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An efficient particle tracking equation with specified spatial step for the solution of the diffusion equation

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Abstract

The traditional diffusive particle tracking equation provides an updated particle location as a function of its previous location and molecular diffusion coefficient while maintaining a constant time step. A smaller time step yields an increasingly accurate, yet more computationally demanding solution. Selection of this time step becomes an important consideration and, depending on the complexity of the problem, a single optimum value may not exist. The characteristics of the system under consideration may be such that a constant time step may yield solutions with insufficient accuracy in some portions of the domain and excess computation time for others. In this work, new particle tracking equations specifically designed for the solution of problems associated with diffusion processes in one, two, and three dimensions are presented. Instead of a constant time step, the proposed equations employ a constant spatial step. Using a traditional particle tracking algorithm, the travel time necessary for a particle with a diffusion coefficient inversely proportional to its diameter to achieve a pre-specified distance was determined. Because the size of a particle affects how it diffuses in a quiescent fluid, it is expected that two differently sized particles would require different travel times to move a given distance. The probability densities of travel times for plumes of monodisperse particles were consistently found to be log-normal in shape. The parameters describing this log-normal distribution, i.e., mean and standard deviation, are functions of the distance specified for travel and the diffusion coefficient of the particles. Thus, a random number selected from this distribution approximates the time required for a given particle to travel a specified distance. The new equations are straightforward and may be easily incorporated into appropriate particle tracking algorithms. In addition, the new particle tracking equations are as accurate and often more computationally efficient than the traditional particle tracking equation.

Keywords: Particle tracking; Diffusive transport; Stochastic processes; Constant spatial step

1. Introduction

With the ever expanding capabilities of computers, particle tracking solutions to various engineering problems are becoming increasingly powerful. Although random walk methods, Monte Carlo simulations, and Fokker–Planck solutions to differential equations have been employed for many years, the availability of inexpensive high speed processors and vast memory stores has allowed the application of these solution techniques to increasingly complex problems (e.g., Uffink, 1988; Valocchi & Quinodoz, 1989; Yamashita & Kimura, 1990; Lu, 2000; Liu, Bodvarrson, & Pan, 2000; Michalak & Kitanidis, 2000; Tsang & Tsang, 2001). For example, James and Chrysikopoulos (1999, 2000) have investigated particle tracking schemes that model the transport of variably sized colloids in both uniform and variable aperture fractures. James and Chrysikopoulos (2001a) have also compared a traditional particle tracking algorithm with analytical solutions for the ideal case of polydisperse colloid transport in a uniform aperture fracture and excellent agreement was shown. Also, Grindrod and Lee (1997) used particle tracking to model the transport of reactive particles in a single, symmetric fracture with a sinusoidally varying aperture. However, for more realistic and involved models accounting for a fracture with a random variable aperture, a distribution of particle sizes, or particle sorption onto the fracture walls, a traditional particle tracking algorithm may not be the most efficient

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solution method (Reimus, 1995). There are often cases when a particle tracking algorithm using a constant time step may lead to both insufficient prediction accuracy and excessive computation time.

Common solution methods to colloid transport problems include analytical solutions, finite element methods, finite difference methods, and traditional particle tracking algorithms; however, each technique has its short comings. Analytical solutions require extensive simplifications to yield tractable problems (Abdel-Salam & Chrysikopoulos, 1994). Finite difference and finite element techniques suffer from numerical dispersion and cannot account for finitely sized particles (Abdel-Salam & Chrysikopoulos, 1995a, b). Although particle tracking methods can account for finitely sized particles, an important case where the traditional particle tracking equation with a constant time step may be insufficient arises in the study of polydisperse colloid transport. Consider the simple example of random diffusion of a plume of polydisperse colloids in a quiescent fluid. Particle tracking theory suggests that the time step should be chosen small enough to represent the time that a particle might take to travel along a certain path before it is forced to significantly deviate from its course through a molecular exchange of kinetic energy (Uffink, 1988). If a constant time step that is appropriate for the median colloid size is applied to all particles in the plume, this constant time step, when applied to colloid particles at each extreme of the size distribution, yields undesirable results. Colloid diameters and corresponding molecular diffusion coefficients can span several orders of magnitude, and as a result, the smallest colloids may travel diffusively too far during this pre-determined time step to meet the desired accuracy. The largest colloids may require an excessive number of time steps to reach the desired time of solution resulting in increased computational cost.

Another case when the traditional particle tracking algorithm may be insufficient is when the transport of colloids is significantly affected by deposition onto formation surfaces. As a colloid travels through a fracture, diffusion across streamlines eventually brings the particle close enough to a fracture wall to have the opportunity to establish a contact at the liquid–solid matrix interface. If a time step was specified, rarely would the particle exactly encounter the sorption site on the fracture wall. Instead, by determining a random diffusive travel time for the particle to reach the fracture wall a known distance away, knowledge of exactly where and when a particle encounters a sorption site is obtained. In either of the above mentioned cases, a particle tracking equation with a pre-determined spatial step yielding a random travel time would achieve the desired predictive accuracy while maximizing computational efficiency.

