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COMPARISON OF APPROACHES TO ACCOUNTING FOR IMPERFECT CONTACTS WHEN DETERMINING THE EFFECTIVE PERMEABILITY OF MATERIAL

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Abstract. The paper develops a complex approach to accounting for imperfect contacts (IC) when determining effective properties of various nature. The IC are assumed to be caused by various factors (microstructure features, process's specifity and so on). To obtain macroscopic properties, we seek a solution of the homogenization problem for the material containing isolated ellipsoidal inhomogeneities when fields are discontinuous at the interphase boundaries. The paper considers, generalizes and compares two existing approaches to accounting for the IC, namely, an approach where IC is modeled by means of a field jump specified in terms of a ratio of field values on the outer and inner sides of the inhomogeneity boundary, and approach, which introduces inhomogeneity with a surface effect. To take into account IC, we have considered an equivalent inhomogeneity with ideal contacts at the boundary. Working the problem on determining the effective diffusional permeability of material provided an example.

Keywords: effective properties, imperfect contact, equivalent inhomogeneity, effective diffusional permeability, homogenization problem

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СРАВНЕНИЕ ПОДХОДОВ К УЧЕТУ НЕИДЕАЛЬНЫХ КОНТАКТОВ ПРИ ОПРЕДЕЛЕНИИ ЭФФЕКТИВНОЙ ПРОНИЦАЕМОСТИ МАТЕРИАЛА

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Аннотация. В работе развивается комплексный подход к учету неидеальных контактов (НК), появление которых вызвано разнообразными факторами (особенности микроструктуры, специфика процесса на мезоуровне и т. п.), при определении эффективных свойств материала различной природы, представляемых тензорами второго ранга. Макроскопические свойства определяются путем решения задачи гомогенизации для материала, состоящего из матрицы и изолированных эллипсоидальных неоднородностей, на границе которых поля не являются непрерывными. Рассмотрены, обобщены и сопоставлены существующие подходы к учету НК: подход, при котором НК моделируют, вводя скачок поля на границе раздела фаз через задаваемое отношение значений поля по обе стороны границы, а также подход, при котором в рассмотрение вводится неоднородность с поверхностным эффектом. С целью учета НК при нахождении эффективных свойств материала, рассматривается эквивалентная неоднородность с идеальными контактами на границе, вклад которой в макроскопическое свойство эквивалентен вкладу исходной неоднородности. В качестве примера решена задача об определении эффективной диффузионной проницаемости материала.

Ключевые слова: эффективные свойства, неидеальный контакт, эквивалентная неоднородность, эффективная диффузионная проницаемость, задача гомогенизации

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Introduction

The properties of a material that is inhomogeneous at the microlevel directly depend on its structure and can be determined within the framework of continuum theory using homogenization methods. Physical fields are introduced into consideration, which, as a rule, are assumed to be continuous at the interphase boundaries. From a physical standpoint, this means that there are "ideal" contacts at the internal boundaries. At the same time, a number of phenomena should be described taking into account the presence of imperfect contacts, which can occur both due to the peculiarities of the microstructure of the material and in connection with the specifics of the described process [1–4].

As a rule, the issues of taking into account imperfect contacts to determine the effective properties are considered in the literature separately, in the context of describing processes that are different in nature. For example, some authors have drawn attention to the need to take into account the phenomenon of segregation when determining effective diffusion coefficients. This phenomenon is understood as the sedimentation of impurities in structural defects, which

is characteristic for mass transfer [2, 5–8]. Such a procedure was implemented by introducing a concentration jump in terms of the ratio of concentrations from the outer and inner sides of the interphase boundary (segregation parameter). Using this approach, the authors of [2] obtained the Voigt–Reuss and Hashin–Shtrikman boundaries for effective impurity mobility by expressing the diffusion flux in terms of a gradient of chemical potential (the potential was assumed to be continuous), after which the effective diffusion coefficients were determined directly. A constant segregation parameter in [5, 6] was introduced into the equations of the modified effective medium method. This parameter was introduced into the equations of effective field methods in [7, 8].

Approaches to accounting for imperfect contacts in determining the effective thermal or electrical conductivity of a material inhomogeneous at the microlevel have been considered separately in the literature [9–12]. It was believed that such contacts appear due to the presence of surface defects (roughness, delamination, etc.). Modeling of imperfect contacts was carried out by considering inhomogeneities with a surface effect (it was assumed that such inhomogeneities were covered with a layer with extreme properties, whose thickness tends to zero). The surface effect was taken into account either by determining the magnitude of the field jump from solving the problem of isolated inhomogeneity in an infinite matrix [9], or by approximating expressions for concentration tensors connecting the average fields inside the inhomogeneity with the applied field [10–12].

The similarity of the equations of diffusion, heat and electrical conductivity allows to make an assumption about the possibility of developing a unified approach to modeling imperfect contacts caused by different factors to determine the effective properties of varying nature for materials.

The goal of this study is to generalize and compare the available approaches to accounting for imperfect contacts in determining the effective properties for cases of materials with spheroidal and ellipsoidal inhomogeneities.

