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Effective Thermoelectric Coefficients for an Exactly Solvable Two-Dimensional Three-Phase Composite

The effective thermoelectric coefficients for a two-dimensional three-phase macroscopically disordered medium are found. To obtain such a value, the method of sequential averaging is used, which allowed using for the method the isomorphism created for the two-phase case. This is compared with the mean-field approximation. It is shown that the conditions that determine the exact values of the effective kinetic coefficients of D-media are insufficient for three-phase media.

Keywords: effective thermoelectric coefficients, two-dimensional three-phase composite, macroscopically disordered medium, heterogeneous materials.

Introduction

There are many approximate methods for calculating effective kinetic coefficients (hereinafter ECCs) for various types of macroscopically disordered media, see monographs [1-4]. The most widely used method is the mean field theory method, the so-called self-consistent method or Bruggeman-Landauer approximation [5, 6]. As a rule, when using approximate methods, the question arises of their accuracy and applicability for different parameter values, for example, with weak or strong heterogeneity of local properties. One possible way to get an answer is to compare the results of the approximate method with the exact solution. Obviously, exact solutions are possible in exceptional (single) cases, otherwise why would we use approximations. Thus, for the problem of calculating the effective conductivity (in the absence

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of thermoelectric phenomena), exact solutions are known for flat-layer media and two-dimensional two-phase Dykhne media [7]. In the same cases, exact solutions are possible for media with thermoelectric phenomena.

Here, a method is proposed, which can be called the method of sequential averaging, which allows obtaining an exact solution for disordered media of a special structure with thermoelectric phenomena for a three-phase medium.

In the first section, a sequential averaging scheme for the effective conductivity problem will be considered and a comparison with the approximate Bruggeman-Landauer method will be given. In the second part, a problem with thermoelectric phenomena will be considered, and an approximate solution of the problem within the framework of the mean field theory approximation will be given.

1. Effective conductivity of a two-dimensional three-phase medium

Here we will use the exact result for a two-phase medium obtained by A.M. Dykhne [7]. In the work it was shown that for a two-phase two-dimensional $m = 2$ medium, with conductivities σ_1 and σ_2 , with their concentration $\frac{1}{2}$ and such their arrangement that the effective conductivity σ_e is a scalar and does not change with mutual change of phases $\sigma_1 \rightleftharpoons \sigma_2$, that is, $\sigma_e(\sigma_1, \sigma_2) = \sigma_e(\sigma_2, \sigma_1)$ there is an exact analytical expression

$$\sigma_e = \sqrt{\sigma_1 \sigma_2} . \tag{1}$$

Such media will be hereinafter referred to as Dykhne media or D-media.

We now consider the following algorithm for constructing a medium from three phases $m = 3$ [8]. Let us take three boxes with numbers 1, 2 and 3, each of which contains a material (phase), with conductivities σ_1, σ_2 and σ_3 , respectively – Fig. 1.

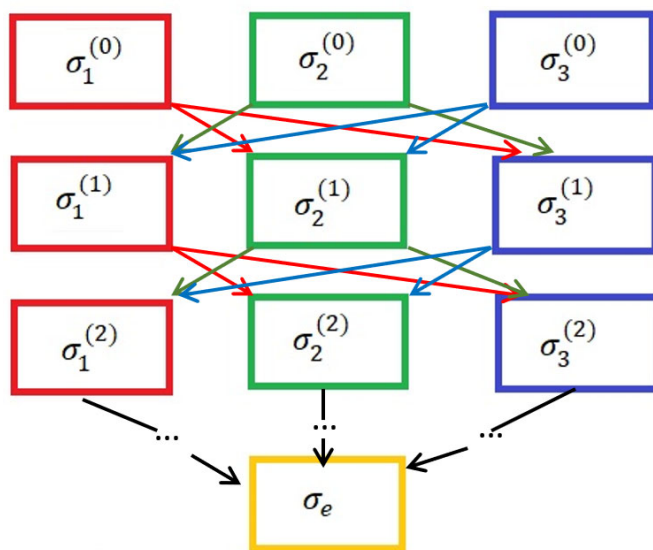


Fig. 1. Sequential averaging scheme

This arrangement of such phases in such boxes will be called a zero step.

