Non-classical transport in highly heterogeneous porous media

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Abstract. The review is presented on non-classical impurity transport in highly heterogeneous media with various correlations of structure characteristics. Regularly heterogeneous, statistically homogeneous double-porous and fractal media are considered. Concentration distributions in the main body and at asymptotically large distances are described. The effects of sorption onto colloids are analyzed.

1. Introduction

In this paper the physical models developed by the authors for non-classical impurity transport are reviewed. By non-classical (anomalous) regimes are implied the regimes for which the size of main body localization depends on time according to

\[ R \sim t^\gamma, \quad \gamma \neq 1/2. \] (1)

We have super-diffusion when \( \gamma > 1/2 \) and sub-diffusion at \( \gamma < 1/2 \). Anomalous transport processes occur in a variety of natural Sciences, such as plasma physics \([1, 2]\), semiconductor physics \([3, 4]\), astrophysics \([5]\), biophysics \([6, 7]\), hydrology \([8, 9]\), and many others \([10]\). A special place in this series is occupied by geological media. An extensive amount of field observations accumulated in the last decades evidences that in many cases classical regularities based on Darcy and Fick laws can not describe contaminant transport processes in geological media so that discrepancies may be of several orders \([11]\). It is to this range of tasks that our studies were oriented.

Historically, the first models describing non-classical transport date back to the 30s of the last century. Apparently, the first was the work of Khinchin and Levy \([12]\), in which the migration of a particle was considered as a result of its successive jumps of varying length and duration. The probabilities of the jumps were determined by the distribution function, and if the given function decreased sufficiently slowly with increasing these parameters, then the mean square displacement of the particle grew with time according to a nonclassical law. Later, this approach was actively used in other models for non-classical transport, such as CTRW \([13]\), equations with fractional derivatives \([14]\), models with non-Markov processes \([15]\), nonlinear Fokker-Planck equation \([16]\).

In our studies, a different approach was adopted. It is based on the averaging of equations in which impurity transport is caused by classical physical mechanisms, taking into account the distribution of inhomogeneities characteristic of a particular type of medium. The specificity of transport in geological environments is that the transport of impurities occurs as a solution in groundwater through channels formed by voids in the rock (pores, fractures). The mechanisms of transport are advection, that is, transport with the speed of local flow, and molecular diffusion in solution. As a result, the equation for the concentration in the channels has the form of a law of conservation of mass, with a flow of the classical form, including advection and diffusion.

To further construct the theory, it is necessary to average this equation over space, or over an ensemble of realizations. At the same time, the following properties of the medium are the key: first,
the geometry of the channels, and secondly, a strong contrast of the properties characterizing the individual regions.

As for the first factor, conditionally all media from the point of view of correlation in the distribution of inhomogeneities can be divided into three classes: 1) the simplest type is regularly heterogeneous sharply contrasting media, such as a fracture in a porous medium; 2) further, statistically homogeneous media - when , starting from a certain averaging scale, the result does not depend on the scale, so that we can introduce average permeability, average velocity, etc., and 3) these are percolation media, or media that, within a certain large scale have fractal properties.

The second factor, fundamental for the emergence of nonclassical regimes, is a sharp contrast between the properties of neighboring regions. Indeed, if we have a fractured-porous medium and the fractures in it are of sufficiently large apertures, then the velocity of percolation through the fractures will be much greater than over the surrounding porous matrix. The difference, as a rule, is so great that in the main approximation we can neglect the leakage in the porous matrix and consider here only the diffusion. As a result, we can assume that the medium consists of two subsystems - strongly and slightly permeable. This approximation is called a double-porous medium [17]. Examples of such a medium are isolated cracks in the porous matrix, fractured-porous media with a continuous distribution of fractures, sandy-clay formations. Next, non-classical transport will be considered in regularly inhomogeneous (section 2), percolation (section 3) and statistically homogeneous sharply contrasting (section 4) media. Section 5 is devoted to an analysis of the effect of sorption effects on transport. We summarize our results in section 6.

2. Regularly heterogeneous media

2.1. Statement of the problem

Transport in regularly inhomogeneous media was considered in the following formulation. The medium consists of two regions - a geometrically regular, strongly permeable region I, which we call a crack, and the rest of the space (region II) filled with a porous but weakly permeable matrix. Two types of cracks were considered: a) a plane-parallel layer of thickness, and b) an infinite cylinder.

