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THERMOELECTRIC COMPOSITES AND RECIPROCITY RELATIONS

The effective kinetic coefficients in macro-inhomogeneous media, their behavior when changing the concentration of component phases and setting the percolation threshold are considered. Combinations of the effective kinetic coefficients were found for which the reciprocity relations are fulfilled. Bibl. 7, Fig. 4, Tabl. 6.
Key words: kinetic coefficients, two-phase medium, single-flow systems, thermoEMF.

Introduction

The main chacreristic of randomly inhomogeneous media are effective kinetic coefficients. When describing, for instance, electrical conductivity - σ_e, when Ohm's law applies

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

Where $\mathbf{j}$ – electric current density and – $\mathbf{E}$ – electric field strength, and local conductivity $\sigma(\mathbf{r})$, where $\langle ... \rangle = 1/V \int ... dV$ is volume average and in case of a two-phase medium the conductivity in the first phase acquires the value σ_1, and in the second phase σ_2. The effective conductivity σ_e is defined as

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

A huge number of articles and monographs [1 – 5] are devoted to the calculation of the effective properties of such media, in particular, the calculation of the effective conductivity, thermoEMF and elastic properties. One of the successful approximate methods that describe the effective conductivity well is the Bruggeman-Landauer approximation [6 – 7], which is often called the mean field approximation - MAE. For effective conductivity, it has the form

$$\frac{\sigma_e - \sigma_1}{2\sigma_e + \sigma_1} p + \frac{\sigma_e - \sigma_2}{2\sigma_e + \sigma_2}(1 - p) = 0 . \tag{3}$$

Where σ_1 and σ_2 are the values of conductivity in the first and second phases.

Now let us turn to thermoelectric phenomena. We shall write down the equations interrelating the electric current density - $\mathbf{j}$, the heat flux - $\mathbf{q}$ and the electric field strength $\mathbf{E}$, as well as the temperature gradient $\mathbf{g} = -gradT$ as follows

$$\mathbf{j} = \sigma\mathbf{E} + \gamma\mathbf{g},$$

$$\mathbf{s} = \gamma\mathbf{E} + \chi\mathbf{g}, \tag{4}$$

where $\gamma = \sigma\alpha$, α is thermoemf, $\chi = \kappa/T$, κ is thermal conductivity and for convenience (symmetry in system (4)) the flux $\mathbf{s} = \mathbf{q}/T$ is introduced.

The effective kinetic coefficients of the thermoelectric system will have the form

$$\langle\mathbf{j}\rangle = \sigma_e\langle\mathbf{E}\rangle + \gamma_e\langle\mathbf{g}\rangle,$$

$$\langle\mathbf{s}\rangle = \gamma_e\langle\mathbf{E}\rangle + \chi_e\langle\mathbf{g}\rangle, \tag{5}$$

As it was shown in [8], the task of calculating effective thermoelectric coefficients can be reduced (in certain cases) to the task of determining the effective electrical conductivity in a system where there are no thermoelectric phenomena. In other words, if we know the solution for the effective coefficient in a single-flow system, we can find out the solution for the effective kinetic coefficients in a two-flow system (for example, with the joint flow of interconnected electric current and heat flux). Later on, this method, called the method of isomorphism, was written in various mathematical versions and generalized to various problems [9 – 11].

**Reciprocity relations for single-flow (conductivity) system**

There is a special class of two-dimensional two-phase media such that the effective conductivity (1) is isotropic and that when the local conductivities of the phases are interchanged, the effective conductivity remains unchanged. One of the many examples of the deterministic structure of such media is a checkerboard, where black cells are one phase, white cells are another, other examples are given in [12]. Randomly inhomogeneous media with half concentration of phases also belongs to this class of media. As was precisely shown in [13, 14], the effective conductivity of such a medium is equal to

$$\sigma_e = \sqrt{\sigma_1\sigma_2} \ . \tag{6}$$

In the case when in randomly inhomogeneous media the concentration of phases is $p = 1/2$, the reciprocity relation holds [13, 14]