$$\sigma_1^{(0)} = \sigma_1, \quad \sigma_2^{(0)} = \sigma_2, \quad \sigma_3^{(0)} = \sigma_3. \quad (2)$$

In the next, first step, we will take an equal share of materials (with conductivities $\sigma_2^{(0)}$ and $\sigma_3^{(0)}$) from boxes 2 and 3 and for a D- medium from them, with conductivity $\sigma_1^{(1)}$, which we will place in a new box 1

$$\sigma_1^{(1)} = \sqrt{\sigma_2^{(0)}\sigma_3^{(0)}} = \sqrt{\sigma_2\sigma_3}. \quad (3)$$

Similarly, we will create materials in new boxes 2 and 3

$$\sigma_2^{(1)} = \sqrt{\sigma_1^{(0)}\sigma_3^{(0)}} = \sqrt{\sigma_1\sigma_3}, \quad \sigma_3^{(1)} = \sqrt{\sigma_1^{(0)}\sigma_2^{(0)}} = \sqrt{\sigma_1\sigma_2}. \quad (4)$$

Continuing this procedure, for the second step $\sigma_1^{(2)}$ we obtain

$$\sigma_1^{(2)} = \sqrt{\sigma_2^{(1)}\sigma_3^{(1)}} = \sqrt{\sqrt{\sigma_1\sigma_3}\sqrt{\sigma_1\sigma_2}} = \sqrt{\sigma_1\sqrt{\sigma_2\sigma_3}} = \sigma_1^{1/2}\sigma_2^{1/4}\sigma_3^{1/4}, \quad (5)$$

And similar for other boxes.

Continuing this procedure of sequential mixing with averaging, for the degrees of specific conductivities of the first box, which were equal in the second step (see (5)) $1/2, 1/4, 1/4$ in the following steps, starting from zero, we obtain

$$\sigma_1 \rightarrow 0, \frac{1}{2}, \frac{1}{4}, \frac{3}{8}, \frac{5}{16}, \frac{11}{32}, \frac{21}{64}, \frac{43}{128}, \dots \quad (6)$$

Degrees $\sigma_2 \rightarrow \frac{1}{2}, \frac{1}{4}, \frac{3}{8}, \frac{5}{16}, \frac{11}{32}, \frac{21}{64}, \frac{43}{128}, \dots \quad (7)$

$$\sigma_3 \rightarrow \frac{1}{2}, \frac{1}{4}, \frac{3}{8}, \frac{5}{16}, \frac{11}{32}, \frac{21}{64}, \frac{43}{128}, \dots \quad (8)$$

The sequence of numerators (6) $a_n = 0, 1, 1, 3, 5, \dots \quad n = 0, 1, 2, 3, \dots$, obeying the recurrence relation $a_n = a_{n-1} + 2a_{n-2}$, $a_0 = 0, a_1 = 1$, is called the Jacobsthal number and, according to the sequence library [9] is not an analytical expression for the n-th number. However, at $n \gg 1$ the value a_n approaches the largest integer from $2^n / 3$. And since the denominator of the sequence (6) is 2^n , then

$$\lim_{n \rightarrow \infty} \sigma_1^{a_n/2^n} = \sigma_1^{1/3}. \quad (9)$$

Similar for (7) and (8).

Thus, the final result of sequential averaging, when an exact expression for the effective value is used at each stage, for all three boxes yields

$$\sigma_e = \sqrt[3]{\sigma_1\sigma_2\sigma_3}. \quad (10)$$

Expression (10) corresponds to qualitative considerations. Indeed, since at each averaging step (4), (5) and subsequent ones the effective conductivity in each of the boxes has the form $\sigma_1^\alpha \sigma_2^\beta \sigma_3^\gamma$, and the final value should not depend on the numbering of the phases,

$$\sigma_e(\sigma_1, \sigma_2, \sigma_3) = \sigma_e(\sigma_2, \sigma_1, \sigma_3) = \sigma_e(\sigma_3, \sigma_2, \sigma_1) = \dots, \quad (11)$$

then $\alpha = \beta = \gamma$, and their sum, due to dimensionality $\alpha + \beta + \gamma = 1$.