The new particle tracking equation derived in this work maximizes computational efficiency and solution accuracy by specifying a priori a spatial step and determining the random time a spherical particle of neutral buoyancy will take to diffusively travel a specified distance. There may be no advective component in the direction in which diffusion is of interest (transport by Poiseuille and Couette flows are prime examples). This work formally presents the methodology used to obtain accurate coefficients for the new particle tracking equation in one dimension and extends this same procedure to obtain equations in two and three dimensions. Results based on the new particle tracking algorithm are validated through comparison with both an analytical solution and results from the traditional particle tracking equation. Furthermore, it is shown that the particle tracking equation derived here is more accurate than the one suggested by Reimus (1995).

2. Model development

2.1. Traditional particle tracking equation

The traditional particle tracking transport equation for the solution of advection–diffusion problems consists of a deterministic (or absolute advective) term, and a stochastic (or diffusive) term that is a function of the random motion of the particle (Thompson, 1993; Kitanidis, 1994). For the case of particle diffusion in the absence of advection considered in this work, the advective term is eliminated and in vector notation the traditional diffusive particle tracking equation is given by

$$\mathbf{X}^m = \mathbf{X}^{m-1} + \mathbf{B}(\mathbf{X}^{m-1}) \cdot \mathbf{Z}(0,1)\sqrt{\Delta t}, \tag{1}$$

where exponent m is the numerical step number; $\mathbf{X}^m = (x^m, y^m, z^m)^{\mathrm{T}}$ is a three-dimensional position vector with x^m, y^m, and z^m representing the Cartesian coordinates of the centroid location of a particle at the numerical step m; $\mathbf{B}$ $(\mathbf{X}^{m-1})$ is a deterministic scaling second order tensor, evaluated at $\mathbf{X}^{m-1}$, that is a function of the spreading of the particle plume; and $\mathbf{Z}(0,1)$ is a vector of three independent random numbers selected from the standard normal distribution. When only molecular diffusion is considered, the terms of the diagonal second order tensor $\mathbf{B}$ $(\mathbf{X}^{m-1})$ are equal to $\sqrt{2\mathscr{D}}$ (Ahlstrom, Foote, Arnett, Cole, & Serne, 1977), and the molecular diffusion coefficient, $\mathscr{D}$, is given by the Stokes–Einstein equation as

$$\mathscr{D} = \frac{kT}{3\pi\eta d_p}, \tag{2}$$

where k is Boltzmann's constant, d_p is the particle diameter, η is the dynamic viscosity, and T the absolute temperature of the suspending fluid. The diffusive particle tracking vector equation (1) may be represented by the following directional particle tracking equations:

$$x^m = x^{m-1} + Z(0,1)\sqrt{2\mathscr{D}\Delta t}, \tag{3a}$$

$$y^m = y^{m-1} + Z(0,1)\sqrt{2\mathscr{D}\Delta t}, \tag{3b}$$

$$z^m = z^{m-1} + Z(0,1)\sqrt{2\mathscr{D}\Delta t}. \tag{3c}$$

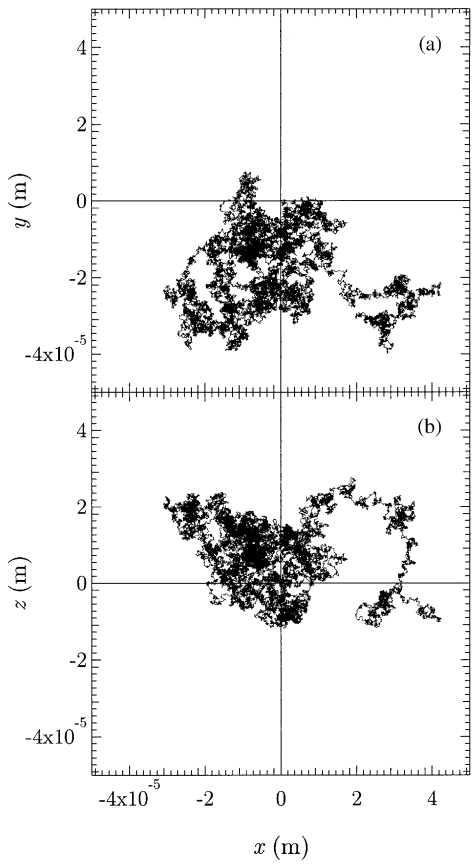

Fig. 1. Pathline projected at (a) the x, y-plane; and (b) the x, z-plane of the Brownian motion for a particle of size $d_p = 1 \times 10^{-6}$ m released at the origin of a spherical volume of water with radius 5×10^{-5} m.

Traditionally, a time step is specified and an updated position vector is marched through time until the desired solution can be examined. In the limit of $\Delta t \to 0$, the particle tracking equation (1) becomes an exact solution to the diffusion equation (Kinzelbach & Uffink, 1988; Thompson & Gelhar, 1990). However, the cost of improved accuracy is increased processor time that is inversely proportional to decreasing Δt. When choosing an appropriate time step, both the accuracy of the solution and computational cost should be considered.