$$\sigma_e(p)\cdot\sigma_e(1-p) = \sigma_1\sigma_2, \tag{7}$$

that is, the product of two functions that depend on the concentration $\sigma_e(p)\cdot\sigma_e(1-p)$ is concentration-independent
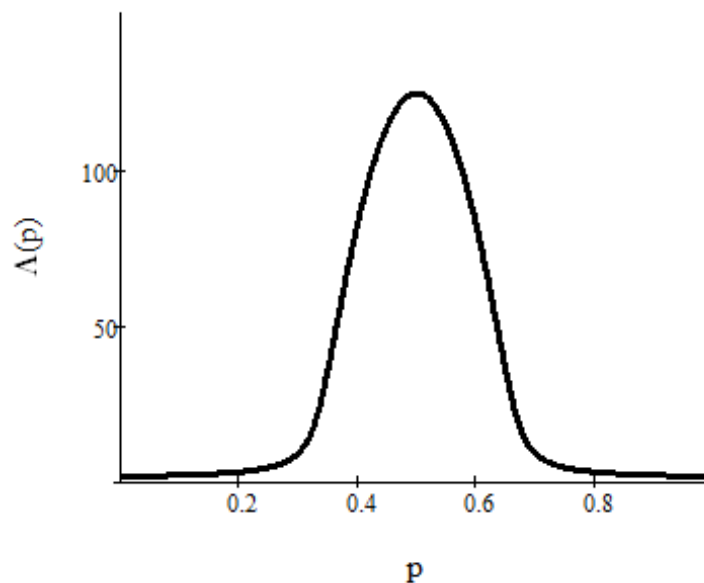
Fig. 1. Concentration dependence of the product of effective conductivities
in a three-dimensional random-inhomogeneous medium.
For example, $\sigma_1 = 1$, $\sigma_2 = 10^{-2}$ *is selected*
(in conventional units)

The analysis of this behavior of randomly inhomogeneous two-phase media by the approximate method of the mean field theory - MAE gives the same result.

Naturally, in randomly inhomogeneous three-dimensional media, as shown in Fig. 1, there is no such behavior.

For the concentration dependence of the effective conductivity, there is a specific parameter - p_c. With great heterogeneity, that is, with a large value of the phase conductance ratio $\sigma_1 / \sigma_2 >> 1$, the effective conductivity experiences a sharp change in behavior during the passage of the concentration through the so-called percolation threshold A sharp change in the behavior of effective conductivity is associated with the appearance in the medium of the so-called infinite cluster, a continuous path along one of the phases through the entire system [1, 5]. In the framework of the mean field approximation - MAE it has the value of the percolation threshold in the three-dimensional case $p_c = 1/3$, and in the two-dimensional case $p_c = 1/2$. At the same time, different values of percolation threshold are observed in different real composites. In this regard, in [15] a modification of MAE was proposed, which allows describing three-dimensional composites with a predetermined percolation threshold $\tilde{p}_c$, not necessarily equal to 1/3.

$$\frac{\dfrac{\sigma_e - \sigma_1}{2\sigma_e + \sigma_1}}{1 + c(p, \tilde{p}_c)\dfrac{\sigma_e - \sigma_1}{2\sigma_e + \sigma_1}}p + \frac{\dfrac{\sigma_e - \sigma_2}{2\sigma_e + \sigma_2}}{1 + c(p, \tilde{p}_c)\dfrac{\sigma_e - \sigma_2}{2\sigma_e + \sigma_2}}(1 - p) = 0, \tag{8}$$

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

$$c(p, \tilde{p}_c) = (1 - 3\tilde{p}_c)\left(\frac{p}{\tilde{p}_c}\right)^{\tilde{p}_c}\left(\frac{1-p}{1-\tilde{p}_c}\right)^{1-\tilde{p}_c}. \tag{9}$$

Later, this approach, which uses the introduction of some term in the Bruggeman-Landauer equation, was generalized to the two-dimensional case, to anisotropic structures, and to describe elastic phenomena [16 – 19].

In the future, we will explore the possibility of the existence of reciprocity relations between media with different percolation thresholds.