Statement of homogenization problem

The effective properties of the material are found by solving the homogenization problem for a representative volume V, which is a particle of a continuous medium at the macro level. Effective properties are expressed using tensor quantities relating the fields that are average in terms of representative volume. As a rule, it is assumed that the homogenized material satisfies the simplest linear governing relations. Due to the similarity of the equations of diffusion, heat and electrical conductivity, below we will limit ourselves to the consideration of the diffusion problem, for which Fick's law holds true:

$$< \mathbf{J} >_V = -\mathbf{D}^{eff} \cdot < \nabla c >_V, \tag{1}$$

where $\mathbf{D}^{eff}$ *is* the effective diffusion permeability tensor (the diffusion tensor of an impurity in a homogenized material), $\mathbf{J}$ is the diffusion flux, c is the concentration, ∇ is the nabla operator, $< ... >_V = \frac{1}{V}\int_V (...) dV$.

To find the fields to be averaged, the stationary diffusion problem is solved. The law of conservation in the absence of internal sources/sinks has the following form:

$$\nabla \cdot \mathbf{J}(\mathbf{x}) = 0, \tag{2}$$

where $\mathbf{x}$ is the radius vector of a point inside volume V.

The flux and concentration gradient at each point of the representative volume are related by a linear governing relation:

$$\mathbf{J}(\mathbf{x}) = -\mathbf{D}(\mathbf{x}) \cdot \nabla c(\mathbf{x}), \tag{3}$$

where $\mathbf{D}(\mathbf{x})$ is the diffusion permeability tensor of the material at point $\mathbf{x}$.

The independence of effective properties from the conditions at the boundary of the representative volume allows to choose them arbitrarily. It is convenient to set a homogeneous Hill condition, which in the case of the diffusion problem has the form $c(\mathbf{x})\big|_\Sigma = \mathbf{G}_0 \cdot \mathbf{x}$. Then the average value of the concentration gradient is completely determined by the boundary condition [13]:

$$< \nabla c >_V = \mathbf{G}_0. \tag{4}$$

The presence of boundaries within volume V (interphase boundaries Γ) requires imposing additional boundary conditions. These conditions will vary depending on the method of accounting for imperfect contacts.

Next, we consider a material consisting of an isotropic matrix characterized by a diffusion permeability tensor $\mathbf{D}_0 = D_0\mathbf{I}$ ($\mathbf{I}$ is the unit tensor), and ellipsoidal inhomogeneities with the volume V_1 with the permeability $\mathbf{D}_1 = D_1\mathbf{I}$, giving the conditions at the interphase boundaries.

One of the simplest methods known from the literature to account for imperfect contact is the introduction of a field jump at the interface between the matrix (+) and the inhomogeneity (−) using a constant ratio of field values from the outer and inner sides of the boundary. In the context of the diffusion problem, either the concentration field or the normal component of the flow can experience a jump. In the first case, the following conditions hold true at the interface of the phases Γ with the external normal $\mathbf{n}_\Gamma$:

$$D_0 \left.\frac{\partial c(\mathbf{x})}{\partial n_\Gamma}\right|_{\mathbf{x}\to\Gamma+} = D_1 \left.\frac{\partial c(\mathbf{x})}{\partial n_\Gamma}\right|_{\mathbf{x}\to\Gamma-}, \; c(\mathbf{x})\big|_{\mathbf{x}\to\Gamma+} = s_c\, c(\mathbf{x})\big|_{\mathbf{x}\to\Gamma-}, \tag{5}$$

where s_c is the segregation parameter; the jump is expressed as $[c] = (s_c - 1)c(\mathbf{x})\big|_{\mathbf{x}\to\Gamma-}$.

If there is a jump in the normal component of the flux J_n, the following conditions can be imposed by introducing the segregation parameter s_f into consideration:

$$D_0 \left.\frac{\partial c(\mathbf{x})}{\partial n_\Gamma}\right|_{\mathbf{x}\to\Gamma+} = s_f\, D_1 \left.\frac{\partial c(\mathbf{x})}{\partial n_\Gamma}\right|_{\mathbf{x}\to\Gamma-}, \; c(\mathbf{x})\big|_{\mathbf{x}\to\Gamma+} = c(\mathbf{x})\big|_{\mathbf{x}\to\Gamma-}. \tag{6}$$

In this case, the jump is defined as $[J_n] = (s_f - 1)\mathbf{n}_\Gamma \cdot \mathbf{J}(\mathbf{x})\big|_{\mathbf{x}\to\Gamma-}$.

Another way of accounting for imperfect contact is used for inhomogeneities with a surface effect. In the general case, inhomogeneities representing confocal ellipsoids are placed in the matrix, for which the conductivity of the inner ellipsoid is $\mathbf{D}_1 = D_1\mathbf{I}$, and the conductivity of the outer layer is $\mathbf{D}_s = D_s\mathbf{I}$.

The semi-major axes of the outer ellipsoid b_1, b_2, b_3 and the inner ellipsoid a_1, a_2, a_3 are related as follows:

$$b_i^2 = a_i^2 + \xi,$$

where $i = 1, 2, 3$; ξ is a constant.