A particle suspended in a quiescent volume of fluid undergoes molecular diffusion in all three dimensions. Fig. 1 shows the (a) x, y-plane and (b) x, z-plane Brownian motion pathlines for a particle with diameter $d_p = 1 \times 10^{-6}$ m that was released at the origin and allowed to diffuse in water at $T = 288.15$ K according to Eqs. (3a), (3b) and (3c) with a time step of $\Delta t = 0.1$ s. The time required for this particle to exit a spherical volume of water with radius 5×10^{-5} m was $\sum \Delta t = 3860$ s.

Tory (2000) has suggested that the times required for many particles to diffusively travel a given distance are log-normally distributed. One might suggest that an intuitive estimation of the time required for a particle to travel a specified distance, $\Delta z = z^m - z^{m-1}$, could be obtained by rearranging the traditional particle tracking equation (3c) to solve for Δt as follows:

$$\Delta t = \frac{(\Delta z)^2}{2\mathscr{D}} \frac{1}{[Z(0,1)]^2}. \tag{4}$$

The preceding equation indicates that Δt is inversely proportional to the square of a standard normally distributed random number. Consequently, any time in the range of 0–∞ is feasible for Δt, with large numbers encountered more often. However, it should be noted that the distribution of numerous times calculated from Eq. (4) is not log-normal because taking the inverse square of standard normal deviates does not yield a log-normal probability density function. In fact, because the probability density is highest at $Z(0,1) = 0$, time steps resulting from Eq. (4) have a high probability of being very large. Computationally, this equates to a non-converging mean and standard deviation for the probability distribution of Eq. (4) as infinite travel times are returned.

2.2. New particle tracking equation

The goal of this work is to generate new particle tracking equations that express a diffusive travel time as a random function of a particle's diffusion coefficient as well as the distance traveled. A detailed description of the model used to generate one-dimensional results follows. Because diffusion is an isotropic process, the diffusive particle tracking equation (1) may be studied by any of the three directional particle tracking equations (3a), (3b) and (3c). In this analysis, Eq. (3c) will be used, but similar results may also be obtained from Eq. (3a) or (3b). The traditional particle tracking equation with an extremely small time step is used to generate histograms of times necessary for plumes of uniquely sized monodisperse particles to travel a preselected distance $|\Delta z|$ with the intent of determining the relationship between the characteristics of the histograms and the parameters, Δz and $\mathscr{D}$.

The one-dimensional particle tracking equation (3a) is employed to simulate the diffusion of a particle plume in the z-direction initially released in water at $T = 288.15$ K and at $z = 0$ (the origin). Each particle is allowed to randomly diffuse until it moves a distance of $\Delta z = \pm 5 \times 10^{-5}$ m from the origin. The selected travel distance is representative of a typical fracture aperture in a fractured rock formation. The numerical time step is chosen to be $\Delta t = 1$ s, so that a large number of time steps (~ 3000 for a particle with diameter $d_p = 1 \times 10^{-6}$ m) will be required for any particle to travel the specified distance, even for the extreme case when the magnitudes of all randomly generated numbers are > 1. The travel time, $\sum \Delta t$, required for a particle to achieve the

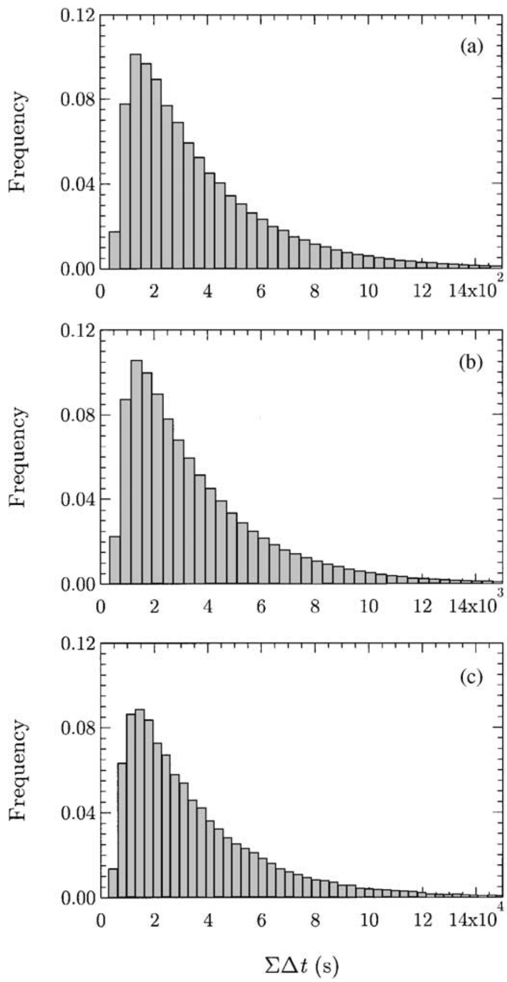

Fig. 2. Histograms of travel times for monodisperse plumes of 500,000 particles of size (a) $d_p = 1 \times 10^{-7}$ m; (b) $d_p = 1 \times 10^{-6}$ m; and (c) $d_p = 1 \times 10^{-5}$ m, traveling a distance of $\Delta z = \pm\, 5 \times 10^{-5}$ m.