Reciprocity relations for the effective kinetic coefficients of thermoelectric composites

To analyze the behavior of the effective kinetic coefficients in thermoelectric media, we use the isomorphism method [20]. Let us first consider the two-dimensional case. In the absence of thermoelectric phenomena (i.e., at $\alpha_1 = 0$, $\alpha_2 = 0$) the system degenerates into two mutually incoherent relations - Ohm's law and Fourier's law, while for each of their effective coefficients (effective conductivity and thermal conductivity) the reciprocity relations are satisfied. In the presence of thermoelectric phenomena, the reciprocity relations are not fulfilled. As can be seen from Fig. 2, the normalized products of effective conductivities and thermoelectric coefficients are no longer constant with a change in concentration

$$\Lambda\sigma(p) = \frac{\sigma_e(p)\sigma_e(1-p)}{\left[\sigma_e(p_c = 1/2)\right]^2}, \Lambda\alpha(p) = \frac{\alpha_e(p)\alpha_e(1-p)}{\left[\alpha_e(p_c = 1/2)\right]^2} \tag{10}$$
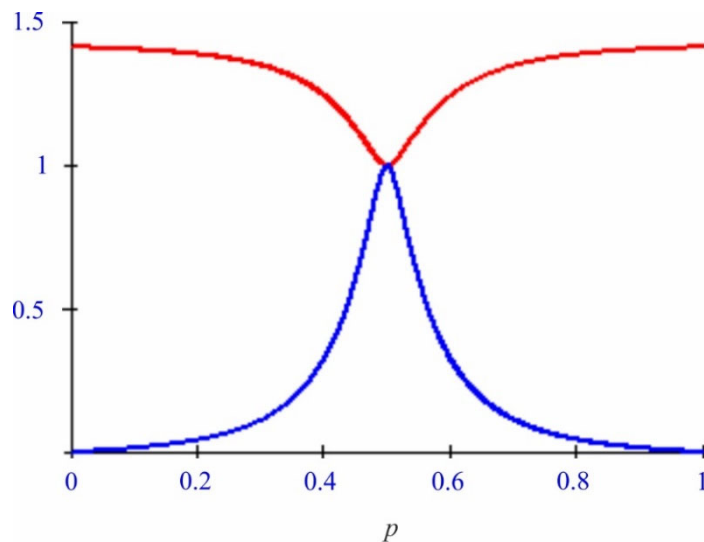

Fig. 2. Two-dimensional case. Concentration dependence of normalized products of conductivity and thermoelectric coefficients - $\Lambda\sigma(p)$ - upper curve and $\Lambda\alpha(p)$ - lower curve. For example, the following values of local kinetic coefficients were chosen: $\sigma_1 = 5 \cdot 10^7 \, \text{Ohm}^{-1} m^{-1}$, $\chi_1 = 0.1 \, W/m$, $\alpha_1 = 0 \, V/K$,
$\sigma_2 = 3.207 \cdot 10^4 \, Ohm^{-1}m^{-1}$, $\chi_2 = 3.3 \cdot 10^{-3} \, W/m$, $\gamma_2 = 6.414 \, V/K$, $T = 300 \, K$

Thus, in thermoelectric systems, the effective kinetic coefficients individually do not satisfy the reciprocity relations. However, as the isomorphism method states, the two-flow problem is reduced to a

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single-flow problem, with one effective kinetic coefficient for which, naturally, the reciprocity relation must hold. This means that it is possible, using isomorphism "in the opposite direction" (moving from a single-flow system to a two-flow system), to find those combinations of effective kinetic coefficients for which a reciprocity relation will take place in a two-flow system.

Isomorphism method

Here we will turn to the version proposed by A. Dykhne [20] (see [5] for details).

We will reduce the local two-flow system (4) to a single-flow system, for this we will add the first equation (4) to the second multiplied by some constant K

$$\mathbf{j} + K\mathbf{s} = (\sigma + K\gamma)\mathbf{E} + (\gamma + K\chi)\mathbf{g}, \tag{11}$$

where σ, α, γ and χ are coordinate-dependent and acquire the values $\sigma_1, \gamma_1, \chi_1 -$ in the first and $\sigma_2, \gamma_2, \chi_2 -$ in the second phases.

