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Transport of large particles in flow through porous media

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There is considerable evidence indicating that significant reduction in the efficiency of many processes in porous media, such as enhancing oil recovery, heterogeneous chemical reactions, deep-bed filtration, gel permeation, and liquid chromatography, is due to the reduction in the permeability of the pore space. This reduction is due to the transport of particles, whose sizes are comparable with those of the pores, and the subsequent blocking of the pores by various mechanisms. In this paper we develop a novel Monte Carlo method for theoretical modeling of this phenomenon. Particles of various sizes are injected into the medium, and their migration in the flow field is modeled by a random walk whose transition probability is proportional to the local pore fluxes. Pores are blocked and their flow capacity is reduced (or vanished) when large particles pass through them (and reduce their flow capacity) or totally block them. The permeability of the medium can ultimately vanish and, therefore, this phenomenon is a percolation process. Various quantities of interest such as the variations of the permeability with process time and the distribution of pore-plugging times are computed. The critical exponent characterizing the vanishing of the permeability near the percolation threshold appears to be different from that of percolation conductivity. The agreement between our results and the available experimental data is excellent.

I. INTRODUCTION

Many operations in the chemical and petroleum industry involve transport and reaction processes in disordered porous media. Examples include multiphase fluid flow in oil and gas reservoirs, filtration processes, wood and coal combustion and gasification, heterogeneous (catalytic) chemical reactions, gel permeation, and liquid chromatography. The efficiency of such processes depends crucially on the availability of open pore space which provides transport paths for the flowing and reacting materials. Thus if the permeability of the porous medium, which is a measure of openness of the pore space, is reduced during such processes, the efficiency of the operation is also reduced, often dramatically. An important reason for the reduction of the permeability is the migration of fine, solid particles whose sizes are comparable with those of the pores of the medium, in the flow field through the porous medium. These particles can be the product of a chemical reaction within the pore space or the result of a process such as etching of the porous medium, which is intended to change the structure of the pore space. Moreover, particles adhering to the pore walls, e.g., diagenetic clays which have been precipitated from formation water, can be released upon contact with the flowing fluids because of the change in the ionic environment or the flow rate. In addition, during the flow of dilute, stable emulsions in fine-grained porous media,[1] a large reduction in the permeability of the porous media is usually observed. This is because the ratio of emulsion size to pore size may be of order of unity and, as a result, the pores of the medium are blocked by the emulsions, a phenomenon which is similar to the reduction of the permeability during the migration of fine particles in flow through a porous medium. Thus size exclusion, by which large particles block smaller pores, is the dominant mechanism of the reduction of permeability of the porous medium. Capture of the particles by the solid surface of the pore by, e.g., an electrostatic interaction between migrating particles and the surface, is another mechanism which results in the reduction of the permeability. In addition, during gel permeation in porous media and liquid chromatography,[2,3] size exclusion plays a fundamental role. In any event, the permeability of the pore space reduces with process time and can ultimately vanish if enough pores are blocked. As such, this phenomenon is a percolation process,[4] although, as we argue below, it may not be an ordinary percolation process and appears to be described by a different set of critical exponents.

Past modeling of this phenomenon has involved[5–8] various geometrical models of the pore space, such as a bundle of parallel capillary tubes with given pore-size and particle-size distributions, or the Carman–Kozeny model for permeability of a porous medium. More recently, Soo, Williams, and Radke[9] presented a filtration model to describe the permeability reduction due to the capture of droplets by the solid surface of the pores. Although these models contain many essential features of transport of fine particles in flow through porous media, such as their deposition on the solid surface of the pores and their possible release from the surface, their predictions of the reduction in the permeability of the porous media are much smaller than the experimental data.[1,10,11] This is because of the fact that models of the pore space that have been employed by these authors cannot represent the effect of the topology (connectedness) of the porous media and the interconnection between the pores. It is well known that such models predict a zero-percolation threshold for the porous medium and, therefore, it is not surprising that they predict a permeability reduction which is much smaller than the observed one. A recent advancement is the work of Sharma and Yortsos,[12] in which the authors employ a network model of pore space to describe the transport of fine particles in a flow field. However, an effective-medium approximation (EMA) is used in order to describe the flow field and make the model analytically tractable. Although the results are in better agreement with the experimental data than those of previous models, their model is not totally satisfactory, since an EMA cannot predict accurately the permeability of the porous medium as the percolation threshold is approached.