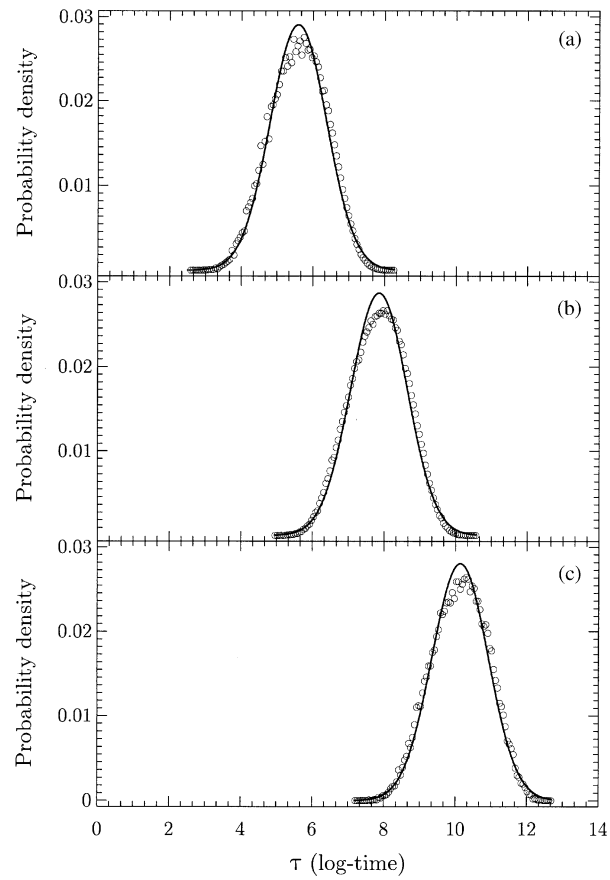

Fig. 3. Probability densities of log-travel times for the same monodisperse plumes used to generate Fig. 2. The solid lines are obtained from the normal pdf with parameters (a) $\mu_\tau = 5.588$ and $\sigma_\tau = 0.786$; (b) $\mu_\tau = 7.848$ and $\sigma_\tau = 0.787$; and (c) $\mu_\tau = 10.133$ and $\sigma_\tau = 0.787$.

specified distance, $|\Delta z| = 5 \times 10^{-5}$ m, is recorded. This particle diffusion process is repeated 500,000 times. Each stochastic trajectory mimics the actual path of an individual particle. Collectively, the trajectories illustrate the overall behavior of a 500,000 particle plume. It should be noted that any possible particle–particle interactions are not accounted for in the present analysis. One hundred unique particle travel time histograms are prepared for particle diameters ranging from 1×10^{-7} to 1×10^{-5} m in increments of 1×10^{-7} m. In view of Eq. (2), with $k = 1.380658 \times 10^{-23}$ kg m^2/s^2 K, $T = 288.15$ K, and $\eta = 1.003 \times 10^{-3}$ kg/m s, the corresponding range of $\mathscr{D}$ is from 4.21×10^{-14} to 4.21×10^{-12} m^2/s. Fig. 2 presents 3 of the 100 travel time histograms generated by this process. Fig. 2a is the histogram generated for the smallest particles considered in this study ($d_p = 1 \times 10^{-7}$ m); Fig. 2b represents the median particle size ($d_p = 1 \times 10^{-6}$ m); and Fig. 2c the largest particles ($d_p = 1 \times 10^{-5}$ m). All histograms are normalized by the number of particles in the plume. Note that the travel times increase proportionally to the increase in particle diameter (decreasing molecular diffusion coefficient).

Taking the log of the travel time for each particle of the plume, $\tau = \ln \sum \Delta t$, and generating the corresponding histogram, it is observed that the normal probability density function (pdf) can consistently fit the distribution of log-travel times obtained by the traditional particle tracking equation (3c). The normal pdf in terms of log-travel times is given by (Banks, Carson II, & Nelson, 1996, p. 209),

$$f(\tau) = \frac{1}{\sigma_\tau \sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\tau - \mu_\tau}{\sigma_\tau}\right)^2\right], \tag{5}$$

where μ_τ and σ_τ are the mean and standard deviation of log-travel times, respectively.

Fig. 3 presents the three probability densities of log-travel times corresponding to the same plumes of 500,000 monodisperse particles used to construct Fig. 2 and supports the suggestion by Tory (2000) that diffusive travel times are log-normally distributed. The solid lines on Fig. 3 represent the normal pdf's generated by Eq. (5). The parameters in Eq. (5), μ_τ and σ_τ, are the calculated arithmetic mean and standard deviation, respectively, of the log-travel times for all particles of a plume. Note that in Fig. 3a, $\mu_\tau = 5.588$ and $\sigma_\tau = 0.786$; in Fig. 3b $\mu_\tau = 7.848$ and $\sigma_\tau = 0.787$; and in Fig. 3c $\mu_\tau = 10.133$ and $\sigma_\tau = 0.787$.