Rewriting (11) in the form

$$\mathbf{j} + K\mathbf{s} = (\sigma + K\gamma)\left(\mathbf{E} + \frac{\gamma + K\chi}{\sigma + K\gamma}\mathbf{g} \right) \tag{12}$$

we can introduce a new "current" $\mathbf{i}$ and a new field $\boldsymbol{\varepsilon}$

$$\mathbf{i} = \mathbf{j} + K\mathbf{s}, \quad \boldsymbol{\varepsilon} = \mathbf{E} + \frac{\gamma + K\chi}{\sigma + K\gamma}\mathbf{g} . \tag{13}$$

In the stationary case under consideration, for fields and currents the following equations hold

$$div\mathbf{j} = 0, \quad div\mathbf{s} = 0, \quad rot\mathbf{E} = 0, \quad rot\mathbf{g} = 0 \tag{14}$$

The new «field»- $\boldsymbol{\varepsilon}$ and «current»- $\mathbf{i}$ must obey similar equations

$$div\mathbf{i} = 0, \quad rot\boldsymbol{\varepsilon} = 0 , \tag{15}$$

which define a single-flow system

$$\mathbf{i} = f \cdot \boldsymbol{\varepsilon} , \tag{16}$$

where $f(\mathbf{r})$ is the kinetic coefficient of a new single-flow system (an analog to conductivity).

To fulfill equations (15), it is necessary that the factor near $\mathbf{g}$ does not depend on the coordinates, that is, that it has the same value in both phases - we denote it by ω. This requirement can be written as follows

$$\frac{\gamma_1 + K\chi_1}{\sigma_1 + K\gamma_1} = \frac{\gamma_2 + K\chi_2}{\sigma_2 + K\gamma_2} = \omega . \tag{17}$$

Equation (17) defines two possible values for the constants K and ω

$$K_{1,2} = \frac{\chi_2\sigma_1 - \chi_1\sigma_2 \pm \sqrt{(\chi_2\sigma_1 - \chi_1\sigma_2)^2 - 4(\chi_1\gamma_2 - \chi_2\gamma_1)(\gamma_1\sigma_2 - \gamma_2\sigma_1)}}{2(\chi_1\gamma_2 - \chi_2\gamma_1)} \tag{18}$$

Writing similar (12, 13, 16) equations for averaged fields and currents, we obtain, in particular, the averaged Ohm's law (for a single-flow medium)

$$\langle \mathbf{i} \rangle = f^e \langle \boldsymbol{\varepsilon} \rangle, \tag{19}$$

where now the role of local conductivity in the first and second phases σ_1, σ_2 will be played by conductivities μ_1, μ_2

$$\mu_1 = \sigma_1 + K\gamma_1, \qquad \mu_2 = \sigma_2 + K\gamma_2 . \tag{20}$$

It should be noted that there are two pairs of local effective kinetic coefficients μ_1 and μ_2, that depend on K_1 and one pair μ_1 and μ_2, that depends on K_2.

Thus, given that two values of the constant are possible (18), law (16) can be written as follows

$$\begin{aligned}
\langle \mathbf{j} \rangle + K_1 \langle \mathbf{s} \rangle &= f_1^e \left(\langle \mathbf{E} \rangle + \omega_1 \langle \mathbf{g} \rangle \right), \\
\langle \mathbf{j} \rangle + K_2 \langle \mathbf{s} \rangle &= f_2^e \left(\langle \mathbf{E} \rangle + \omega_2 \langle \mathbf{g} \rangle \right),
\end{aligned} \tag{21}$$

where it is taken into account that constant ω similar to constant K can take on two values and

$$f_1^e = f^e(\mu_1, \mu_2, K_1, \omega_1), \qquad f_2^e = f^e(\mu_1, \mu_2, K_2, \omega_2). \tag{22}$$