The goal of this paper is to develop a Monte Carlo method for studying the transport of fine particles in flow through a porous medium and its effect on permeability. Our method is closely related to the Monte Carlo method that we developed[13] earlier to study hydrodynamic dispersion in porous media, although there are also significant differences between the two methods because the two problems are totally unrelated. We consider only the reduction of the permeability of the porous medium due to the size-exclusion mechanism and ignore any other factor that can affect the permeability of the pore space. In a future paper,[14] we will investigate the effect of other mechanisms of permeability reduction. However, we show here that there is excellent qualitative agreement between our results and the experimental data. This indicates that the dominant mechanism of the reduction of the permeability of a porous medium may be the size-exclusion phenomenon, as mentioned above.

The plan of this paper is as follows. In Sec. II we give the details of our Monte Carlo method. Section III contains the results and the discussion of our Monte Carlo simulations of the problem. The paper is summarized in Sec. IV where we also discuss how an analytical model may be developed to predict the Monte Carlo results.

II. MONTE CARLO SIMULATIONS METHOD

The porous medium is represented by a two-dimensional square network. The bonds of the network, which represent the pore throats of the porous medium, are assumed to be cylindrical capillary tubes which have no converging or diverging section. The radii R of the tubes are distributed according to a statistical distribution, which, in the present paper, is assumed to be the Rayleigh distribution

$$f(R)=2\alpha^2 R\exp(-\alpha^2 R^2)\ , \tag{1}$$

where α^{-1} is a characteristic pore radius. This distribution mimics qualitatively the pore-size distributions determined experimentally by several investigators.[15,16] The sites of the network, which represent the pore bodies of the porous medium, are assumed to be large enough in order to be able to contain the fine particles, although the effect of their sizes on the flow field is ignored. Creeping flow within each bond is assumed, and the pressure distribution in the network is computed. This is done by solving a standard Kirchhoff-law formulation in which the flow rate in each bond is the product of the pressure difference between its nodes and its hydrodynamic conductance, which is proportional to R^4. The boundary conditions are constant pressures imposed at the entrance and exit plane of the network ($X=0$ and $X=L$, respectively), and matched periodic conditions in the Y direction. From the pressure distribution and the bond radii, the average flow velocity and the flow rate in each bond is calculated and the permeability of the entire network is determined.

After the flow field is determined, solid particles are injected into the network at random at plane $X=0$. This can be done in several ways. For example, one can inject a single particle into the network at time $t=0$, the second particle at time Δt, and the Nth particle at time $(N-1)\Delta t$. Alternatively, we can inject simultaneously a set of M particles into the network at time $t=0$, the second set at time Δt, and so on, where M can vary from set to set. Finally, we may choose the time intervals between two consecutive injections from a probability density function. The results presented in this paper are with the second method. After each time interval, we inject M particles into the network. We take M to be a random variable uniformly distributed in the interval $(0,L)$. This is because of the fact that at any given time a different number of solid particles may enter the porous medium.

The particles are assumed to be effectively spherical (circular in two dimensions) the effective radii of which are distributed according to a particle-size distribution which, in the present paper, is either a uniform distribution in the interval (0,1) or the Rayleigh distribution. Within each pore, a particle moves with the mean flow velocity of the pore. Because the size of a particle is finite, its motion can disturb the flow pattern within the pore. However, we neglect such disturbances because taking them into account would make the computations extremely complicated. We also neglect the possible capture of the particles by the solid surface of the pore; these issues will be considered in a future paper.[14]

When a particle arrives at a node, it leaves into one of the attached bonds which is open to flow (i.e., it has not been plugged yet). The transition probability for going from one pore into another is assumed to be proportional to the fraction of the flow rate departing from the node through that node. After a particle selects one of the available pores, its effective radius r_p is compared with that of the pore R. If $r_p<R$, the particle is allowed to move into that pore and to be carried with the flow. However, if $r_p\geq R$, the entrance to the pore is blocked by the solid particle and the pore is completely plugged. This is effectively equivalent to removing the pore from the network (i.e., setting its radius to be zero) and, therefore, the phenomenon is a percolation process. This procedure is then repeated for all the particles in the network. Once the plugged pores are identified, a new flow field is computed, taking into account the presence of all solid particles in the system and their effect on the flow

