Influence of fractal substructures of the percolating cluster on transferring processes in macroscopically disordered environments

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Abstract. The presented work belongs to the issue of searching for the effective kinetic properties of macroscopically disordered environments (MDE). These properties characterize MDE in general on the sizes which significantly exceed the sizes of macro inhomogeneity. The structure of MDE is considered as a complex of interpenetrating percolating and finite clusters consolidated from homonymous components, topological characteristics of which influence on the properties of the whole environment. The influence of percolating clusters’ fractal substructures (backbone, skeleton of backbone, red bonds) on the transfer processes during crossover (a structure transition from fractal to homogeneous condition) is investigated based on the offered mathematical approach for finding the effective conductivity of MDEs and on the percolating cluster model. The nature of the change of the critical conductivity index \( t \) during crossover from the characteristic value for the area close to percolation threshold to the value corresponded to homogeneous condition is demonstrated. The offered model describes the transfer processes in MDE with the finite conductivity relation of «conductive» and «low conductive» phases above and below percolation threshold and in smearing area (an analogue of a blur area of the second-order phase transfer).

Description of the physical phenomena, in particular – transfer processes, in composite (heterogeneous) materials, which represent macroscopically disordered environments (MDE), is an important scientific-technical issue in different fields of the industry. To solve this problem, it is necessary to define (forecast), using the known physical properties, concentration and distribution of components in composite, effective kinetic transfer processes coefficients (thermal and electrical conductivity, dielectric and magnetic inductive capacitance, etc.), which characterise environment in general (on the sizes which significantly exceed the defining sizes of macro inhomogeneity, what does not require averaging over realizations [1])

MDE with highly heterogeneous properties of components are of special interest. Characteristic of such MDE is a cardinal change of system’s physical features upon transition through percolation threshold, upon which percolating (infinite) cluster is appeared; this cluster is a geometrical analogue of new condition emergence and appearance of order parameter in the theory of the second-order phase transfer [1 - 5]. Characteristics that inherent to MDE are fractal, they possess a scaling and sedately depend on proximity to percolation threshold at the systems close to percolation threshold. Above the percolation threshold, starting from the moment of occurrence, on scales that are less than correlation length percolating cluster and its substructures (backbone, skeleton of backbone, red
bonds, etc.) possess fractal dimensions (the values of which are presented in the works on the theory of percolation) that are less than dimensionality of space [2, 3]. Percolating cluster ceases to be fractal, becomes homogeneous and acquires dimensionality of space on scales that are bigger than correlation length. There is no percolating cluster below percolation threshold and the structure consisted from conductive phase elements (nodes, bonds, particles, etc.) represents a complex of finite clusters with different dimensions. While studying effective properties of such environments, the emphasis is usually on investigation of transition from one structure MDE to another one in the immediate vicinity to percolation threshold in smearing area (an analogue of a smearing area of the second-order phase transfer) [1, 4]. However, conductivity behavior in a distance from percolation transition, where qualitative change of structure happens upon transition from fractal state to homogeneous one (crossover), which, in our opinion, has not been studied enough, is also of interest. That is why this issue requires further development in the light of design of the unified model (approach) to forecast effective conductivity on the whole range of determining parameters (concentration, phase conductivity) changes, which takes in account an influence of percolating cluster’s fractal substructures on transfer processes not only in the area close to percolating threshold and in smearing area but also upon crossover.

The work is limited to consideration of two-component third-dimensional MDE. The model is based on presenting MDE as aggregates of different combinations of interpenetrative and interdependent percolating clusters and finite clusters compiled of components’ (phases’) elements (nodes, bonds, particles), which possess equal probability of distribution in the area and have the following characteristics: θ₁, θ₂ - volumetric concentration and λ₁, λ₂ - conductivity of the first and the second components, respectively. It is accepted that λ₁ ≥ λ₂, ν = λ₂/λ₁ ≤ 0. The model that describes transfer processes in MDE with finite conductivity relation of «conductive» and «low conductive» phases above and below percolation threshold is considered.