Now we find from (22) the expressions for $\langle \mathbf{j} \rangle$ and $\langle \mathbf{s} \rangle$

$$\begin{aligned}
\langle \mathbf{j} \rangle &= \frac{K_2 f_1^e - K_1 f_2^e}{K_2 - K_2} \langle \mathbf{E} \rangle + \frac{K_2 f_1^e \omega_1 - K_1 f_2^e \omega_2}{K_2 - K_2} \langle \mathbf{g} \rangle, \\
\langle \mathbf{s} \rangle &= \frac{f_1^e - f_2^e}{K_2 - K_2} \langle \mathbf{E} \rangle + \frac{f_1^e \omega_1 - f_2^e \omega_2}{K_2 - K_2} \langle \mathbf{g} \rangle
\end{aligned} \tag{23}$$

Thus, knowing the dependence of the effective kinetic coefficients of a single-flow system on the local kinetic coefficients and concentration, it is possible to obtain similar dependences for the effective kinetic coefficients of a thermoelectric (two-flow) system from (23). Indeed, comparing (23) and (5), σ_e, α_e and χ_e can be written as

$$\sigma_e = \frac{K_2 f_1^e - K_1 f_2^e}{K_2 - K_2}, \qquad \alpha_e = \frac{K_2 f_1^e \omega_1 - K_1 f_2^e \omega_2}{K_2 f_1^e - K_1 f_2^e}, \qquad \chi_e = \frac{f_1^e \omega_1 - f_2^e \omega_2}{K_2 - K_2} . \tag{24}$$

Reciprocity relations of thermoelectric composites

The isomorphism method described above explains why the effective kinetic coefficients do not directly give the reciprocity relation. According to [14], the reciprocity relations hold for single-flow two-dimensional randomly heterogeneous media. In this case, for the single-flow system (16), they must be present for the effective coefficients f_1^e and f_2^e and we write them in the form

$$
f^e\left(\mu_1,\mu_2,K_1,p\right)\cdot f^e\left(\mu_1,\mu_2,K_1,1-p\right)=f^e\left(\mu_1,\mu_2,K_1,1/2\right)^2,
$$
$$
f^e\left(\mu_1,\mu_2,K_2,p\right)\cdot f^e\left(\mu_1,\mu_2,K_2,1-p\right)=f^e\left(\mu_1,\mu_2,K_2,1/2\right)^2 \tag{25}
$$

or in abbreviated notations

$$
f_1^e\left(p\right)\cdot f_1^e\left(1-p\right)=f_1^e\left(1/2\right)^2, \qquad f_2^e\left(p\right)\cdot f_2^e\left(1-p\right)=f_2^e\left(1/2\right)^2. \tag{26}
$$

Thus, the effective kinetic coefficients of thermoelectric systems are a function of local kinetic coefficients, found constants (18), effective coefficients of single-flow systems $f_1^e\left(p\right)$, $f_2^e\left(p\right)$.

Let us now use (24) and find the expressions for $f_1^e\left(p\right)$, $f_2^e\left(p\right)$ through the effective kinetic coefficients of thermoelectric systems, for instance, in the form

$$
f_1^e\left(p\right)=\sigma_e+K_1\sigma_e\alpha_e, \quad f_2^e\left(p\right)=\sigma_e+K_2\sigma_e\alpha_e. \tag{27}
$$

According to (26) and (27) we can write the reciprocity relations for the effective kinetic coefficients of thermoelectric system for K_1

$$
\sigma_e\left(p\right)\left[1+K_1\alpha_e\left(p\right)\right]\cdot\sigma_e\left(1-p\right)\left[1+K_1\alpha_e\left(1-p\right)\right]=const \tag{28}
$$

And similarly for the second case of constant K_2

$$
\sigma_e\left(p\right)\left[1+K_2\alpha_e\left(p\right)\right]\cdot\sigma_e\left(1-p\right)\left[1+K_2\alpha_e\left(1-p\right)\right]=const \tag{29}
$$