Let us now compare the result of the obtained value $\sigma_e(10)$, according to the use of the exact result (1) of the effective conductivity with the approximate expression, which can be obtained from the Bruggeman-Landauer approximation

$$\frac{\sigma_e - \sigma_1}{\sigma_e + \sigma_1} p_1 + \frac{\sigma_e - \sigma_2}{\sigma_e + \sigma_2} p_2 + \frac{\sigma_e - \sigma_3}{\sigma_e + \sigma_3} p_3 = 0, \quad p_1 + p_2 + p_3 = 1. \quad (12)$$

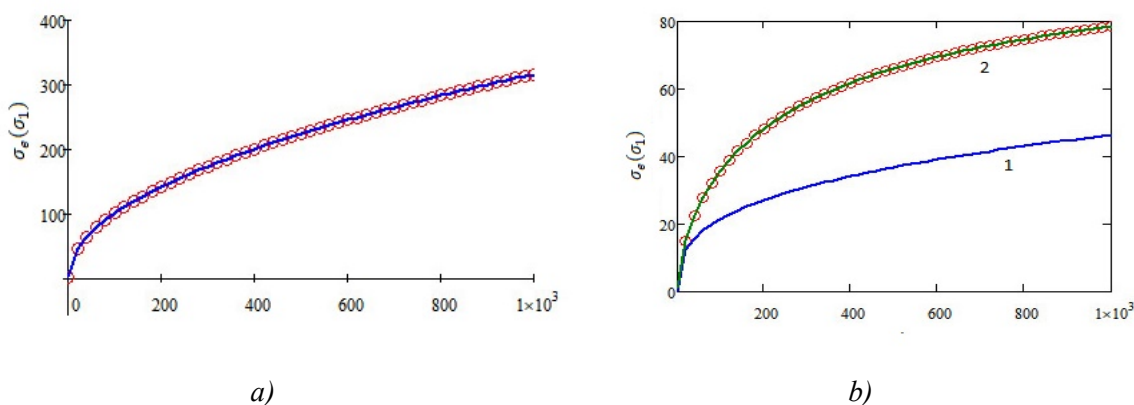


Fig. 2. Dependences of the effective conductivity σ_e on the conductivity values of the first phase. a – for the three-phase case, 1 – exact solution (10), 2 – approximate (13), b – for the two-phase case

At equal concentration of phases

$$\frac{\sigma_e - \sigma_1}{\sigma_e + \sigma_1} + \frac{\sigma_e - \sigma_2}{\sigma_e + \sigma_2} + \frac{\sigma_e - \sigma_3}{\sigma_e + \sigma_3} = 0. \quad (13)$$

Fig. 2 shows the dependences of the effective conductivity on the value of one of the phases for the exact solution (10) according to the solution of the Bruggeman-Landauer equation (13) for two- and three-phase cases.

As can be seen from Fig. 2 *b*, the Bruggeman-Landauer approximation coincides with the exact analytical solution of Dykhne (1). This is, of course, due to the fact that the conditions formulated for D-media – 1) two-dimensionality, 2) two-phase nature, 3) isotropy of the effective value, 4) equal concentration of phases and independence of the effective conductivity from mutual replacement of phases – are sufficient for an unambiguous determination of the effective conductivity. A large number of different structures meet these conditions [4]. For example, one formed by sequential averaging of two-dimensional polycrystals, which in turn are formed from flat-layer structures, also gives the exact value (10) [8].

As can be seen from Fig. 2 *a*, for the three-dimensional case similar conditions leading to one universal expression are insufficient. The Bruggeman-Landauer approximation, which implies that the effective conductivity is isotropic, does not depend on the mutual change of phases, and their concentration is the same, gives a result different from the result obtained by sequential averaging.