Let us consider case a). In both regions, the transfer is determined by the diffusion mechanism, so that for the concentration \( n(\vec{r}, t) \) in the region I the equation is valid

\[
\frac{\partial n}{\partial t} = D \Delta n ,
\]  

and for the concentration \( c(\vec{r}, t) \) in the region II we have
\[
\frac{\partial c}{\partial t} = d \Delta c .
\]  \hspace{1cm} (3)

There is a strong inequality \( D >> d \). On the boundary of two regions, the conditions for equality of concentrations and normal flows are valid. At the initial time, the entire impurity is concentrated in the crack at the origin. This model was first proposed by A.M. Dykhne, therefore we call it the Dykhne model [18].

2.2. Behavior of the main body of impurity localization

Qualitative analysis has shown that the nature of the transfer in such a system depends on the time interval. At short times, \( t << t_i \), where

\[
t_i = a^2/4d ,
\]  \hspace{1cm} (4)

the impurity particles do not yet have time to go beyond the crack, and the transport along the crack occurs according to the classical diffusion law with a coefficient \( D \). However, with time, particles will spend in the fracture less and less time, and basically they will be in the matrix. The fraction of time that the particles spend in the fracture and diffuse in the fracture plane with a coefficient \( D \) will be approximately equal to \( \tau(t) \approx \frac{a}{\sqrt{dt}} \). In accordance with this, the size of the impurity cloud in the crack plane will grow according to

\[
R^2 \approx \int_{t_i} D\tau'(t') dt' \approx D\sqrt{t_i} .
\]  \hspace{1cm} (5)

The transfer along the matrix at this stage gives a very small contribution, and thus the matrix acts as a trap. This leads to the appearance of a nonclassical, sub-diffusion regime: \( R \sim t^{1/4} \). Similarly, for a fast medium in the form of an infinite cylinder, the size of the impurity cloud along the axis of the cylinder increases according to the law

\[
R \approx \sqrt{D\tau_1 \ln(t/t_i)} .
\]  \hspace{1cm} (6)

At long times, the fraction of time spent by the particles in the crack is negligible, so that the particle practically always spends in a slow medium. As a result, the transfer is determined by the properties of the slow medium, that is, classical diffusion, but with a small coefficient \( d \).

Similarly, a model can be considered in which, along with diffusion, advection with constant velocity \( u \) is also taken into account in the crack [19]. In this case, the behavior of the system depends on the Peclet number \( Pe = \frac{ua}{d} \), which determines the relative speed of the diffusion and advection processes. When \( Pe << 1 \), the impurity behavior is described by the same laws as in the simple Dykhne model. When \( Pe >> 1 \) new non-classical regimes arise which are quasi-diffusive ones. They are characterized by the fact that the average displacement of particles increases with time according to the same laws as the size of the cloud. The velocity of the front motion depends on the shape of the crack. For a plane crack, the displacement of the front is proportional to the root of time, and for an infinite cylinder, to the logarithm of time.

2.3. Concentration asymptotics at large distances

All the above regimes can be obtained from the exact solution of the problem, which confirms the results obtained above on the basis of semi-qualitative arguments [20, 21]. But an exact solution also makes it possible to calculate the behavior of the concentration at asymptotically large distances from the source, \( r >> R(t) \). The calculation shows that the asymptotical behavior at large distances, again, depends on the time interval. At short times, the concentration decreases due to Gauss, and is characterized by a high diffusion coefficient, \( D \). When in the main cloud the transport regime changes to sub-diffusion, the asymptotics becomes two-stage. The near segment is described by a stretched
exponential and is determined by sub-diffusion, and the far segment is described by a Gaussian exponential with a large diffusion coefficient. Namely, it is determined by the mode from the previous time interval. At the latest times, the asymptotics become three-stage: the two remote parts are the same as in previous time interval, and the nearest part is determined by the current regime of slow classical diffusion. The structure of the asymptotics at large times is shown schematically in Fig. 2.

Thus, from the Dykhne model it is possible to draw the following conclusions: in sharply contrasting media, the weakly permeable regions serve as traps that greatly retard the rate of impurity transport. This slowing leads to the appearance of sub-diffusion regimes, which essentially depend on the geometry of the medium and the mechanisms of transport. The change in the transport regimes that occurs in time in the main cloud leads to the formation of a multi-stage distribution of the concentration at asymptotically large distances. The following regularity was established for the concentration distribution in asymptotics: the more distant parts of the asymptotics are determined by the earlier modes of transport in the main cloud.

![Figure 2. Multi-stage structure of concentration at asymptotically far distances from the source.](image)

### 3. Percolation media

Now we consider the media in which the system of channels with high permeability has the form of a percolation cluster. In nature, this situation is often found in fractured formations, as well as in porous media, if there is a strong scatter of pores in size. A peculiarity of percolation media is that in the range of large spatial scales up to the correlation length $\xi$, they are fractal and have the property of scale invariance. At scales greater than correlation length $\xi$, they are statistically homogeneous.