field, and the permeability of the network is computed. Particle injection is continued until there is no longer any significant change in the network permeability, or until the network is completely plugged and its permeability vanishes. Alternatively, one can stop the injection of the particles after a certain amount of them have been injected into the system, and study the structure of the resulting network. The extent of the pore plugging and the decrease in the permeability of the network depend crucially on the size distribution for the pores and particles and the method of injection of the particles into the network. These factors also influence the time at which the network becomes macroscopically plugged and its permeability vanishes; these are discussed below. This Monte Carlo method is very similar to that used for the study of hydrodynamic dispersion,[13] although the two problems are totally different. We used networks of sizes $L=40$ and 60, and we typically averaged the results over ten different realizations of the network.

III. RESULTS AND DISCUSSION

We have carried out extensive Monte Carlo simulations of the process described above. We have used two different values of Δt in our simulations, $\Delta t=1000$ and 5000. The units of time depend on the units of volumetric flow rate in the network. Changing Δt enables one to change the effective concentration or volume fraction of the solid particles in the pore space. Small values of Δt mean a large concentration of the solid particles (since for a given process time, more particles have entered the pore space), and *vice versa*. Figure 1 shows the variations of the permeability of the network with the process time. The permeability of the network is normalized with its value when no pore has plugged yet (i.e., its permeability at $t=0$). The results are for the Rayleigh distribution for the pore sizes with $\alpha=1$, and a uniform particle size distribution. As can be seen, the permeability drops sharply after a relatively short time, and the decrease is sharper for the smaller value of Δt. After some time, the permeability reaches its steady-state value and changes very little with the process time. This means that with the pore-size and particle-size distributions that have been used, almost no pore can be plugged after time $t \simeq 2\times 10^4$, because there is not a large enough particle to plug a relatively large pore. In order to be able to compare directly the results with the experimental data,[1,10,11] we present in Fig. 2 the variations of the permeability of the network with the pore volume injected τ. Here τ is defined by $\tau=Qt/V$, where Q is the total volumetric flow rate in the network at time t and V the total volume of the unplugged network at $t=0$. The results are in qualitative agreement with the experimental data.[1,10,11] although our results show a steeper decrease in the permeability of the network than those of the experimental data.

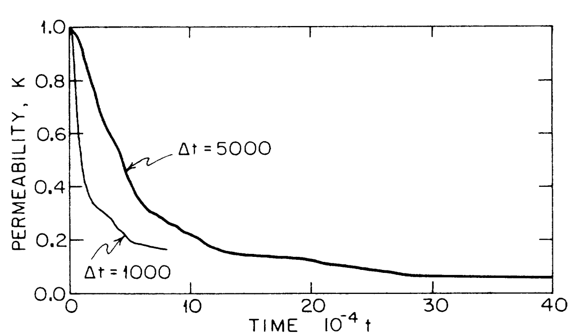

FIG. 1. Variation of the permeability of the network with process time for various values of Δt.

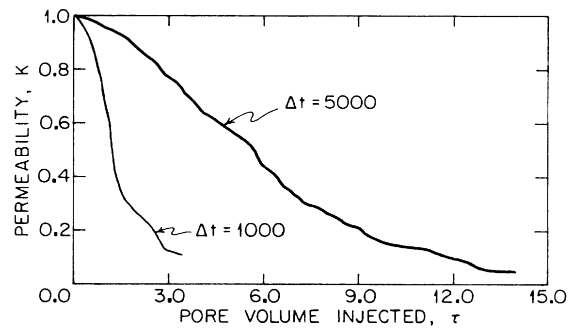

FIG. 2. Dependence of the permeability of the network on the pore volume injected, for various values of Δt.

In order to interpret the results in terms of the concepts of percolation theory, the variations of the permeability of the network with the fraction q of the unplugged pores at time t are shown in Fig. 3. Two points are worth noting. The shapes of these curves are totally different from those of percolation conductivity (the analog of permeability) which have been obtained for the square network; see, e.g., Kirkpatrick.[17] Whereas percolation conductivity decreases essentially linearly with the fraction of conducting bond (i.e., the analog of unplugged pores in our study), except very close to the percolation threshold $p_c=\frac{1}{2}$, the permeability of the network in our study decreases more slowly with q, and the decrease in the permeability is not even linear. Moreover, the percolation threshold q_c of our network (i.e., the point at which the permeability effectively vanishes) appears to be higher than that of the percolation conductivity. This is because in our simulations the bonds of the network are not blocked completely at random. Those pores that are in the direction of the average (macroscopic) flow have larger fluid fluxes and, therefore, are more likely to be selected by the solid particles