Any normally distributed random number, $Z(\mu_\tau, \sigma_\tau^2)$, can be generated from the standard normal distribution, $Z(0,1)$, by employing the following relationship (Banks et al., 1996):

$$Z(\mu_\tau, \sigma_\tau^2) = \mu_\tau + \sigma_\tau Z(0,1). \tag{6}$$

Because it is evident from Fig. 3 that a normal pdf closely approximates the log-travel times for a plume of particles, a random number generated by Eq. (6) can be used to determine the log-travel time necessary for a particle with known diffusion coefficient to travel a specified distance. Assuming that the mean, μ_τ, and standard deviation, σ_τ, of log-travel times may be expressed as functions of the variables, Δz and $\mathscr{D}$, then taking the inverse log of a single normally distributed number generated from $Z(\mu_\tau, \sigma_\tau^2)$ is equivalent to selecting a particle travel time from the histogram of such travel times (see Fig. 2).

As seen in Fig. 3, the mean log-travel time varies with the particle diameter (i.e., molecular diffusion coefficient) used in each numerical simulation. For the units of mean log-travel time to be log-seconds, the relationship for the mean of the log-travel times must be linear with respect to $\ln[(\Delta z)^2/\mathscr{D}]$ and of the form

$$\mu_\tau = \alpha \ln\left[\frac{(\Delta z)^2}{\mathscr{D}}\right] + \beta, \tag{7}$$

where α is the slope and β the y-intercept of a linear least-squares fit. The preceding equation may be rearranged as follows:

$$\mu_\tau = \ln\left\{\left[\frac{(\Delta z)^2}{\mathscr{D}}\right]^\alpha\right\} + \beta. \tag{8}$$

For the units of the mean log-travel time, μ_τ, to be consistent in the preceding equation, α must be unity. Further, the form of the particle tracking equation (3c) suggests that travel times are proportional to $(\Delta z)^2$ and inversely proportional to $\mathscr{D}$ (and therefore proportional to d_p). Using a linear least-squares procedure, we fit Eq. (8) to the 100 numerically determined data points for μ_τ, thereby specifying the 95% confidence interval for the y-intercept, $\beta = -0.978 \pm 0.012$. Consequently, the mean of the log-travel times can be expressed as

$$\mu_\tau = \ln\left[\frac{(\Delta z)^2}{\mathscr{D}}\right] - 0.978. \tag{9}$$

Fig. 4 shows the 100 numerically determined values for μ_τ together with the best linear fit. The correlation coefficient between the numerical data points and Eq. (9) is $R^2 \approx 1$, indicating a near perfect fit.

Simulation results show that the values of the standard deviation of the log-travel times for each particle plume are independent of Δz and $\mathscr{D}$. The arithmetic mean and variance of the standard deviations of the log-travel times are 0.787 and 0.001, respectively, yielding a 95% confidence limit for the standard deviation in log-travel times of

$$\sigma_\tau = 0.787 \pm 0.002. \tag{10}$$

The standard deviation of log-travel times may be viewed as a measure of the difference between the fastest and slowest log-travel times for each particle plume, or the range of log-travel times. While these differences are constant on the log scale, the differences between the fastest and slowest actual travel times are inversely proportional to the molecular diffusion coefficient and proportional to the particle diameter (see Fig. 2). For example, the range of travel times for 95% of the particles to travel the specified distance may be derived from properties of the normal distribution as $\exp(\mu_\tau + 1.96\sigma_\tau) - \exp(\mu_\tau - 1.96\sigma_\tau)$. Comparing the range of travel times for the values used in Fig. 3a and b shows that the ratio of these ranges is 0.101, essentially the ratio of the diameters of the particles used for these simulations. Larger particles take a longer time to diffusively travel a specified distance while having a correspondingly larger range of travel times.

Substituting Eqs. (9) and (10) into Eq. (6) yields

$$Z(\mu_\tau, \sigma_\tau^2) = \ln\left[\frac{(\Delta z)^2}{\mathscr{D}}\right] - 0.978 + 0.787 Z(0,1). \tag{11}$$

Taking the inverse log of the previous equation yields a log-normally distributed travel time. Consequently, the

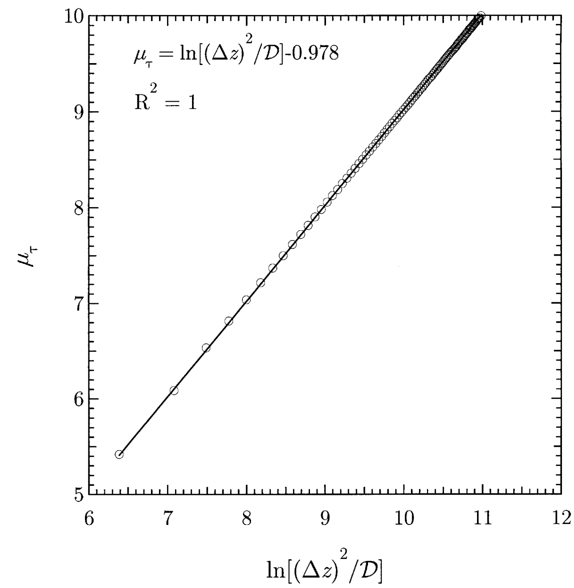

Fig. 4. Variation of the mean log-travel time, μ_τ, as a function of the log of the ratio $(\Delta z)^2/\mathscr{D}$. The equation for the least-squares fit (solid line) of the numerically determined values (open circles) and the corresponding correlation coefficient are shown.