Perfect contacts take place at the inner boundaries Γ_a of the inner ellipsoid of volume V_a with the outer normal $\mathbf{n}_{\Gamma_a}$ and Γ_b of the outer ellipsoid of volume V_b with the outer normal $\mathbf{n}_{\Gamma_b}$:

$$\begin{aligned} &D_0 \left.\frac{\partial c(\mathbf{x})}{\partial n_{\Gamma_b}}\right|_{\mathbf{x}\to\Gamma_b+} = D_s \left.\frac{\partial c(\mathbf{x})}{\partial n_{\Gamma_b}}\right|_{\mathbf{x}\to\Gamma_b-}, \; c(\mathbf{x})\big|_{\mathbf{x}\to\Gamma_b+} = c(\mathbf{x})\big|_{\mathbf{x}\to\Gamma_b-}, \\ &D_s \left.\frac{\partial c(\mathbf{x})}{\partial n_{\Gamma_a}}\right|_{\mathbf{x}\to\Gamma_a+} = D_1 \left.\frac{\partial c(\mathbf{x})}{\partial n_{\Gamma_a}}\right|_{\mathbf{x}\to\Gamma_b-}, \; c(\mathbf{x})\big|_{\mathbf{x}\to\Gamma_a+} = c(\mathbf{x})\big|_{\mathbf{x}\to\Gamma_a-}. \end{aligned} \tag{7}$$

To take into account the surface effect, it is necessary to pass to the limit at

$\xi \to 0$, as well as (in the context of the diffusion problem) either at $D_s \to 0$ or at $D_s \to \infty$. In the first case, corresponding to insulating coating, it is convenient to introduce an equivalent surface resistance into consideration

$$\beta = \frac{V_s}{D_s S_a} = \frac{4\pi\left(a_1^2 a_2^2 + a_1^2 a_3^2 + a_2^2 a_3^2\right)}{6 a_1 a_2 a_3 S_a} \lim_{\xi \to 0, D_s \to 0} \frac{\xi}{D_s}, \tag{8}$$

where $V_s = \lim_{\xi \to 0} V_b - V_a$, S_a is the surface area of the inhomogeneity with the volume V_a.

In the second case, corresponding to conductive coating, it is convenient to introduce an equivalent surface permeability

$$\lambda = \frac{D_s V_s}{S_a} = \frac{4\pi\left(a_1^2 a_2^2 + a_1^2 a_3^2 + a_2^2 a_3^2\right)}{6 a_1 a_2 a_3 S_a} \lim_{\xi \to 0, D_s \to \infty} \xi D_s. \tag{9}$$

The inhomogeneity with imperfect contacts can be formally replaced by an equivalent inhomogeneity with perfect contacts, which affects the effective properties in the same way as the initial one. To carry out such a replacement, it is necessary to determine the properties $\mathbf{D}^*$ that an equivalent inhomogeneity should possess. These properties will vary depending on the method of accounting for imperfect contact.

The introduction of equivalent inhomogeneity has the advantage that it becomes possible to use existing homogenization methods developed under the assumption of continuity of fields at the real interphase boundary. In this case, it is sufficient to substitute the corresponding diffusion coefficients of the impurity inside the inhomogeneity into expressions known from the literature. Since it is sufficient to take into account the presence of imperfect contacts at the stage of determining the diffusion permeability of equivalent inhomogeneity in this manner, here we will limit ourselves to qualitative and quantitative analysis of expressions for $\mathbf{D}^*$.

Note that imperfect contact, modeled by setting a concentration jump or by considering inhomogeneity with *insulating* coating, may occur when an impurity is aggregated at the interphase boundary. On the other hand, imperfect contact, which is modeled by setting another jump, namely, the normal component of the flux, or by considering inhomogeneity with *conductive* coating, may occur when additional diffusion paths are formed along the interphase boundary.

In view of this, it is of interest for each of these cases to compare two approaches to modeling imperfect contacts:

by setting the field jump in terms of the segregation parameter;

by considering an inhomogeneity with a surface effect.

The effective property can be expressed as a function of various microstructural parameters. This article uses the approach developed by Sevostyanov and Kachanov [13], where the role of the microstructural parameter is played by the sum of the tensors of the contribution of inhomogeneities. Below, we give the expressions for these tensors in the presence of imperfect contacts in the material, modeled using the approaches discussed above.

Contribution tensors

The contribution tensors are determined assuming that the inhomogeneities are isolated. If the concentration is set at the boundary of the representative volume, then the average gradient c over the representative volume is fully determined, while the average flux depends on the microstructure; it can be represented as a sum

$$< \mathbf{J} >_V = -\mathbf{D}_0 \cdot \mathbf{G}_0 + \Delta\mathbf{J}, \tag{10}$$

where $\Delta\mathbf{J}$ is the additional flux due to the presence of inhomogeneity.

Such an additional flux is a linear function of the applied field:

$$\Delta\mathbf{J} = -\frac{V_1}{V} \mathbf{H}^D \cdot \mathbf{G}_0, \tag{11}$$

where $\mathbf{H}^D$ is the tensor of the contribution of inhomogeneity to the diffusion permeability.

The contribution tensor can be found by solving the Eshelby problem for diffusion. The latter has an analytical solution only for ellipsoidal inhomogeneity. In this case, the contribution tensor can be expressed in terms of the concentration tensor, which linearly relates the field inside the inhomogeneity with the applied field.

Thus, to find the contribution tensor, it is necessary to solve the problem of averaging fields and find the concentration tensor. The presence of imperfect contacts should be taken into account at both stages.

A brief description of both stages is provided below.

Field averaging

We determine the average fields in the case of perfect contacts at the matrix/inhomogeneity interface, which in the framework of this study corresponds to a material with equivalent inhomogeneity, as well as modeling of imperfect contacts by various approaches.