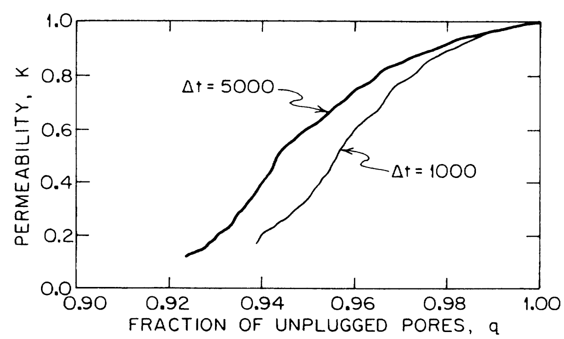

FIG. 3. Permeability of the network as a function of the fraction of unplugged pores for various values of Δt.

and be plugged. Moreover, the blockage of a pore depends on its effective radius and the effective size of the particle that is encountering it. Thus we may expect a higher percolation threshold than that of the percolation conductivity, consistent with our results. The shape of the permeability curve might also indicate that near q_c, one has

$$K \simeq (q - q_c)^{\mu} \,, \tag{2}$$

where μ is presumably a universal exponent. We expect μ to be different from that of percolation conductivity, whose best current estimate is about 1.3,[18] and in fact we believe $\mu < 1$; this is discussed below. Our results do, however, indicate that the phenomenon of pore plugging is sensitive to the morphology of the pore space (i.e., its geometry, represented by the pore-size distribution, and topology), the particle-size distribution, and the concentration of the solid particles.

Figure 4 represents the distributions of the distances, along the direction of macroscopic average velocity that the solid particles travel before they cause any plugging. Although the two distributions for the two values of Δt are qualitatively similar, the distance that is traveled by the particles is larger for larger values of Δt. This is in agreement with experimental observations[1,10,11] that for less concentrated systems (i.e., larger values of Δt in our model) the particles travel a larger distance before they cause any pore plugging. It is also intuitively clear that, if the system is highly concentrated, more pores are plugged and, therefore, it is more difficult for the particle to travel before any pore plugging takes place.

To investigate the effect of mean-flow orientation on the pore-plugging process, we applied a pressure difference at 45° to the two pore directions and thereby produced flow in the diagonal direction. In this case, by virtue of the pore orientations, two bonds at each node carry fluxes into the node and the two other carry fluxes out, i.e., they are directed away. Thus we obtain a fully directed square network in the language of percolation theory, though the full directedness of the network is not intrinsic but is flow induced and dynamical. Moreover, no pore in a particle direction is more likely to be plugged because, on the average, all pores that are directed away from a node carry the same flux. Therefore, we may expect the percolation threshold of the oriented network at which the permeability vanishes to be lower than that of the usual square network discussed below.

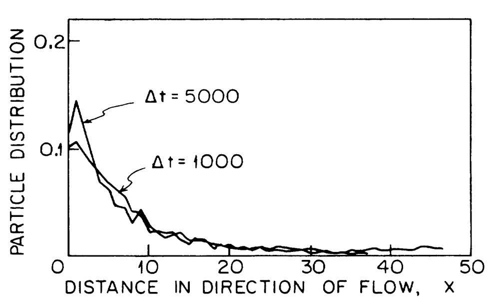

FIG. 4. Distribution of the distances, along the direction of macroscopic mean velocity, that the particles travel before they cause any pore plugging.

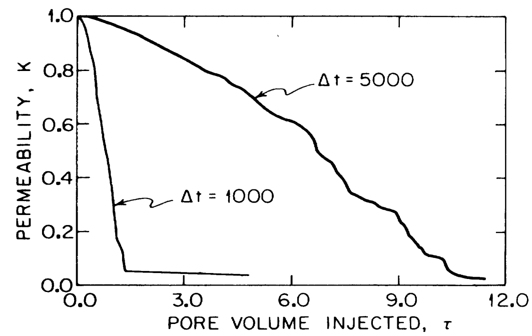

FIG. 5. Reduction of the permeability of the oriented network with the injected pore volume for various values of Δt.