time step necessary for a particle to travel a specified distance, Δz, may be written as

$$\Delta t = \exp[Z(\mu_\tau, \sigma_\tau^2)] = \exp\left\{\ln\left[\frac{(\Delta z)^2}{\mathscr{D}}\right] - 0.978 + 0.787Z(0,1)\right\}. \quad (12)$$

A new particle tracking equation is obtained by recasting the preceding equation to describe the current time of a particle as a function of its previous time, specified travel distance, and particle diffusion coefficient as follows:

$$t^m = t^{m-1} + \exp\left\{\ln\left[\frac{(z^m - z^{m-1})^2}{\mathscr{D}}\right] - 0.978 + 0.787Z(0,1)\right\}. \quad (13)$$

Because $Z(0,1)$ ranges over all real numbers, time steps can be anywhere from 0 to ∞; however, with six significant digits, the standard normal distribution can computationally only have values between ± 7.25. To find the time for a particle to move a specified distance, $\Delta z = z^m - z^{m-1}$, select a single value from the standard normal distribution and substitute it into Eq. (13). It should be noted that Reimus (1995) has presented, without proof, the following particle tracking equation:

$$\Delta t = \exp\left\{\ln\left[\frac{(\Delta z)^2}{2\mathscr{D}}\right] - 0.2 + 0.79Z(0,1)\right\} = \exp\left\{\ln\left[\frac{(\Delta z)^2}{\mathscr{D}}\right] - 0.89 + 0.79Z(0,1)\right\}. \quad (14)$$

Although Eqs. (13) and (14) are similar, the coefficients of Eq. (14) are well outside the confidence intervals presented for the coefficients of Eq. (13). For the mean, the coefficients presented by Reimus (1995) are over 14 standard deviations from the revised estimate and approximately three from the revised estimate of the standard deviation.

2.3. Extension to two and three dimensions

The same procedure that was used to specify the new particle tracking equation in one dimension may be used to determine the form of constant spatial step particle tracking equations appropriate for travel distances in two and three dimensions. Defining a spatial step, Δr, the time required for a plume of colloids, released at the origin, to exit a disk (two dimensions) or a sphere (three dimensions) with radius r can be derived. The constant spatial step particle tracking equation in two dimensions is

$$t^m = t^{m-1} + \exp\left\{\ln\left[\frac{(r^m - r^{m-1})^2}{\mathscr{D}}\right] - 1.578 + 0.649Z(0,1)\right\}, \quad (15)$$

and in three dimensions is

$$t^m = t^{m-1} + \exp\left\{\ln\left[\frac{(r^m - r^{m-1})^2}{\mathscr{D}}\right] - 1.939 + 0.575Z(0,1)\right\}. \quad (16)$$

To implement either of the preceding equations in a particle tracking algorithm, another uniformly distributed random number must be selected that describes the angle on the disk where the particle lands (and one more for the azimuth on a sphere).

3. Verification

Consider a uniform fracture with aperture $b = 5 \times 10^{-5}$ m that is saturated with water (288.15 K) that is flowing with a Poiseuille velocity profile with maximum centerline velocity of $U_{\max} = 1 \times 10^{-6}$ m/s. A polydisperse colloid plume of 10,000 particles with neutral buoyancy and log-normally distributed diameters with arithmetic mean 1 μm (mean of the log-diameter is −14.11) and standard deviation 0.9 μm (standard deviation of the log-diameters is 0.77) is instantaneously injected at the fracture inlet at time zero. The times required for each of the 10,000 particles to travel 8 m are used to generate a cumulative normalized particle breakthrough curve. Three methods are used to obtain these breakthrough curves: the first method employs the traditional particle tracking algorithm outlined by James and Chrysikopoulos (1999) with a constant time step, the second method utilizes the new particle tracking algorithm with a constant spatial step derived in this work, while the third method employs an analytical solution for polydisperse particle transport in a water saturated, uniform aperture fracture (James & Chrysikopoulos, 2001a).