2. Thermoelectric properties of a two-dimensional three-phase medium

In the case of thermoelectric phenomena, Ohm's law, written in local and averaged form

$$\mathbf{j} = \sigma(r)\mathbf{E}, \quad \langle \mathbf{j} \rangle = \sigma^e \langle \mathbf{E} \rangle, \quad (14)$$

should be generalized and include heat flux – \mathbf{q} and temperature gradient – $\mathbf{g} = -gradT$

$$\begin{aligned} \mathbf{j} &= \sigma\mathbf{E} + \gamma\mathbf{g}, \\ \mathbf{s} &= \gamma\mathbf{E} + \chi\mathbf{g}, \end{aligned} \quad (15)$$

where $\gamma = \sigma\alpha$, $\chi = \kappa/T(1+ZT)$, $\mathbf{s} = \mathbf{q}/T$, $Z = \sigma \cdot \alpha^2 / \kappa$, α is the Seebeck coefficient, κ is thermal conductivity and for convenience (symmetry in system (15)) the flux $\mathbf{s} = \mathbf{q}/T$ is introduced.

Let us write this system in a convenient form for further use, in matrix notation:

$$\begin{aligned} \mathbf{j} &= A_{11}\mathbf{E} + A_{12}\mathbf{g} \\ \mathbf{s} &= A_{12}\mathbf{E} + A_{22}\mathbf{g} \end{aligned} \quad (16)$$

or

$$\begin{pmatrix} \mathbf{j} \\ \mathbf{s} \end{pmatrix} = \begin{pmatrix} A_{11} & A_{12} \\ A_{12} & A_{22} \end{pmatrix} \begin{pmatrix} \mathbf{E} \\ \mathbf{g} \end{pmatrix}, \quad \begin{pmatrix} \mathbf{j} \\ \mathbf{s} \end{pmatrix} = \hat{\mathbf{A}} \begin{pmatrix} \mathbf{E} \\ \mathbf{g} \end{pmatrix}, \quad (17)$$

where $\hat{\mathbf{A}}$ is the tensor of thermoelectric coefficients

Accordingly

$$\left\langle \begin{pmatrix} \mathbf{j} \\ \mathbf{s} \end{pmatrix} \right\rangle = \hat{\mathbf{A}}^e \left\langle \begin{pmatrix} \mathbf{E} \\ \mathbf{g} \end{pmatrix} \right\rangle. \quad (18)$$

$\hat{\mathbf{A}}^e$ is the tensor of the effective thermoelectric coefficients.

For D – media, as in the case of a purely conductive problem, there is an exact analytical solution [10, 8], which in matrix form can be written as

$$\hat{\mathbf{A}}^e = \left(Det\hat{\mathbf{A}}_1 Det\hat{\mathbf{A}}_2 \right)^{1/4} \frac{\frac{\hat{\mathbf{A}}_1}{\sqrt{Det\hat{\mathbf{A}}_1}} + \frac{\hat{\mathbf{A}}_2}{\sqrt{Det\hat{\mathbf{A}}_2}}}{\sqrt{Det\left(\frac{\hat{\mathbf{A}}_1}{\sqrt{Det\hat{\mathbf{A}}_1}} + \frac{\hat{\mathbf{A}}_2}{\sqrt{Det\hat{\mathbf{A}}_2}} \right)}}, \quad i = 1, 2 \quad (19)$$

Index 1 and 2 specify the first and second phases.

The expression for the effective thermoelectric coefficients (19) can be obtained from the same symmetry considerations that were formulated and used in [7] to find the exact value of the effective conductivity in the absence of thermoelectric phenomena. Another way to solve this problem is also possible. In the works [11, 12, 10], see also [4, 13, 14, 15] it is shown how, having a known analytical expression (dependence on the value of phases and concentration) of the effective conductivity of a two-phase medium, to obtain an expression for the effective thermoelectric coefficients. Such a method, called the isomorphism method, allows, for example, to obtain (19) based on $\sqrt{\sigma_1\sigma_2}$. Unfortunately, this method works only for the two-phase case. For the three-phase case, this method does not work and it remains only to use approximate methods, for example, mean field theory.