Figure 5 represents the reduction of the permeability of the oriented network with the injected pore volume. This figure shows that, at least for larger values of Δt, the permeability of the oriented network decreases more slowly than that of the unoriented one. This may be expected on intuitive grounds because the plugging of the pores of the oriented network is more random than those of the unoriented network. As a result, the permeability of the network should decrease more slowly as more pores are plugged. This figure is in excellent agreement with the experimental data,[1,10,11] and it may indicate that the dominant mechanism of pore plugging during the transport of fine particles, in flow through a porous medium is the size-exclusion mechanism, and that other possible mechanisms, such as the capture of fine particles by the surface of the pores, may be of secondary importance. Of course, capture of a solid particle by the surface of the pores depends on several factors. It depends primarily on the fluid flux within a pore. If the flux is large, the particle is quickly transported to the end of the pore, and it is not very likely that the particle and the pore walls can interact with one another and, thus, the particle may not be captured by the surface of the pores. Capture of a particle depends also on the ratio r of particle size to pore size. If this ratio is close to unity, it is more likely that the particle would be captured by the surface of the pores. More-

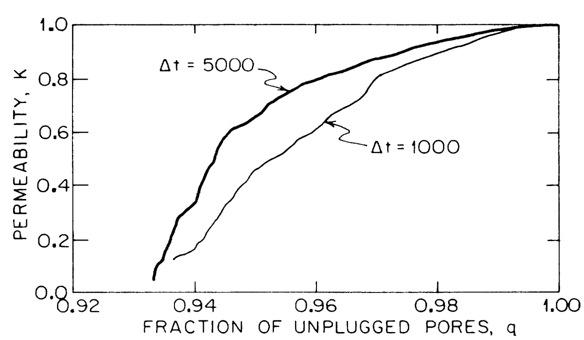

FIG. 6. Permeability of the oriented network as a function of the fraction of unplugged pores.

over, if r is close to unity, the transport of fine particles can have a strong effect on the flow field within the pore, which can affect the capture of the particles. Another factor that influences the capture of the particles by the pore walls is the strength of the interaction between the particle and the pore walls. These matters will be discussed in a future paper.[14]

Figure 6 represents the variations of the permeability of the network with the fraction q of unplugged pores. A comparison between Fig. 6 and Fig. 3 shows again that the permeability of the oriented network decreases more slowly with q than that of the unoriented network. The shape of the curves in Fig. 6 shows a more dramatic departure from that of percolation conductivity.[17] It also indicates that $\mu < 1$, in contrast to the exponent of percolation conductivity (or permeability, since the two exponents must be the same in two dimensions), which is always greater than unity.

Other important quantities are the distribution and the average of times that the particles spend in the porous medium before they cause any plugging. Knowledge of these quantities may help one to predict the time at which the porous medium becomes macroscopically disconnected and its permeability vanishes. In Fig. 7, we present the distribution of plugging times, i.e., the times at which a given particle, after entering the porous medium, plugs a pore. As can be seen, this distribution is long tailed and, while most particles plug a pore after a relatively short time, some of the pores are plugged after a long time. Because the particle-size distribution is uniform in the interval (0,1), while the pore size distribution is given by Eq. (1) with $\alpha = 1$, pores whose effective sizes are close to unity are very difficult to plug and their plugging times are very large.

Finally, to investigate the effect of pore-size and particle-size distributions on the results, we calculated the reduction in the permeability of the network if both particle and pore sizes are distributed according to Eq. (1), with $\alpha = 1$ for the pore-size distribution and $\alpha = \frac{3}{2}$ for the particle-size distribution. The variations of the permeability with the process time are shown in Fig. 8. As can be seen, the permeability decreases very sharply and vanishes at a relatively short time. The shape of the curve is also somewhat different from those discussed above. This indicates that the permeability reduction is sensitive to the pore-size and particle-size distributions, as discussed above.

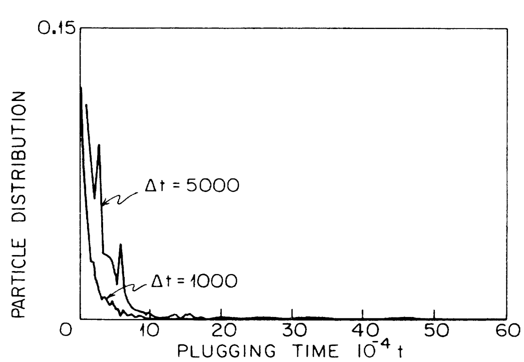

FIG. 7. Distribution of pore-plugging times for $\Delta t = 10^3$.