Above a percolation threshold the model describes effective conductivity through four-parametric approximated correlation [6]

\[ \Lambda_e = \Lambda_{01} + \Lambda_{02} \nu + \nu(1-\Lambda_{01} - \Lambda_{02})^2 / \left( \nu(\theta_1 - \Lambda_{01}) + (\theta_2 - \Lambda_{02}) \right) \]  \hspace{1cm} (1)

\[ \Lambda_{01} = \lambda_{01}/\lambda_1, \quad \Lambda_{02} = \lambda_{02}/\lambda_2, \quad \lambda_{01} - \text{conductivity of the first component’s percolating cluster (under condition } \lambda_2 = 0 \text{), } \lambda_{02} - \text{conductivity of the second component’s percolating cluster (under condition } \lambda_1 = 0 \text{).} \]

Conductivities \( \Lambda_{01} \) and \( \Lambda_{02} \) consider a topology of percolating clusters and they are integral non-dimensional characteristics of MDE structure.

The formula (1) is a «cover» for a definition of MDE’s effective conductivity. An accuracy of calculations depends on used values of percolating cluster’s conductivity (\( \Lambda_{01} \) and \( \Lambda_{02} \)), which should take into account a nature of percolating cluster and its substructure formation, which depends on physical and mechanical components’ properties, technology of consolidation processes and other practical features. Such data usually can be obtained through observational [7, 8] or model approaches [1, 9, 10, 11]. By virtue of symmetry of the given MDE the influence of structural characteristics of percolating cluster on its conductivity will be investigated by the example of conductive phase’s percolating cluster’s formation.

One of the main characteristics of percolating cluster is its density (power) that, when it is close to percolation threshold, is described by exponential function

\[ \theta_1 \propto (\theta_1 - \theta_c)^\beta, \] \hspace{1cm} (2)

\( \theta_c \) - percolation threshold (a minimal critical concentration of component, upon which percolating cluster arises), \( \beta = \nu(d - d_f) = 0.417 \pm 0.003 - \text{fractal critical index, } d_f = 2.54 \pm 0.008 - \text{fractal dimension of percolating cluster, } d = 3 - \text{dimension of 3-dimensional space} \) [2].

Its another characteristic is contiguity (contact) \( C_c \) - ratio of specific lengths of contact and complete percolating cluster particles’ surfaces that may be represented as a ratio of specific non-
dimensional surfaces of contact \( \alpha_t \) and rated cross-section \( \theta_t \) of percolating cluster \( C_t = \alpha_t/\theta_t \) or as a common non-dimensional contact cross-section of one particle belonged to percolating cluster [8]. In accordance with the law of multiplication of probabilities, percolating cluster contiguity equals the product of probability \( \theta_t \) of contact and instantaneous probability \( z_t \) of this contact preservation: 
\[
C_t = \theta_t z_t \quad [8].
\]
Under component’s particles’ equality of rights \( z_t = \theta_t \), consequently \( C_t = \theta_t^2 \).

It is known from percolation theory that only active part of percolating cluster is involved in transfer processes. This active part is backbone, density of which, when it is close to percolation threshold, is described by correlation
\[
\theta_0 \propto (\theta_t - \theta_c)^b, \quad (3)
\]
\[b = \sqrt{(d - d_b)} = 1.002 \pm 1\] - fractal critical index of backbone, \( d_b = 1.885 \pm 0.015 \) - fractal dimension of backbone [2]. In backbone, the value of contact cross-section coincides with critical cross-section \( \alpha_{01} \), in which directional tension or processes concentrate [8], and its contiguity (critical) \( C_{01} = \alpha_{01}/\theta_{01} \) is a ratio of specific non-dimensional surfaces of critical cross-section \( \alpha_{01} \) and rated cross-section \( \theta_{01} \). This contiguity can be also considered as a concentration of critical cross-section in cross-section of backbone phase. Under component’s particles’ equality of rights \( C_{01} = \theta_{01}^2 \).