The traditional particle tracking transport equation consists of a non-stochastic or absolute term, the advection, and a stochastic term representing the random molecular diffusion (Thompson & Gelhar, 1990; Thompson, 1993; Kitanidis, 1994). For the case of particles flowing in a uniform aperture fracture, the appropriate particle tracking equations are

$$x^m = x^{m-1} + U_{\max}\left[1 - 4\left(\frac{z^{m-1}}{b}\right)^2\right]\Delta t + Z(0,1)\sqrt{2\mathscr{D}\Delta t}, \quad (17a)$$

$$z^m = z^{m-1} + Z(0,1)\sqrt{2\mathscr{D}\Delta t}. \quad (17b)$$

The preceding particle tracking model assumes that every particle undergoes an incremental movement during each time step. As a particle diffuses across streamlines it samples different portions of the parabolic velocity profile and thus is subject to different velocities at t^{m-1} and t^m. Implicit to the particle tracking theory is the assumption that over a single time step a particle travels with the velocity with which it began and that changes in the velocity field over a time step do not compromise the accuracy of the solution. The colloids are introduced at the inlet side of the fracture flow domain ($x = 0$) and distributed according to the local volumetric flow rate (Reimus, 1995; James & Chrysikopoulos, 2000). When a particle encounters a wall, it is reflected back as in a mirror image. That is, the final x-location remains unchanged, whereas the final z-coordinate is set a distance away from the wall equal to the distance that the particle would have obtained if it had penetrated the rock matrix plus the particle diameter. For example, if a particle of $d_p = 5 \times 10^{-7}$ m initially diffuses to a z-location of 2.53×10^{-5} m (2.5×10^{-5} m being the location of the fracture wall), its reflected z value would be 2.42×10^{-5} m. A large number of particles is used in an effort to reduce random noise. A time step of $\Delta t = 0.9$ s was selected for use in the traditional particle tracking equation because, according to the new particle tracking equation (13), the mean diffusive travel time necessary for the smallest particle of the polydisperse plume ($d_p = 1 \times 10^{-8}$ m) to move a distance equal to one-fourth of the aperture is 0.9 s. It should be noted that if Eqs. (17a) and (17b) are used with a time step of larger than about 5 s, then the smallest particles of the plume may diffuse distances so great as to compromise the accuracy of the solution (i.e., an adequate sampling of all velocities within the fracture may not be taken before the particle exits the fracture). A cumulative normalized breakthrough curve is generated by tracking the number of particles that exit the fracture at 8 m.

Another particle breakthrough curve was also generated using the new particle tracking algorithm with a constant spatial step equal to one-fourth of the aperture ($\Delta z = 1.25 \times 10^{-5}$ m). Colloids are again placed in the fracture as a function of the flow rate. Note that for the case considered here molecular diffusion is the only transport mechanism in the z-direction; consequently, a constant spatial step in z may be used to determine the time step. In the x-direction, the particle tracking equation (17a) is used to simulate particle motion; however, the spatial step in the z-direction is specified and the corresponding time step, to be used in Eq. (17a), is calculated from Eq. (13). The appropriate particle tracking equation in the z-direction is given by

$$z^m = z^{m-1} \pm \Delta z, \tag{18}$$

where the direction of the displacement, $\pm\Delta z$, is determined from the sign of a standard normally distributed random number, $Z(0, 1)$. Fracture walls are treated as reflective boundaries, that is, when a particle encounters a wall, it is reflected back as in a mirror image. A cumulative normalized breakthrough curve is generated by tracking the number of particles that exit the fracture at 8 m.

A cumulative normalized particle breakthrough curve at $x = 8$ m is also generated from the following analytical solution (James & Chrysikopoulos, 2001a):

$$\bar{n}_{d_p}(x,t) = \int_0^{\infty} \frac{n_{\rm pdf}}{(4\pi D_{\rm eff} t)^{1/2}} \exp\left[-\frac{(x - U_{\rm eff} t)^2}{4 D_{\rm eff} t}\right] \mathrm{d}d_p, \tag{19}$$

where $\bar{n}_{d_p}(x,t)$ is the average concentration of polydisperse particles at any x-location along the fracture, and $n_{\rm pdf}$ is the particle size log-normal pdf for the polydisperse particles instantaneously injected as a plane source at the fracture inlet, $U_{\rm eff}$ is the effective velocity of the particle plume defined as (James & Chrysikopoulos, 2001b)

$$U_{\rm eff} = \frac{2}{3} U_{\max} \left[1 + \frac{d_p}{b} - \frac{1}{2}\left(\frac{d_p}{b}\right)^2\right] \tag{20}$$

and $D_{\rm eff}$ is the effective dispersion coefficient of the plume expressed by (James & Chrysikopoulos, 2001b)

$$D_{\rm eff} = \mathscr{D} + \frac{2}{945} \frac{U_{\max}^2 b^2}{\mathscr{D}} \left(1 - \frac{d_p}{b}\right)^6. \tag{21}$$

The cumulative particle breakthrough curves determined from each solution method considered are indistinguishable and therefore not shown. However, the computational time required to produce each solution is quite different. Using a PC with an 866 MHz Pentium III processor, the traditional particle tracking model (Eqs. (17a) and (17b)) required 32,071 s of CPU time to generate a solution while the new particle tracking scheme (Eqs. (13), (17a) and (18)) required 3100 s, an order of magnitude faster. Even with the maximum $\Delta t = 5$ s, the traditional particle tracking equation is still less efficient than the new one by approximately a factor of two. It is apparent that the new particle tracking scheme is computationally efficient and suffers no serious loss of accuracy.