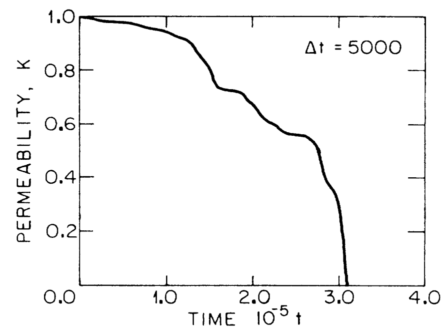

FIG. 8. Time dependence of the permeability of the network if particle- and pore-size distributions are both given by the Rayleigh distribution [Eq. (1)].

IV. SUMMARY AND DISCUSSION

We have developed a novel Monte-Carlo-simulation approach to the problem of reduction of permeability of a porous medium due to the plugging of its pores by large solid particles. The results are in excellent qualitative agreement with the available experimental data.[1,10,11] Since in our simulations we considered the plugging of the pores only by the size-exclusion mechanism, the agreement between our results and the experimental data may indicate that during the migration of fine-particles in flow through a porous medium, size exclusion is the dominant mechanism of pore plugging.

Sharma and Yortsos[12] have developed an EMA to estimate the permeability K of the network as a function of the fraction of the unplugged pores q. For a square network, their equation for K becomes

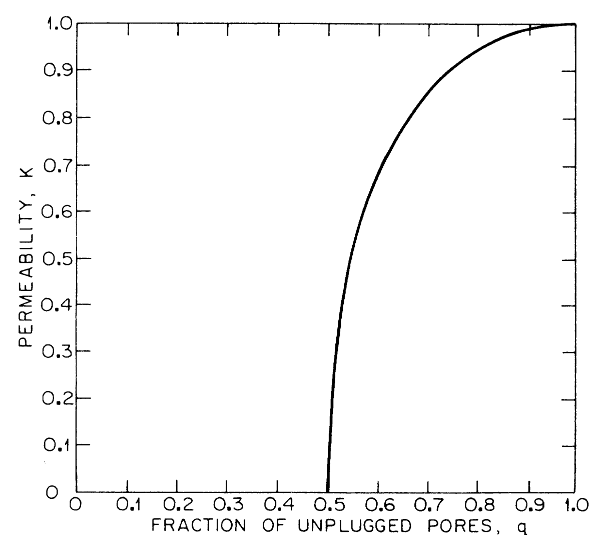

FIG. 9. Permeability of the network as predicted by the effective-medium approximation.

$$\int_A^\infty f(R)\frac{K-R^4}{K+R^4}dR = q-1 , \tag{3}$$

where $A = R^*/r_p^*$, and R^* and r_p^* are the mean pore size and particle size, respectively. The predictions of (3) are shown in Fig. 9. They are qualitatively similar to the Monte Carlo results, but the EMA predictions of the permeability reduction are much steeper than those predicted by our Monte Carlo simulations. Asymptotically, an EMA predicts that $K \simeq (q-q_c)^\mu$, with $\mu = \frac{1}{2}$. Although this value of μ may not be exact, it does indicate that the value of permeability exponent in the present problem is different from that of percolation conductivity.

The close agreement between our results and the experimental data may indicate that, at least to a high degree of accuracy, the concentration C of the particles may obey a convective-diffusion equation (CDE)

$$\frac{\partial C}{\partial t} + V_a \nabla C = D_L \frac{\partial^2 C}{\partial x^2} + D_T \nabla_2^2 C ,$$

where V_a is the mean velocity of the flow, D_L and D_T are some effective diffusion coefficients measuring the spread of the particles in the longitudinal (macroscopic mean flow) and transverse (perpendicular to the macroscopic mean flow) directions, respectively, and ∇_2^2 is the Laplacian in transverse direction. This is because of the fact that the random walk of a particle in a flow field can be described[13] by a CDE. We will investigate this in a future paper.[14]

In a future paper,[14] we will consider other mechanisms of permeability reduction during transport of fine particles in flow through porous media. We will also attempt to estimate the exponent μ in order to establish the universality class of the permeability of the network in the present problem.

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