Here we show how, for the considered case of obtaining the effective conductivity of a three-phase medium using the exact solution of the two-phase case ($\sqrt{\sigma_1\sigma_2} \rightarrow \sqrt[3]{\sigma_1\sigma_2\sigma_3}$), the thermoelectric problem can be generalized.

Let us introduce the notation

$$\hat{Q}(\hat{a}, \hat{b}) = (\text{Det}\hat{a} \cdot \text{Det}\hat{b})^{1/4} \frac{\frac{\hat{a}}{\sqrt{\text{Det}\hat{A}_1}} + \frac{\hat{A}_2}{\sqrt{\text{Det}\hat{A}_2}}}{\sqrt{\text{Det}\left(\frac{\hat{A}_1}{\sqrt{\text{Det}\hat{A}_1}} + \frac{\hat{A}_2}{\sqrt{\text{Det}\hat{A}_2}}\right)}}. \quad (20)$$

Let us now apply the process of sequential averaging and draw a mixing scheme similar to that used above to obtain the effective conductivity. Consider three boxes (see Fig. 1) containing material, with tensors of local kinetic coefficients \hat{P} , \hat{T} and \hat{S} . At the zero step

$$\hat{P}^{(0)} = \hat{A}_1, \quad \hat{T}^{(0)} = \hat{A}_2, \quad \hat{S}^{(0)} = \hat{A}_3, \quad (21)$$

that is, at the zero step in the boxes there is material with local coefficients corresponding to the first, second and third phases, respectively.

At the first step, the boxes contain material with effective properties obtained according to (20)

$$\hat{P}^{(1)} = \hat{Q}(\hat{T}^{(0)}, \hat{S}^{(0)}), \quad \hat{T}^{(1)} = \hat{Q}(\hat{P}^{(0)}, \hat{S}^{(0)}), \quad \hat{S}^{(1)} = \hat{Q}(\hat{P}^{(0)}, \hat{T}^{(0)}), \quad (22)$$

and at the $n+1$ step

$$\hat{P}^{(n+1)} = \hat{Q}(\hat{T}^{(n)}, \hat{S}^{(n)}), \quad \hat{T}^{(n+1)} = \hat{Q}(\hat{P}^{(n)}, \hat{S}^{(n)}), \quad \hat{S}^{(n+1)} = \hat{Q}(\hat{P}^{(n)}, \hat{T}^{(n)}). \quad (23)$$

At $n \rightarrow \infty$ matrices in the boxes coincide with each other, and their values are the values of the effective thermoelectric coefficients of the three-phase medium.

$$\hat{A}^e(\hat{A}_1, \hat{A}_2, \hat{A}_3) = \lim_{n \rightarrow \infty} \hat{P}^{(n)} = \lim_{n \rightarrow \infty} \hat{T}^{(n)} = \lim_{n \rightarrow \infty} \hat{S}^{(n)}. \quad (24)$$

Fig. 30 *a* shows the dependence of the effective coefficients A_{11}^e, A_{12}^e и A_{22}^e on the value of thermoEMF α . The value of the first phase was chosen to be close to the thermoelectric material Bi₂Te₃ $\sigma_1 = 10^5 \Omega^{-1} \text{m}^{-1}$, $\alpha_1 = 2 \cdot 10^{-4} \text{V/K}$, $\kappa_1 = 1 \text{W/m}\cdot\text{K}$, at $T = 300 \text{K}$ the figure of merit of the second phase $Z_1 T = 1.2$ [16]. Let us note that for the sake of example we have considered the case of changing the Seebeck coefficient of only the first phase, all other thermoelectric coefficients of all phases – conductivity, thermoEMF of the second and third phases and thermal conductivity do not change. According to (15)-(16) it means that with a change in α_1 the matrix coefficients $A_{12} = \sigma_1 \alpha_1$ and $A_{22} = \frac{\kappa_1}{T} \left(1 + \frac{\sigma_1 \alpha_1^2}{\kappa_1} T \right)$ change. For the second and third phases $B_{11} = 5 \cdot 10^6$, $B_{12} = 10^{-3}$, $B_{22} = 0.13$, $C_{11} = 1$, $C_{12} = 0.01$, $C_{22} = 0.01$ were selected.