Note that the percolation clusters themselves have an internal structure - they consist of a backbone that connects remote parts of the clusters providing long-range transport, and dead ends that are finite and join the core at a single point.

The equation for the microscopic concentration $\hat{c}$ during the transport through the percolation cluster is the standard conservation law, where the flux is determined by advection in a random velocity field:

$$\frac{\partial \hat{c}}{\partial t} + \text{div}(\hat{V}\hat{c}) = 0$$

(7)

Our goal is to describe the impurity transport in term of the concentration averaged over the ensemble of realizations, $\bar{c} = \langle \hat{c} \rangle$. The velocity field was considered to be incompressible, which is valid in describing the flow of groundwater. As the first step, an isotropic medium with a correlation length tending to infinity was considered. In this case, the velocity averaged over the ensemble of realizations is zero, so that the main quantity determining the transport is the velocity correlator.
Due to the property of scale invariance, under the transformation of similarity $\vec{r} \rightarrow \lambda \vec{r}$, an arbitrary quantity $A$ transforms according with the relation $A \rightarrow \lambda^{\Delta} A$. The value $\Delta$ is called as the scale dimension of the quantity $A$. Applying this property to the pair velocity correlation function $\langle \dot{V}(\vec{r})\dot{V}(\vec{r}) \rangle$, we conclude that in an isotropic homogeneous space this function has the form

$$\langle \dot{V}(\vec{r})\dot{V}(\vec{r}) \rangle \propto V^2 \left( \frac{a}{|\vec{r}_1 - \vec{r}_2|} \right)^{2h},$$

where $h$ is the velocity scale dimension.

Turning to the description of the concentration averaged over the ensemble of realizations, we arrive at an integro-differential equation, the study of which was carried out in different ways: on the basis of an analysis of scale dimensions [22], and using the Feynman diagram technique [23]. Both methods lead to similar results, which consist in the following.

The impurity transport mode depends on the rate of decrease of the correlation function, i.e. on the value of parameter $h$. When $h > 1$, the transport corresponds to the classical diffusion regime, so that the concentration is described by a Gaussian profile, including the asymptotically large distances. If $h < 1$, the super-diffusion regime is realized, so that the size of the impurity cloud increases with time according to the law

$$R \approx (a^\beta V t)^{\frac{1}{1-h}}.$$  

In this case, the decrease of concentration asymptotics is described by a compressed exponential, that is, it turns out to be faster than in the classics [22].

For the applicability of the random advection model to real media, the influence of the following factors was considered: the finite value of the correlation length, the anisotropy of percolation media, and the effect of traps (dead ends and a weak permeable porous matrix surrounding percolation cluster).

If the correlation length is finite, $\xi < \infty$, the behavior of the impurity is described as follows [24]. At times $t < t_\xi$, when the size of the cloud of impurity is smaller than $\xi$, the transport mode is the same as in an infinite fractal medium. Therefore, the transport under the condition $h < 1$ occurs in the super diffusion regime, according to (10). At times $t > t_\xi$ when the size of the cloud exceeds $\xi$, the medium becomes statistically homogeneous, and, consequently, the transport becomes a classical diffusion. The effective diffusivity $D_\xi$ and the characteristic time $t_\xi$ are given by the equalities

$$D_\xi \approx \frac{\xi^2}{t_\xi}, \quad t_\xi \approx \frac{\xi}{Va^\beta}.$$  

Asymptotics of the concentration at large distances were also obtained. At times less than $t_\xi$, the decrease with distance was described by a compressed exponent, as before. At long times, $t >> t_\xi$, the asymptotics are two-stage. The near part corresponds to the current regime of classical diffusion, and the distant part corresponds to the previous regime of super-diffusion. In this case, the behavior in the asymptotics corresponds to the general laws obtained for regularly inhomogeneous media.