According to [11], non-dimensional conductivity of percolating cluster \( \Lambda_{01} \) can be represented as a dependence on two non-dimensional topological characterizations - density (power) \( \theta_0 \) and critical cross-section (under coincidence of contact and critical cross-sections) \( C_{01} \) of percolating cluster’s active part, which is involved in transfer processes
\[
\Lambda_0 = \theta_0 \sqrt{C_{01}}. \quad (4)
\]
Then we will deduce from (4) with provision for (3)
\[
\Lambda_{01} = \theta_{01} \sqrt{C_{01}} = \theta_{01}^2 \propto (\theta_t - \theta_c)^{2b}. \quad (5)
\]
In percolation theory, percolating cluster conductivity is described by formula
\[
\Lambda_{01} \propto (\theta_t - \theta_c)^t, \quad (6)
\]
t – critical index of conductivity, the value of which, according to different data, varies from 1.6 to 2.0 for third-dimensional space [1 – 4, 9]. From the expressions (5) and (6) follow \( t \approx 2b \approx 2 \), what provides support for the opted approach to a definition of percolating cluster conductivity. Scatter of values of critical index \( t \) in our opinion, can be explained by processes of formation of percolating cluster’s fractal structure. At the moment of emergence of percolating cluster, a fraction of elements (particles) in it is negligibly small and consists of (in the main) single bonds, under disruption of which transfer process ceases – red bonds. Density of red bonds is described by correlation \( \theta_{0red} \propto (\theta_t - \theta_c)^{b_{red}} \), where \( b_{red} = \sqrt{(d - d_{red})} - fractal critical index of red bonds, d_{red} = 1.143\) - fractal dimension of red bonds [2]. Cross-section (contact) of percolating cluster \( C_1 \), which can be considered as a common non-dimensional contact cross-section of one particle belonged to percolating cluster, will be coincide with non-dimensional critical cross-section of particle belonged to red bonds. That is why \( C_1 \) will be critical cross-section for red bonds. Then we will deduce from (4)
\[
\Lambda_{01} = \theta_{0red} \sqrt{C_1} = \theta_{0red} \theta_1 \propto (\theta_t - \theta_c)^{b_{red} + b}. \quad (7)
\]
Similarly, it is possible to obtain an equation also for a further phase of percolating cluster development that is a formation of a skeleton of backbone which consists of red bonds.
\[
\Lambda_{01} = \theta_{sk} \sqrt{C_1} = \theta_{sk} \theta_1 \propto (\theta_t - \theta_c)^{b_{sk} + b}. \quad (8)
\]
b_{sk} = \sqrt{(d - d_{sk})} - fractal critical index of skeleton of backbone, \( d_{sk} = 1.37 \) - fractal dimension of skeleton of backbone [2].
In a distance from a percolation threshold, on a scale coming to the correlation length \([1 - 4]\) for the percolating cluster with fractal dimension \(d_f\), contact contiguity of the percolating cluster coincides with critical contiguity of backbone

\[
\Lambda_{01} = \theta_{01} \theta_1 \propto (\theta_1 - \theta_c)^{t_{\beta} + b}.
\]  

(9)