According to the Ostrogradsky–Gauss theorem,

$$< \nabla c >_V = \frac{1}{V}\int_\Sigma \mathbf{n}_\Sigma c(\mathbf{x})\, d\Sigma, \quad < \mathbf{J} >_V = \frac{1}{V}\int_\Sigma \mathbf{n}_\Sigma \cdot \mathbf{J}(\mathbf{x})\mathbf{x} d\Sigma, \tag{12}$$

where $\mathbf{n}_\Sigma$ is the external normal to the surface Σ of the representative volume V.

Expressions (12) can be conveniently rewritten taking into account the interphase boundaries; in this case, the corresponding surface integrals should be added and subtracted. Then, with perfect contacts at the interface of the inhomogeneity of volume V_1, we obtain the known formulas:

$$< \nabla c >_V = \left(1 - \frac{V_1}{V}\right) < \nabla c >_{V_0} + \frac{V_1}{V} < \nabla c >_{V_1}, \quad < \mathbf{J} >_V = \left(1 - \frac{V_1}{V}\right) < \mathbf{J} >_{V_0} + \frac{V_1}{V} < \mathbf{J} >_{V_1}, \tag{13}$$

where $< ... >_{V_0} = \frac{1}{V_0}\int_{V_0} (...) \mathrm{d}V_0$, $< ... >_{V_1} = \frac{1}{V_1}\int_{V_1} (...) \mathrm{d}V_1$.

The case of a material with inhomogeneity with a coating of finite thickness characterized by finite properties is a particular case of a three-phase material with perfect contacts at its inner boundaries.

In this case, the average fields follow the expressions

$$\begin{gathered} < \nabla c >_V = \left(1 - \frac{V_b}{V}\right) < \nabla c >_{V_0} + \frac{V_a}{V} < \nabla c >_{V_a} + \frac{V_s}{V} < \nabla c >_{V_s}, \\ < \mathbf{J} >_V = \left(1 - \frac{V_b}{V}\right) < \mathbf{J} >_{V_0} + \frac{V_a}{V} < \mathbf{J} >_{V_a} + \frac{V_s}{V} < \mathbf{J} >_{V_s}, \end{gathered} \tag{14}$$

where $< ... >_{V_a} = \frac{1}{V_a}\int_{V_a} (...) \mathrm{d}V_a$ and $< ... >_{V_s} = \frac{1}{V_s}\int_{V_s} (...) \mathrm{d}V_s$.

In the presence of a concentration jump at the interface, the average concentration gradient should be determined as follows [6]:

$$< \nabla c >_V = \left(1 - \frac{V_1}{V}\right) < \nabla c >_{V_0} + \frac{V_1}{V} < \nabla c >_{V_1} + \frac{1}{V}\int_\Gamma \mathbf{n}_\Gamma [c] d\Gamma, \tag{15}$$

whereas the average flux is calculated using Eq. (13).

If the concentration jump is set based on the segregation parameter under condition (5), then it is convenient to rewrite Eq. (15) in the following form:

$$< \nabla c >_V = \left(1 - \frac{V_1}{V}\right) < \nabla c >_{V_0} + s_c \frac{V_1}{V} < \nabla c >_{V_1}. \tag{16}$$

The presence of a jump in the normal flux component leads to the need to use the following formula for the average flux [6]:

$$< \mathbf{J} >_V = \left(1 - \frac{V_1}{V}\right) < \mathbf{J} >_{V_0} + \frac{V_1}{V} < \mathbf{J} >_{V_1} + \frac{1}{V}\int_\Gamma [J_n] \mathbf{x} d\Gamma, \tag{17}$$

in this case, the average concentration gradient is determined by Eq. (13).

In the particular case, when the jump of the normal component of the flux is given in accordance with condition (6), the average flux is determined by the expression

$$< \mathbf{J} >_V = \left(1 - \frac{V_1}{V}\right) < \mathbf{J} >_{V_0} + s_f \frac{V_1}{V} < \mathbf{J} >_{V_1} . \tag{18}$$

Expressing $< \nabla c >_{V_0}$ in terms of $\mathbf{G}_0$, we obtain the following representations for the average flux:
for the material with equivalent inhomogeneity,

$$< \mathbf{J} >_V = -\mathbf{D}_0 \cdot \mathbf{G}_0 - \frac{V_1}{V}\left(\mathbf{D}^* - \mathbf{D}_0\right) \cdot < \nabla c >_{V_1}, \tag{19}$$

for the material with inhomogeneity and coating of finite thickness (characterized by finite properties),

$$< \mathbf{J} >= -\mathbf{D}_0 \cdot \mathbf{G}_0 - \frac{V_a}{V}\left(\mathbf{D}_1 - \mathbf{D}_0\right) \cdot < \nabla c >_{V_a} - \frac{V_s}{V}\left(\mathbf{D}_s - \mathbf{D}_0\right) \cdot < \nabla c >_{V_s} . \tag{20}$$