Consider now anisotropic fractal media. In a general case, the anisotropy of geological media is due to the gravity or the pressure gradient. In the presence of anisotropy, a fractal medium behaves differently under the scale transformations along and across the anisotropy axis. Therefore, it is necessary to introduce an additional index $\beta$ for the coordinates in the basal plane, so that instead of $r \rightarrow \lambda r$, we consider the transformation $\{z \rightarrow \lambda z, \quad r_z \rightarrow \lambda^\beta r_z\}$. The study [25] showed that, depending on the parameter $\beta$, two cases are possible, namely, strong and weak anisotropy. When the
anisotropy is weak \((h < 1, \frac{1}{1+h} < \beta < \frac{2}{1+h})\), the transport goes in the super-diffusion regime in all directions, albeit at different rate:

\[
R_h(t) - (a^h Vt)^{\frac{1}{\beta}} \quad \text{and} \quad R_\perp(t) - (b^{(1+h)-1} Vt)^{\frac{1}{(1+h)}}
\]

(11)

In the case of strong anisotropy \((h < 1, \beta > \frac{2}{1+h})\), the transport in the longitudinal direction is still super-diffusion, and in the basal plane it is the classical diffusion:

\[
R_h(t) - (a^h Vt)^{\frac{1}{\beta}} \quad \text{and} \quad R_\perp(t) - \sqrt{D_\perp t}
\]

(12)

It is also important to note that in the presence of anisotropy, an anomalous drift takes place, when the average particle displacement increases in accordance with the superdiffusion law, so as is the size of the impurity cloud along the anisotropy axis \(\langle z \rangle \sim \sqrt{\langle z^2 \rangle} = R_{\parallel}(t)\).

In real percolation environments, traps are always contained. One can suggest at least two sources of traps. First, as already indicated, this is due to the very structure of the percolation cluster, which, along with the backbone, has also dead ends. Secondly, the percolation cluster itself is often immersed in a porous, although weakly permeable medium. In this case, the term describing the exchange of impurities between the backbone and the traps enters the transport equation for the concentration averaged over the ensemble of realizations, in a manner analogous to that in the Dykhne model.

This term has the shape

\[
Q(r,t) = \frac{\partial}{\partial t} \int_0^t \varphi(t-t') c(r,t') dt',
\]

(13)

where the properties of the integral kernel are determined on the basis of the properties of scale invariance of the medium [26, 27]. It follows that there is a large time interval, when the kernel has a power-law form \(\varphi(t) \sim \frac{1}{t^\alpha} \left(\frac{t}{t_0}\right)^\alpha\), where \(\alpha\) characterizes the decay of the kernel in the self-similar interval. At large times, \(t > t^*\), when the traps are saturated, the kernel decreases exponentially with time. As a result, the index determining the rate the impurity cloud growth is determined both by the properties of the velocity field and by the properties of the traps [28]:

\[
R(t) \approx (a^h Vt^*)^{\frac{1-\alpha}{\beta}} \propto t^{\frac{1-\alpha}{\beta}}.
\]

(14)

Depending on the relationship between the parameters (but under the condition \(h < 1\)), the transport can occur in both super- and sub-diffusion modes.

To check the theory's conclusions, a comparison was made between the results of the theory and experiment [29]. The experiment [29] was carried out in fractured crystalline rocks. Injection and pumping out of the tracer were produced in a converging weakly dipole configuration. As tracers were chosen heavy water, bromides, pentafluorobenzoic acid, the diffusion properties of which are very different. Concentration behavior at large times is the same for all tracers. Consequently, the diffusion exchange of particles between a weakly permeable matrix and a strongly permeable medium does not affect the impurity transport processes. Hence, to compare the conclusions of the theory with experiment, an interpolation formula was proposed that describes the behavior of the concentration at some distance from the source as a function of time

\[
c(r,t) = \left(\frac{t_0}{t}\right)^{\frac{3}{2h}} \exp \left( -B \left(\frac{t_0}{t}\right)^{\frac{1}{h}} \right).
\]

(15)
On the basis of this formula, the results of the theory were compared with experiment. This comparison is shown in Fig. 3.

![Comparison of the theory (formula (15)) with the results of tracer experiments [29].](image)

**Figure 3.** Comparison of the theory (formula (15)) with the results of tracer experiments [29].

4. **Statistically homogeneous sharply contrasting media**

An important (and perhaps most common) class of highly heterogeneous media are sharply contrasting, statistically homogeneous media. An example is fractured porous media when the fractures are uniformly distributed in a porous matrix. To describe transport in such media, there is a Gerke-Genuhent model [30], in which the impurity exchange between the fast and slow subsystems is considered in the "mean-field approximation". This implies that the exchange fluxes are determined by the difference in local average concentrations in blocks and fractures. However, it is not difficult to estimate that even for small blocks of the order of 10 cm, the time for concentration equalizing on the block sizes is quite large (tens and hundreds of years). And for an adequate description of transport during this time, it is necessary to take into account the remaining concentration gradients on the scales of individual blocks. For this, we developed a nonequilibrium model of double porosity [31, 32], in which the following results were obtained.