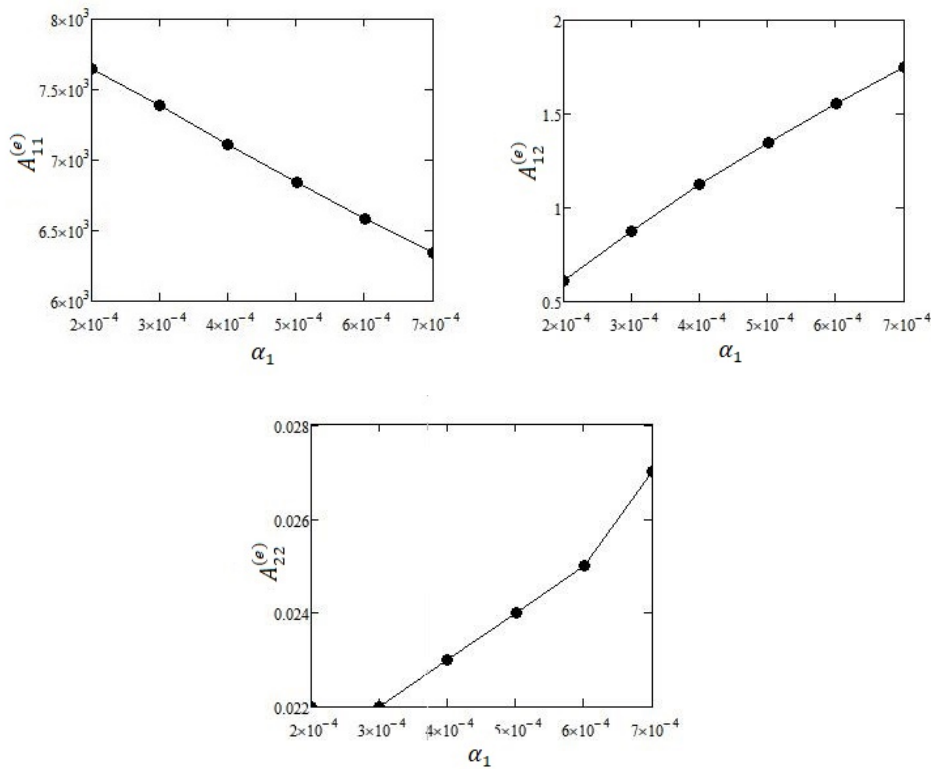


Fig. 3. Dependence of the effective kinetic coefficients on the thermoEMF α_1 of the first phase obtained by sequential averaging method

Let us now compare the effective thermoelectric coefficients obtained by the method of sequential averaging with the result of calculation by the approximate method – a generalization of the Bruggeman-Landauer approximation to the thermoelectric case.

$$\frac{\hat{A}^e - \hat{A}_1}{\hat{A}^e - \hat{A}_1} p_1 + \frac{\hat{A}^e - \hat{A}_2}{\hat{A}^e - \hat{A}_2} p_2 + \frac{\hat{A}^e - \hat{A}_3}{\hat{A}^e - \hat{A}_3} p_3 = 0, \quad (25)$$

where like in (12,13) $p_1 = p_2 = p_3 = 1/3$.