From comparison the equation (6) with (7), (8), (9), respectively for the considered cases, follows \(t = 2.042, t = 1.843, t = 1.472\). The obtained result shows that the critical index of conductivity \(t\) is the variable depending on formation of the conductive part of the percolating cluster which is possible to be presented as a function \(t = f(\theta_1 - \theta_c)\). When finding such dependence, it is necessary to consider that the termination of the crossover comes at different values of concentration. It is explained by hierarchical structure of the percolating cluster for which at achieving of a state when, for example, the percolating cluster in general is not fractal, its "internal" structure (backbone, skeleton of backbone, red bonds) still remains fractal, and at achieving in general of a homogeneous condition of backbone, its "internal" structure (skeleton of backbone, red bonds) still remains fractal and so on. In works \([10, 11]\) approach of finding the limits of fractal conditions existence of the percolating cluster and the backbone is offered. The critical value of the conductive component concentration defines border of a fractal percolating cluster existence as a "top" percolation threshold \(\theta^*_c = \theta_c (1 - \beta)^{-1}\), which is defined by the universal critical indicator \(\beta\), that does not depend on percolation type but only on a problem space dimension and value \(\theta_c\) is found. Use of value \(\theta^*_c\) has allowed to obtain an equation for calculation of the fractal percolating cluster density \(\theta_1(\theta_1) = \theta^*_c (\theta_1 - \theta_c)^0/\theta^*_c (\theta_1 - \theta_c)^b\). At the same place options of finding the range of fractal skeleton existence are considered. In figure 1 the correlation \(t = f(\theta_1 - \theta_c)\) obtained on the basis of comparison the critical index of conductivity values \(t\) to data of the percolating analysis of the percolating cluster behaviour and its substructures from the percolation threshold to the termination of the crossover is presented.

Figure 1. Correlation \(t = f(\theta_1 - \theta_c)\)
A type of the correlation was approximated by Gaussian normal probability distribution. For unambiguity equation for finding a conductivity of the percolating cluster was written the following \( \Lambda_{01} = (1 - \vartheta) - (\vartheta_1 - \vartheta) \) and value of a percolation threshold is \( \vartheta_c = 0.156 \) [11]. In view of symmetry of the structure of the taken MDE, \( \Lambda_{02} \) is defined by analogy with \( \Lambda_{01} \).

The analysis of a formula (1) shows that in the range \( \vartheta_c \leq \vartheta \leq 1 \) (above a percolation threshold) it maintains limit transitions: \( \nu = 1 \rightarrow \Lambda_{\nu} = 1 \), \( \nu = 0 \) (both for a case at \( \vartheta_2 = 0 \), and at \( \vartheta_1 \rightarrow \infty \)) \( \rightarrow \Lambda_{\nu} = \Lambda_{01} \) and at the corresponding combinations of values \( \Lambda_{0i} \) and \( \nu \) it is also transformed in a row of the familiar formulas: at \( \Lambda_{02} = \vartheta_2 - \) to a formula for parallel layered systems to a stream, and at \( \Lambda_{01} = 0 \) and \( \Lambda_{02} = 0 \) for layered systems that are perpendicular to a stream, at \( \Lambda_{01} = \vartheta_1^2 \) for systems with the interpenetrating components, at \( \nu = 0 \) and \( \Lambda_{02} = 0 \) is transformed to the equation \( \Lambda_{0i}(\vartheta_i) = \vartheta_i \) applied to calculation of porous bodies conductivity with the isolated isometric pores [6, 9]. Below the percolation threshold (\( \vartheta_i < \vartheta_c \)) the formula (1) does not consider formation and growth of finite clusters to the size of a percolating cluster and describes conductivity only of MDE with isolated inclusions of the conductive component.

At the creation of model that is lower than a percolation threshold (\( \vartheta_i < \vartheta_c \)) a formula (1) has been taken as a basis. It is proceeded from that at the moment of the percolating cluster’s conductive component emergence (strictly at \( \vartheta_i = \vartheta_c \)) its density will be \( \vartheta_i^i = \vartheta_c^i \) (taking into account the preservation probability of meeting the elements included into the percolating cluster, according to the theory of physics and statistics of porous bodies [8]), and its resistance will be \( R_i = \vartheta \vartheta_c^2 \). Taking into account an introduction of a conditional (“displaced”) percolating cluster with the density \( \vartheta_c^2 \) (instead of the conductive percolating cluster) consisting of two consistently connected parts which elements are only of the first component with the density \( \vartheta_c^2 \) and the resistance \( \nu \vartheta_c^2 \), and of elements only of the second component with the density and the resistance \( \vartheta_c^2 - \vartheta_1^2 \), a correlation is obtained

\[
\Lambda_{\nu} = \lambda_{\nu} = \frac{\vartheta_i^4}{\nu \vartheta_i^2 + (\vartheta_i^2 - \vartheta_1^2) + (\vartheta_i^2 - \vartheta_1^2)} + \frac{\vartheta_i^2 - \vartheta_1^2}{\sqrt{(\vartheta_i^2 - \vartheta_1^2) + (\vartheta_1^2 - \vartheta_2^2)}} .
\]