There is a large time interval during which the transport is described by anomalous regimes: along the average velocity, the transport occurs in the quasi-diffusion regime, when the average displacement \( \langle r \rangle \) and the size of the impurity cloud \( R \) grow in proportion to the root of time

\[
\langle r \rangle \approx R \approx \sqrt{D_u t}, \quad D_u = u^2 t_1,
\]

and in the transverse direction there is power sub-diffusion, described by the formula (5). In these formulas \( t_1 \) is the introduced above characteristic time of the beginning of the action of the traps, and \( u \) is the average velocity of groundwater filtration. At small and long times, the transport occurs in the classical advection-diffusion regime, but with different mean velocities and an effective dispersion coefficient.

5. **Sorption effects**

The advantage of our approach is that when describing non-classical regimes, it is possible to take into account in a natural way additional processes that affect the transfer, for example, sorption processes. It is known that sorption of impurities in the matrix during transport in porous media leads to a significant slowdown of the transport. For example, for long times, the average transport velocity and the effective dispersion coefficient decrease \( K \) times, where \( K = 1 + k_D \) is a so-called delay factor, and
the distribution coefficient $k_p$ relates the impurity concentration in the solution with the impurity concentration adsorbed in the matrix [33]. On the other hand, if colloidal particles are present in the medium (microscopic particles ranging in size from tens of nanometers to tens of microns, which may be present in groundwater, adsorb and carry impurities), this can lead to a significant acceleration of transport in double porosity media. The reason for this is that, because of the large dimensions of these particles compared to the pore sizes in a weakly permeable matrix, the process of their escape into the matrix is suppressed. As a result, they are freely transported with groundwater velocity through the fractures and carry the impurity adsorbed on them.

The account of a colloidal subsystem in describing impurity transport was considered for different types of double porous media [34-36]. An important parameter in the model is the distribution coefficient $\sigma$, which connects the impurity concentration in the solution with the concentration adsorbed on colloids under equilibrium conditions

$$c_{eq} = \sigma m_{eq}. \quad (17)$$

The calculation shows that the set of possible transport modes is preserved, but the effective transport parameters and the time interval boundaries change. The colloids exert the greatest influence at $\sigma \ll 1$. In Fig. 4 the results of calculations of the average displacement of impurity particles for systems with colloids (upper curve) and without them (the lower curve) for statistically homogeneous double-porous media are shown. It is evident that the presence of colloids greatly accelerates the transport process. Accounting for this process is especially important when assessing the rates of environmental pollution.

![Figure 4](image.png)

**Figure 4.** Average displacement of impurity for its migration in a statistically homogeneous double-porous medium in the presence of colloids. $t_s$ corresponds to the characteristic time of equalization of the impurity concentration at the scales of the porous blocks.

### 6. Conclusions

In this paper, we consider models of nonclassical transport in strongly heterogeneous media with a sharp contrast of properties and various types of correlations in the distribution of structural media characteristics. Three possible media types are analyzed: regularly heterogeneous, statistically homogeneous double-porous and percolation media. It is shown that in a general case the transport regime in the main body region of impurity localization depends on the time interval. This is due to the fact that the fraction of time the impurity particles spend in the channels with rapid transport and in
the traps depends on time interval. The exponential decrease in concentration at asymptotically large
distances takes place for all types of anomalous transport. The change in the transport regimes in the
main body region leads to a multi-step asymptotics, the more distant parts being determined by earlier
transfer regimes.

For regularly heterogeneous media consisting of a well conducting region bounded in one or
two directions and surrounded by a weakly permeable matrix, quasi-diffusion, power-law, and
logarithmic subdiffusion modes can be realized.

For percolation media, with a sufficiently slow decrease in the correlator of flow velocity
fluctuations, the transport occurs first in the super-diffusion regime, and then, in later times, the super-
diffusion regime is changed by the classical diffusion regime. The effective diffusivity is of the order
of the product of the average advection velocity and the correlation length of the medium.

The transport mode in anisotropic media with fractal properties depends on the degree of
anisotropy. For weak anisotropy, super-diffusion takes place in all directions. For strong anisotropy,
the classical diffusion regime is realized in the transverse direction. In addition, an anomalous drift
takes place in anisotropic media, so that the average displacement of particles increases in accordance
with the super-diffusion law.

For statistically homogeneous sharply contrasting media, there are long time intervals when the
transport along the flow velocity occurs in the quasi-diffusion mode (the average particle displacement
and the size of the impurity cloud grow with time according to the root law), and sub-diffusion takes
place in a plane perpendicular to the mean velocity.

Sorption on colloids in sharply contrasting media can lead to a significant acceleration of the
transfer process.

Acknowledgment
The authors acknowledge the support from the Russian Foundation for Basic Research (RFBR) under the Project 18-08-01229-a.

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