The following representations hold true for a material with a inhomogeneity with a field jump determined by the segregation parameter occurring at the interface:
if there is a jump in concentration,

$$< \mathbf{J} >_V = -\mathbf{D}_0 \cdot \mathbf{G}_0 - \frac{V_1}{V}\left(\mathbf{D}_1 - s_c \mathbf{D}_0\right) \cdot < \nabla c >_{V_1}, \tag{21}$$

if there is a jump in the normal flux component,

$$< \mathbf{J} >_V = -\mathbf{D}_0 \cdot \mathbf{G}_0 - \frac{V_1}{V}\left(s_f \mathbf{D}_1 - \mathbf{D}_0\right) \cdot < \nabla c >_{V_1} . \tag{22}$$

Representation of the contribution tensors in terms of the concentration tensors

The average concentration gradients included in expressions (19)–(22) can be expressed for the case of ellipsoidal inhomogeneity in terms of the applied field $\mathbf{G}_0$, for which the concentration tensors Λ_*, Λ_a, Λ_s, Λ_c, Λ_f are introduced, satisfying the equalities
$< \nabla c >_{V_1} = \mathbf{\Lambda}_* \cdot \mathbf{G}_0$ (for equivalent inhomogeneity),

$< \nabla c >_{V_a} = \mathbf{\Lambda}_a \cdot \mathbf{G}_0$ and $< \nabla c >_{V_s} = \mathbf{\Lambda}_s \cdot \mathbf{G}_0$ (for inhomogeneity with coating),

$< \nabla c >_{V_1} = \mathbf{\Lambda}_c \cdot \mathbf{G}_0$ (for inhomogeneity with a concentration jump, determined in terms of the segregation parameter, at the interface),

$< \nabla c >_{V_1} = \mathbf{\Lambda}_f \cdot \mathbf{G}_0$ (for inhomogeneity with a jump in the normal component of the flux, determined in terms of the segregation parameter, at the interface).

Expressions for these concentration tensors were obtained in [8, 10–14]. Taking into account these expressions and Eqs. (10), (11), we limit ourselves here to giving the final expressions for the contribution tensors of inhomogeneities:

$$\mathbf{H}^D = D_0 \sum_{i=1}^{3} \frac{D_{ii}^* - D_0}{D_{ii}^* A_i + D_0\left(1 - A_i\right)} \mathbf{e}_i \mathbf{e}_i \tag{23}$$

(for equivalent inhomogeneity with perfect contacts [13]);

$$\mathbf{H}^D = D_0 \sum_{i=1}^{3} \frac{D_1 - D_0 - D_0 D_1 \beta \dfrac{S_a}{V_a} A_i}{A_i D_1 + \left(1 - A_i\right) D_0 + \left(1 - A_i\right) D_0 D_1 \beta \dfrac{S_a}{V_a}\left(A_i - \dfrac{F_i}{H}\right)} \mathbf{e}_i \mathbf{e}_i \tag{24}$$

(for inhomogeneity with insulating coating);

$$\mathbf{H}^D = D_0 \sum_{i=1}^{3} \frac{D_1 - D_0 + \lambda \frac{S_a}{V_a}\left(1 - A_i\right)}{A_i D_1 + \left(1 - A_i\right) D_0 + A_i \lambda \frac{S_a}{V_a}\left(1 - A_i + \frac{F_i}{H}\right)} \mathbf{e}_i \mathbf{e}_i \tag{25}$$

(for inhomogeneity with conductive coating)

$$\mathbf{H}^D = D_0 \sum_{i=1}^{3} \frac{D_1 - s_c D_0}{A_i D_1 + s_c D_0 \left(1 - A_i\right)} \mathbf{e}_i \mathbf{e}_i \tag{26}$$

(in the presence of a concentration jump determined in terms of the segregation parameter s_c [8]),

$$\mathbf{H}^D = D_0 \sum_{i=1}^{3} \frac{s_f D_1 - D_0}{A_i s_f D_1 + D_0 \left(1 - A_i\right)} \mathbf{e}_i \mathbf{e}_i \tag{27}$$

(in the presence of a jump in the normal component of the flux, determined in terms of the segregation parameter s_f).

In the case of spheroidal inhomogeneity, for $a_1 = a_2 = a$, $\gamma = a_3/a$, the following equalities hold true:

$$A_1 = A_2 = f_0(\gamma),\ A_3 = 1 - 2 f_0(\gamma),$$

$$F_1 = F_2 = \frac{1}{a^2} \frac{1}{2} \left(\frac{1}{\gamma^2} f_0(\gamma) - \left(1 - 2 f_0(\gamma)\right) \right),$$

$$F_3 = -\frac{1}{a^2} \left(\frac{1}{\gamma^2} f_0(\gamma) - \left(1 - 2 f_0(\gamma)\right) \right),$$

where

$$f_0(\gamma) = \frac{1 - g(\gamma)}{2\left(1 - \gamma^{-2}\right)},\ g = g(\gamma) = \begin{cases} \dfrac{1}{\gamma\sqrt{1-\gamma^2}} \arctan \dfrac{\sqrt{1-\gamma^2}}{\gamma}, \gamma \le 1 \\ \dfrac{1}{2\gamma\sqrt{\gamma^2-1}} \ln \left(\dfrac{\gamma + \sqrt{\gamma^2-1}}{\gamma - \sqrt{\gamma^2-1}} \right), \gamma \ge 1. \end{cases}$$

In the case of spherical inhomogeneity, $F_1 = F_2 = F_3 = 0$, $f_0(\gamma) = 1/3$.