Numerical analysis for two-dimensional thermoelectric systems

Let us consider several specific examples of the behavior of the effective kinetic coefficients and reciprocity relations. To do this, we normalize expressions (27 - 28) (recall that for the two-dimensional case in the standard MAE approximation the percolation threshold is equal to $p_c=1/2$)

$$
\Lambda_1\left(p\right)=\frac{\sigma_e\left(p\right)\left[1+K_1\alpha_e\left(p\right)\right]\cdot\sigma_e\left(1-p\right)\left[1+K_1\alpha_e\left(1-p\right)\right]}{\left\{\sigma_e\left(p\right)\left[1+K_1\alpha_e\left(p\right)\right]\right\}^2},
$$

$$
\Lambda_2\left(p\right)=\frac{\sigma_e\left(p\right)\left[1+K_2\alpha_e\left(p\right)\right]\cdot\sigma_e\left(1-p\right)\left[1+K_2\alpha_e\left(1-p\right)\right]}{\left\{\sigma_e\left(p\right)\left[1+K_2\alpha_e\left(p\right)\right]\right\}^2}. \tag{30}
$$

The expressions f^e will be found in the two-dimensional MAE approximation for a single-flow system

$$\frac{\dfrac{f_e - \mu_1}{f_e + \mu_1}}{1 + c(p, \tilde{p}_c)\dfrac{f_e - \mu_1}{f_e + \mu_1}} p + \frac{\dfrac{f_e - \mu_2}{f_e + \mu_2}}{1 + c(p, \tilde{p}_c)\dfrac{f_e - \mu_2}{f_e + \mu_2}}(1 - p) = 0, \tag{31}$$

where in the two-dimensional case the term similar to the Sarychev-Vinogradov term has the form

$$c(p, \tilde{p}_c) = (1 - 2\tilde{p}_c)U(\mu_1, \mu_2)\left(\frac{p}{\tilde{p}_c}\right)^{\tilde{p}_c}\left(\frac{1 - p}{1 - \tilde{p}_c}\right)^{1 - \tilde{p}_c}, \tag{32}$$

and

$$U(\mu_1, \mu_2) = \begin{cases} 1, & \mu_1 > \mu_2 \\ 0, & \mu_1 = \mu_2 \\ -1, & \mu_1 < \mu_2 \end{cases} \tag{33}$$

Substituting the obtained f_1^e and f_2^e in (20), we find $\Lambda_1(p)$ and $\Lambda_2(p)$. Fig.3 shows the concentration behaviour of $\Lambda\sigma(p)$, $\Lambda\alpha(p)$ and $\Lambda_1(p)$ from (10) and (30). Naturally, in (10) there is the effective conductivity from (24). As can be seen from the figure, the functions $\Lambda\sigma(p)$, $\Lambda\alpha(p)$ depend on the concentration, in contrast to $\Lambda_1(p)$, which is practically independent of the concentration, which can be called one of the reciprocity relations for thermoelectric phenomena.
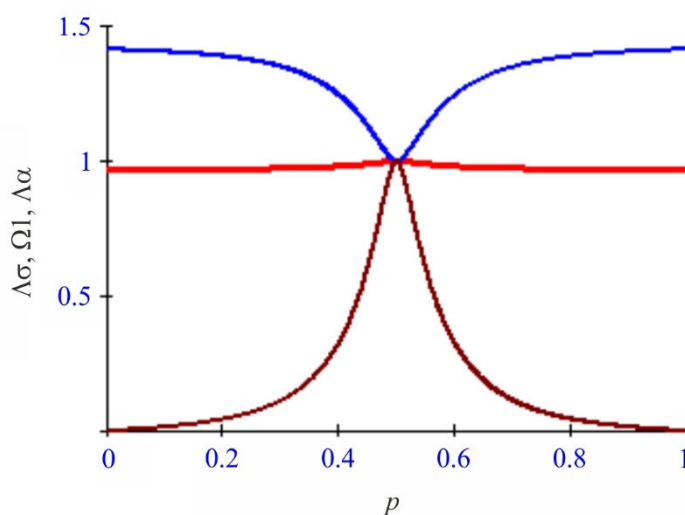