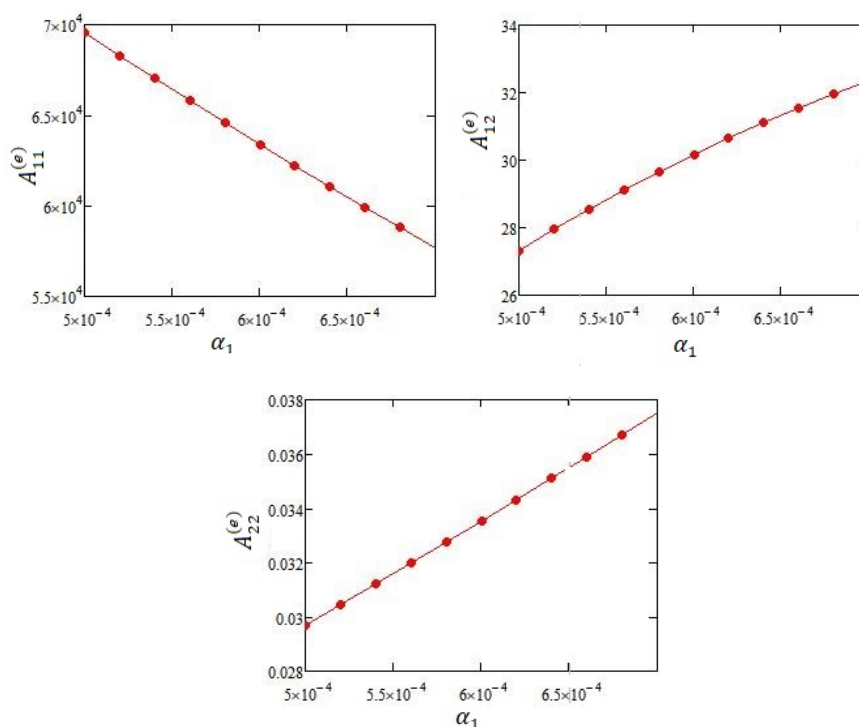


Fig. 4. Dependence of the effective kinetic coefficients on the thermoEMF α_1 of the first phase obtained from the Bruggeman-Landauer approximation

As can be seen from Fig. 4, the dependences obtained using the approximate method qualitatively exhibit the same patterns – increase or decrease – but different numerical values. This is similar to what occurs when comparing calculations of the single-flow effective conductivity problem.

Discussion

The paper formulates and applies a method of sequential averaging. This method allows for the calculation of effective kinetic coefficients, including those for thermoelectric phenomena.

First, as is known from the "construction" itself, the resulting medium has a non-standard structure. Yes, for the conductivity problem, it yields an effective conductivity value with a non-standard dependence on local conductivity values. For example, given equal phase concentrations, with mutual phase exchange, if the local conductivity of one phase approaches zero, the effective conductivity value also approaches zero. The medium ceases to conduct.

Second. The sequential averaging method for thermoelectricity, where a two-phase medium is considered at each averaging stage, allows for the use of an exact isomorphism method, which is fundamentally impossible for a three-phase medium. Thus, the "bypass" of sequential averaging (mixing) allows for the use of the "two-phase" method for the three-phase case.

Third. It is shown that when using the conditions formulated for D-media – 1) two-dimensionality, 2) two-phase, 3) isotropy of the effective value, 4) equal concentration of phases and independence of the effective conductivity from the mutual replacement of phases

– sufficient for determining (specifying) the effective conductivity, it is insufficient for a three-phase medium. Two examples of three-phase media with different effective conductivities are given, in which these conditions are fulfilled. The same statement applies to the determination of the effective kinetic coefficients in the presence of thermoelectric phenomena. It should be noted that in the study of the effective elastic coefficients [17] such conditions are not sufficient for the two-phase case.

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Ефективні термоелектричні коефіцієнти для точно розв'язуваного двовимірного трифазного композиту

Знайдені ефективні термоелектричні коефіцієнти для двовимірно трифазного макроскопічно неупорядкованого середовища. Для отримання такого значення використаний метод послідовного усереднення, який дозволив використати для методу ізоморфізм, що був створений для двофазного випадку. Дане порівняння з наближенням середнього поля. Показано, що умови, що визначають точні значення ефективних кінетичних коефіцієнтів D – середовищ, для трифазних середовищ є недостатньо.

Ключові слова: ефективні термоелектричні коефіцієнти, двовимірний трифазний композит, макроскопічно неупорядковане середовище, гетерогенні матеріали.