Note that according to the conclusions presented in monograph [9], where only spherical inhomogeneities were considered, the presence of insulating coating leads to a concentration jump at the matrix/inhomogeneity interface, and the presence of a conductive layer leads to a jump in the normal flux component, determined by solving the problem for composite inhomogeneity at the passage to the limit. This corresponds to the physical understanding of the phenomenon modeled, as noted above.

For the correct implementation of the procedure for comparing two approaches to modeling imperfect contact (by setting the field jump with the appropriate segregation parameter and by considering the inhomogeneity with the appropriate type of surface effect), we will determine what diffusion permeability $\mathbf{D}^*$ that an equivalent inhomogeneity whose contribution to the macroscopic property coincides with the contribution of inhomogeneity with imperfect contact modeled within the framework of different approaches should possess.

Equivalent inhomogeneity

Let us start by considering imperfect contact when an impurity is deposited as sediment at the matrix/inhomogeneity interface. When such a contact is modeled by setting a concentration jump in terms of the segregation parameter, it follows from the equality of the contribution tensor defined by expression (26) and the contribution tensor of equivalent inhomogeneity described by Eq. (23) that

$$\mathbf{D}^* = D^*\mathbf{I} = D_1/s_c\,\mathbf{I}, \tag{28}$$

that is, a material of equivalent inhomogeneity is isotropic.

Evidently, the components of the tensor $\mathbf{D}^*$ depend only on the segregation parameter and the diffusion permeability of the inhomogeneity, but do not depend on its shape. An increase in the segregation parameter leads to a decrease in the diffusion permeability of the equivalent inhomogeneity. In the absence of impurity sedimentation (at $s_c = 1$) $D^* = D_1$. Depending on whether the impurity is deposited at the interphase boundary from the outside or inside, the segregation parameter takes the values $s_c > 1$ or $s_c < 1$, respectively.

In the first case, $D^* < D_1$, which reflects the physics of the process, since the impurity penetrates the inhomogeneity to a lesser extent and, in order to achieve the same effect when considering an equivalent inhomogeneity, it is necessary to reduce its permeability.

In the second case, $D_* > D_1$, which is also physically justified, since the equivalent inhomogeneity should be more permeable to the impurity due to its accumulation inside the «real» inhomogeneity with imperfect contact.

At $s_c \to \infty$ $D^* \to 0$; this is due to the fact that the entire impurity accumulates outside the inhomogeneity and it is impermeable to the diffusant.

At $s_c = D_1/D_0$, we have $D^* = D_0$, i.e., a jump in concentration

$$[c] = \left(D_1 - D_0\right)/D_0\, c\left(\mathbf{x}\right)\Big|_{\mathbf{x}\to\Gamma-}$$

allows to ignore the presence of inhomogeneity when finding effective properties.

In the case of using the second approach to modeling imperfect contact, it follows from the equality of the inhomogeneity contribution tensor with equivalent surface resistance (see Eq. (24)) and the equivalent inhomogeneity contribution tensor (see expression (23)) that

$$\mathbf{D}_* = \sum_{i=1}^{3} D_1 \frac{1 - R\dfrac{S_a a_1}{V_a}\dfrac{D_0}{D_1}\dfrac{F_i}{H}\left(1 - A_i\right)}{1 + R\dfrac{S_a a_1}{V_a}\left(A_i - \dfrac{F_i}{H}\left(1 - A_i\right)\right)}\mathbf{e}_i\mathbf{e}_i; \tag{29}$$

Here, for convenience, a dimensionless parameter of equivalent surface resistance $R = D_1\beta/a_1$ is introduced.

The diffusion permeability tensor of equivalent inhomogeneity, expressed by Eq. (29), is generally orthotropic, and its symmetry group is determined by the shape of the inhomogeneity. In the absence of the surface effect (at $R = 0$), the tensor is isotropic and $\mathbf{D}^* = \mathbf{D}_1$ ($D_{11}^* = D_{22}^* = D_{33}^* = D_1$). In general, the diffusion coefficients D_{ii}^* can take values both greater and smaller than D_1. We should note that such features as redirection of the diffusion flux due to negative values of the components of the tensor $\mathbf{D}^*$, as well as the infinite permeability of equivalent inhomogeneity, can formally appear at certain values of structural characteristics (the ratio of the diffusion coefficients of the impurity in the matrix and in the inhomogeneity, the parameters of the inhomogeneity's shape, the value of equivalent surface resistance). Such cases require separate qualitative and quantitative studies, which is beyond the scope of this paper.

Expression (29) is significantly simplified in the case of spherical inhomogeneity: then the equivalent inhomogeneity is characterized by an isotropic tensor

$$\mathbf{D}^* = D_1/\left(1 + R\right)\mathbf{I}.$$

It follows from comparing this expression with expression (28) that two approaches to modeling imperfect contacts at the boundary of spherical inhomogeneities coincide when

$$s_c = 1 + R. \tag{30}$$

Let us turn to the consideration of imperfect contact, when additional diffusion paths are present in the material at the interphase boundary. When such a contact is modeled by setting the jump of the normal flux component in terms of the segregation parameter, it follows from the equality of the contribution tensors defined by expressions (27) and (23) that

$$\mathbf{D}^* = D^*\mathbf{I} = D_1 s_f \mathbf{I}. \tag{31}$$

The diffusion tensor $\mathbf{D}^*$, defined by Eq. (31), depends only on the segregation parameter and on the diffusion permeability of the inhomogeneity and does not depend on its shape.