The formula (10) maintains limit transitions for cases: \( \vartheta_i = 0 \rightarrow \Lambda_{\nu} = 1 \); \( \nu = 1 \rightarrow \Lambda_{\nu} = 1 \); at \( \nu = 0 \) (\( \lambda_{\nu} \neq 0 \) and \( \vartheta_i = \vartheta_c \) the condition \( \Lambda_{\nu} \rightarrow \infty \) is also fulfilled. From formulas (1) and (10) it follows that \( \Lambda_{\nu} = 0 \) (\( \lambda_{\nu} = 0, \lambda_1 \neq \infty \)) and \( \left(\Lambda_{\nu}\right)^{-1} = 0 \) (\( \lambda_{\nu} \neq 0, \lambda_1 = \infty \)) at \( \nu = 0 \) and \( \vartheta_i = \vartheta_c \). At \( \nu \neq 0 \) equations (1) and (10) allow to find the correlation \( \Delta = f(\nu) \) (at \( \nu = 0 \rightarrow \Delta = 0 \)) considering that in the smearing area \( \vartheta_c - \Delta \leq \vartheta_i \leq \vartheta_c + \Delta \) [1] a condition \( \Lambda_{\nu} = \left(\Lambda_{\nu}\right)^{-1} \) has to be fulfilled.

Thus, the offered model for calculation of effective conductivity of MDE "works" in a whole area of changing the basic parameters:

- below a percolation threshold, out of the smearing area \( 0 \leq \vartheta_i \leq \vartheta_c - \Delta \), effective conductivity is calculated by a formula (10);
- above a percolation threshold, out of the smearing area \( \vartheta_c + \Delta \leq \vartheta_i \leq 1 \), effective conductivity is calculated by a formula (1);
- in the smearing area \( \vartheta_c - \Delta \leq \vartheta_i \leq \vartheta_c + \Delta \) effective conductivity is calculated from a condition

\[
\lambda_{\nu} \Lambda_{\nu} \vartheta_i + \Delta = \lambda_{\nu} \Lambda_{\nu} \vartheta_c + \Delta = \lambda_{\nu} \Lambda_{\nu} \vartheta_i - \Delta .
\]

Calculations of effective MDE’s conductivity in a concentration change range \( 0 \leq \vartheta_i \leq 1 \) (covers all stages of MDE structure formation) and correlation of components’ conductivity \( 0 \leq \nu \leq 1 \)
(including threshold cases \( \lambda_2 = 0, \lambda_4 \neq \infty \) and \( \lambda_2 \neq 0, \lambda_4 = \infty \)) have been made with the aid of the offered model. Comparison of the calculations’ results shows that the results have a good coherence with the known observational and model results that were obtained during the investigation of conductivity of MDE, a structure of which is characteristic for a certain stage of its formation: below percolation threshold, in smearing area, above percolation threshold, in crossover area and in area of homogeneous condition. Inaccuracy is comparable with scatter of observational values, resulting from technological fluctuation of determining parameters. The conclusion illustrates a universality of the given approach.

The offered model allows to predict values of effective transferring coefficients in the wide range of changing the basic parameters, to consider the real structural characteristics of MDE connected with physical and mechanical properties of components, technology of consolidation processes, a percolation deviation from standard [5] and other features of MDE formation by using the values \( \Lambda_{bi} \) obtained in either the experimental or model ways. It can be also useful while forecasting the other effective kinetic properties.

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