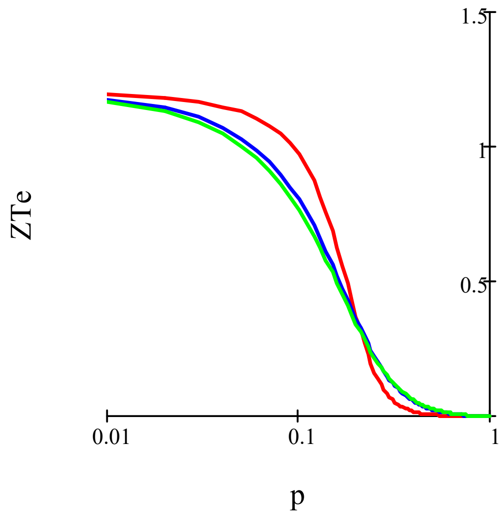


Fig.6 Dependence of the effective figure of merit Z_eT on the concentration of the first phase for different values of parameter λ (from top to bottom in the left part) the parameter is equal to 0, 9, 15

Conclusion

Using the modification of the term proposed in [11], the behaviour of the effective figure of merit Z_eT with a different set of local kinetic coefficients was considered. As it turned out, with constant values of local figures of merit $Z_1T = \text{const}$, $Z_2T = \text{const}$ the value Z_eT changes with a change in the set of kinetic coefficients. It is interesting to consider a similar modification of the term for the task of determining effective elastic moduli.

This approach makes it possible to describe composites with different percolation thresholds within the framework of the mean-field theory. Note that the values of the effective coefficients, even far from the percolation threshold, depend on the value of the percolation threshold. Composites with nanoparticles exhibit [16] unusual percolation threshold values from the point of view of standard mean-field theory and percolation theory (there is, among other things, a difference between the experimentally obtained percolation threshold values for, for example, electrical conductivity and elasticity). The approach proposed in this paper makes it possible (formally, without elucidating the physical reason for such a phenomenon) to describe the effective properties of such composites.

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ТЕРМОЕЛЕКТРИЧНІ КОМПОЗИТИ З РІЗНИМИ ПОРОГАМИ ПРОТІКАННЯ

Розглядається модифікація наближення середнього поля для опису поведінки ефективних кінетичних коефіцієнтів, у тому числі для термоелектричних композитів. Запропонована модифікація дозволяє описувати випадково-неоднорідні середовища з різними порогами протікання при довільних значеннях локальних кінетичних коефіцієнтів. Бібл. 16. рис. 6.
Ключові слова: термоелектричні композити, кінетичні коефіцієнти.

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