An increase in the segregation parameter leads to an increase in the diffusion permeability of the equivalent inhomogeneity. In the absence of surface defects (at $s_f = 1$), $D^* = D_1$. At $s_f \to \infty$, the equivalent inhomogeneity is characterized by infinite permeability, regardless of the properties of the inhomogeneity (in this case, the entire impurity will instantly diffuse over the surface). In the case when $s_f = D_0/D_1$, the equality $D^* = D_0$ is satisfied.

When imperfect contact is modeled using the second approach, the equality of the contribution tensors defined by expressions (25) and (23) gives the following result:

$$\mathbf{D}_* = \sum_{i=1}^{3} D_1 \frac{1 + K\dfrac{S_a a}{V_a}\left(1 - A_i + \dfrac{F_i}{H}\right)}{1 + K\dfrac{S_a a}{V_a}\dfrac{D_1}{D_0}\dfrac{F_i}{H}A_i}\mathbf{e}_i\mathbf{e}_i, \tag{32}$$

where a dimensionless parameter of equivalent surface permeability $K = \lambda/(D_1 a_1)$ is introduced.

The diffusion permeability tensor of equivalent inhomogeneity, defined by expression (32), is generally orthotropic. In the absence of the surface effect (at $K = 0$), $\mathbf{D}^* = \mathbf{D}_1$. In the presence of the surface effect, the diffusion coefficients D_{ii}^* can take values both greater and smaller than D_1. In a certain range of values of structural characteristics, as in the case of an insulating coating, components D_{ii}^* can take values less than zero, which means from a physical standpoint that flux is redirected, as well as goes to infinity. Both cases require separate research beyond the scope of this study.

Expression (32) in the case of spherical inhomogeneities has the form

$$\mathbf{D}_* = D_1\left(1 + 2K\right)\mathbf{I},$$

it follows from here, taking into account Eq. (31), that two approaches to modeling imperfect contacts are equivalent when

$$s_f = 1 + 2K. \tag{33}$$

To summarize, the following qualitative differences can be observed between the two approaches to modeling imperfect contacts.

1. Taking into account imperfect contact by setting the field jump in terms of a constant segregation parameter, the symmetry group of the diffusion permeability tensor of equivalent inhomogeneity coincides with that for the initial inhomogeneity (in particular, it was shown above that the isotropy of the tensor $\mathbf{D}_1$ implies the isotropy of the tensor $\mathbf{D}^*$; we presented a more detailed study of the general anisotropic case in [8]). As a result, the components of the diffusion permeability tensor of equivalent inhomogeneity depend only on the physical properties of the inhomogeneity and the segregation parameter. In the case of modeling imperfect contact by considering inhomogeneity with a surface effect, the components of the tensor $\mathbf{D}^*$ depend both on the properties of the coating and the material of the inhomogeneity and on its shape.

Two approaches to modeling imperfect contacts produce the same results only in the case of a material with spherical inhomogeneities, provided that either equality (30) or (33) is satisfied, depending on the type of imperfect contact.

2. When taking into account imperfect contact by setting the field jump in terms of a constant segregation parameter, the components of the diffusion permeability tensor of equivalent inhomogeneity linearly depend either on the quantity $(s_c)^{-1}$, or on the parameter s_f. In the case of modeling imperfect contact by considering an inhomogeneity with a surface effect, the components of the tensor $\mathbf{D}^*$ depend non-linearly on the characteristics of the coating R or K (or their inverse quantities). At the same time, these dependencies, firstly, are different for different directions, and secondly, can take negative values at certain values of the characteristics of the structure, which, apparently, means redirection of the diffusion flux, as well as going to infinity. These cases need to be further investigated for compliance with the physical meaning of the modeled phenomenon.

Simulation results for imperfect contacts

Let us conduct quantitative analysis of the effect of the method of accounting for imperfect contact at the interface on the diffusion permeability of equivalent inhomogeneity using the example of a polycrystal.

A polycrystal is considered a two-phase material consisting of a matrix that models grain boundaries and elongated spheroidal inhomogeneities that model grains of lower diffusion permeability [7, 15]. For certainty, we take the values $D_1/D_0 = 0{,}2$, $\gamma = a_3/a = 100$ ($a_1 = a_2 = a$). In polycrystals, imperfect contacts can occur for various reasons, which should be modeled in different ways. Let us briefly describe them.

1. The phenomenon of segregation, which is characteristic of diffusion and which is understood as the sedimentation of impurities along grain boundaries from the outside, can be modeled either by setting a concentration jump using the segregation parameter s_c (I), or by considering an insulating coating with equivalent resistance R (II). Let us assume that $s_c = 1 + R$, which, on the one hand, is true for the case of a material with spherical inhomogeneities, on the other hand, satisfies the condition $s_c = 1$ at $R = 0$ in the case of perfect contacts in a material with inhomogeneities of arbitrary shape.

2. Due to cracking along grain boundaries, additional accelerated diffusion paths can be formed; they can be taken into account either by setting the jump of the normal flux component in terms of the segregation parameter s_f(III), or by considering conductive coating characterized by equivalent conductivity K (IV). For the same reasons as when choosing the dependence $s_c(R)$, we assume that $s_f = 1 + 2K$.