Fig.3 Concentration dependences $\Lambda\sigma(p)$, $\Lambda\alpha(p)$ and $\Lambda_1(p)$ with an unshifted percolation threshold
$\tilde{p}_c = 1/2$. For example, the following values of local kinetic coefficients were chosen:
$\sigma_1 = 5 \cdot 10^7 \, Ohm^{-1}m^{-1}$, $\chi_1 = 0.1 \, W/m$, $\alpha_1 = 0 \, V/K$, $\sigma_2 = 3.207 \cdot 10^4 \, Ohm^{-1}m^{-1}$,
$\chi_2 = 3.3 \cdot 10^{-3} \, W/m$, $\gamma_2 = 6.414 \, V/K$, $T = 300 \, K$

Note that when the percolation threshold shifts, that is, even with a small deviation of $\tilde{p}_c$ from 1/2, the expression $\Lambda_1(p)$ (and, of course, $\Lambda_2(p)$), which was constant, begins to significantly depend on the concentration.
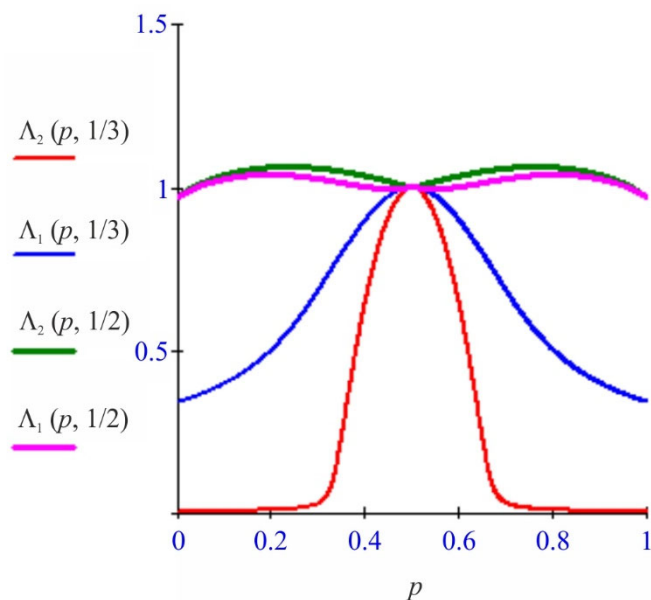

Fig. 4 Concentration dependences $\Lambda_2(p, p_c = 1/2), \Lambda_1(p, p_c = 1/2)$,
$\Lambda_1(p, p_c = 1/3), \Lambda_2(p, p_c = 1/3)$ *- top down*

In the three-dimensional case, when (8) and (9) hold, the products (30) for the unshifted percolation threshold, i.e. when $\tilde{p}_c = 1/3$, cease to be independent of the concentration, which is natural in general, since a strong dependence on the concentration is also observed in the single-flow case - Fig. 4. However, when the percolation threshold is shifted, when choosing $\tilde{p}_c = 1/2$ the reciprocity relations are approximately fulfilled. The three-dimensional case deserves a separate detailed analysis.

Conclusions

The reciprocity relations written for the effective conductivity (single-flow system) can be generalized to the case of thermoelectric phenomena in the two-dimensional case. In the three-dimensional form, the canonical mean field theory (Bruggeman-Landauer approximation) shows that there are no such relations. In the case of a shifted percolation threshold, in the three-dimensional case, although approximate, the reciprocity relations take place.

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ТЕРМОЕЛЕКТРИЧНІ КОМПОЗИТИ ТА СПІВВІДНОШЕННЯ ВЗАЄМНОСТІ

Розглянуто ефективні кінетичні коефіцієнти у макронеоднорідних середовищах, їхня поведінка при зміні концентрації фаз компонентів та завданні порога протікання. Знайдено комбінації ефективних кінетичних коефіцієнтів, за яких виконуються співвідношення взаємності. Бібл. 20, рис. 4.
Ключові слова: кінетичні коефіцієнти, двофазне середовище, однопотокові системи, термоЕРС.

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