The dependences of the diffusion permeability of equivalent inhomogeneity in the presence of segregation, i.e., in the case of imperfect contact modeled by methods I and II, are shown in Fig. 1,*a*. An increase in the parameter R leads to a decrease in the components of the tensor $\mathbf{D}^*$. This, in turn, should subsequently (with further application of homogenization methods not considered in this study) lead to a decrease in the effective permeability of the material.

Note that the parameter R can formally take values from zero to infinity. To carry out quantitative analysis, however, we limited ourselves to considering a smaller range in which the flux does not change direction to the opposite, which would be the case with negative values of the coefficients D^*_{ii} and which, as noted above, requires additional analysis.

It is also worth noting that when using approach II, there is a difference in the behavior of the decreasing curves of the diffusion coefficients D^*_{33} along the symmetry axis of the inhomogeneity and the coefficients $D^*_{11} = D^*_{22}$ in the isotropy plane.

With the selected set of structure parameters, the coefficients $D^*_{11} = D^*_{22}$ change in the same way as the components of the isotropic tensor $\mathbf{D}^*$ introduced using approach I.

Fig. 1,*b* shows the dependences of the diffusion permeability of equivalent inhomogeneity in the presence of cracking, i.e., in the case of imperfect contact modeled by methods III and IV.

An increase in the parameter K leads to an increase in the components of the tensor $\mathbf{D}^*$, which vary in different ways, depending on the method of modeling imperfect contact, as well as on the direction in the case of approach IV. In the future, this type of change in the permeability of equivalent inhomogeneity should lead to an increase in the effective permeability of the material. Parameter K, like parameter R, can formally take values from zero to infinity, while at a certain value of K the component D^*_{33} will go to infinity, which, as discussed above, requires additional analysis beyond the scope of this paper.

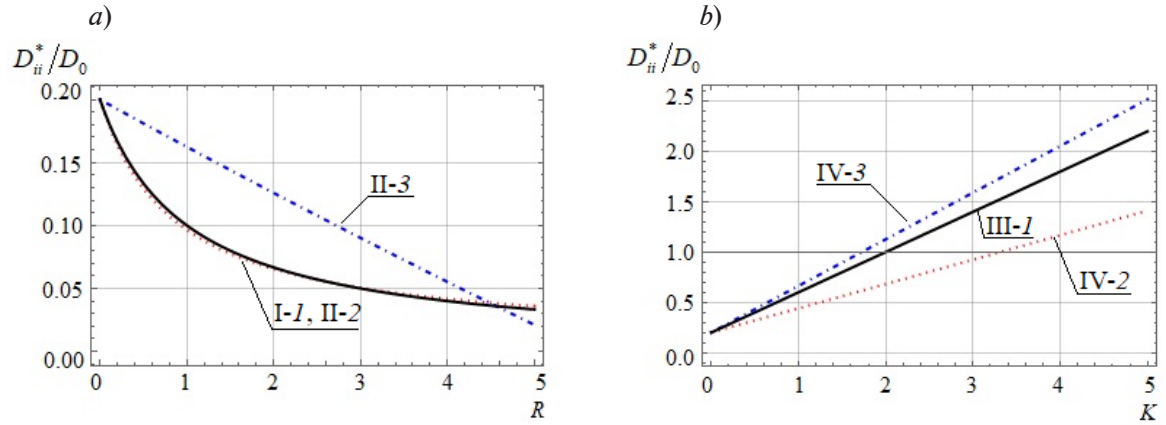

Fig. 1. Dependences of diffusion coefficients of the impurity in equivalent inhomogeneity on parameters $R(a)$ and $K(b)$ for the case of imperfect contact, simulated by methods I, II (a) and III, IV (b) (see explanations in the text).
The following diffusion coefficients are shown: $D_{11}^* = D_{22}^* = D_{33}^*$ (solid lines I-*1* and III-*1*) using approaches I and III; $D_{11}^* = D_{22}^*$ (dashed lines II-*2* and IV-*2*) and D_{33}^* (lines II-*3* and IV-*3*) using approaches II and IV

It is important that the components D_{ii}^* take values both smaller and larger than D_0, depending on the value of K (D_{ii}^*/D_0 can be either smaller or larger than unity).

Thus, the method of accounting for imperfect contact at the interface of the matrix and non-spherical inhomogeneity has a direct impact on the effective properties of the material. To choose the optimal approach, it is necessary to compare the results of numerical simulation with experimental data. The problem of such a comparison, in turn, involves difficulties in identifying the characteristics of the structure and requires separate study.

Conclusion

The paper proposes a generalization of the approaches available in the literature to modeling imperfect contacts at the interphase boundary of a material that is inhomogeneous at the microlevel in determining its effective properties of various nature.

It is taken into account that such contacts can occur in the material for different reasons: due to the particular internal structure and in connection with the specifics of the described process, which affects the physical interpretation of the model, but does not affect the mathematical framework used. A specific example of a diffusion problem was considered. The general case of a material with ellipsoidal inhomogeneities is considered and two approaches to modeling imperfect contacts are compared: by introducing a field jump (concentration or normal flux component) in terms of a constant segregation parameter and by considering inhomogeneity with a surface effect (respectively, with the presence of insulating or conductive coating).

We confirmed that the two approaches are equivalent only in the case of a material with spherical inhomogeneities, while in other cases these methods give qualitatively and quantitatively different results.

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