Fig. 5 compares the cumulative normalized breakthrough curves for the constant spatial step particle tracking algorithms using the new particle tracking equation (13) and Eq. (14) presented by Reimus (1995). It is shown that the new particle tracking equation may improve accuracy. Eq. (14) has an upwardly biased time step owing to the constant -0.89 as opposed to -0.978 in Eq. (13). For larger selections of the spatial step, the larger time step obtained from Eq. (14) will not allow all colloids to sample all portions of the velocity profile before exiting the fracture, thereby affecting particle breakthrough times. If a smaller constant spatial step is used in the equation suggested by Reimus (1995), the cumulative breakthrough curves coincide; however, the computation cost increases.

4. Summary

Particle tracking methods are able to solve increasingly complex contaminant transport problems with the

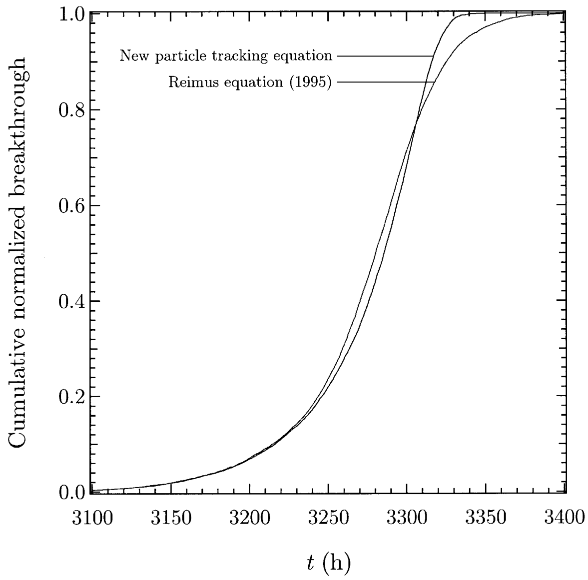

Fig. 5. Comparison of cumulative normalized particle breakthrough curves determined by the new particle tracking equation, Eq. (13), developed in this work and Eq. (14) suggested by Reimus (1995) for $\Delta z = 1.25 \times 10^{-5}$ m.

rapid advances in computing power. In cases where a constant time step is inappropriate (e.g., polydisperse particle plumes or reactive particles), it may be necessary to determine the (random) time a particle takes to diffusively travel a specified distance. However, it is not possible to simply retrieve the time step directly from the traditional particle tracking equation. Because the size of a particle affects how it diffuses in a quiescent fluid, differently sized particles require different times to travel a given distance. Histograms of travel times for plumes of monodisperse particles were consistently log-normal in shape. Thus, probability densities of the log-travel times are normally distributed. The parameters describing these normal distributions (i.e., mean and standard deviation of the log-travel times), are functions of the distance specified for travel and the diffusion coefficient of the particles. A constant standard deviation of log-travel times was found in each numerical simulation regardless of the parameter values, Δz and $\mathscr{D}$. Using a least-squares method, a linear relationship was found between the mean of the log-travel times and $\ln[(\Delta z)^2/\mathscr{D}]$. Employing the expressions obtained for the mean and standard deviation of the log-travel times, a new one-dimensional particle tracking equation with specified spatial step was determined. Appropriate particle tracking equation with specified spatial step applicable to two and three dimensions are also presented. Using both the traditional and the new particle tracking algorithms to model polydisperse colloid transport in a fracture, a comparison of computational times proves that the new particle tracking scheme derived here is more efficient than the traditional particle tracking method. Cumulative normalized particle breakthrough curves from both constant spatial step and constant time step particle tracking compare favorably with an available analytical solution. The new particle tracking scheme is quite robust and may be applicable to particle tracking techniques where it is more appropriate to specify a spatial step than a temporal step.

Notation

b	fracture aperture (L)
$\mathbf{B}$	deterministic scaling tensor ($Lt^{-1/2}$)
d_p	particle diameter (L)
D_{eff}	effective dispersion coefficient for a particle plume (L^2t^{-1})
$\mathscr{D}$	molecular diffusion coefficient (L^2t^{-1})
$f(\tau)$	normal probability density function (dimensionless)
k	Boltzmann's constant ($ML^2t^{-2}T^{-1}$)
m	time/spatial step number (dimensionless)
$\bar{n}_{d_p}$	number concentration of polydisperse particles averaged across the fracture (L^{-3})
n_{pdf}	probability density function of polydisperse particles (L^{-4})
r	radius of the disk or sphere used to generate the new particle tracking equations in two and three dimensions (L)
Δr	specified two or three dimensional spatial step (L)
R^2	correlation coefficient (dimensionless)
t	time (t)
Δt	time step, equal to $t^m - t^{m-1}$ (t)
T	absolute temperature of the suspending fluid (T)
U_{eff}	effective velocity of a particle plume (Lt^{-1})
$U_{\max}$	maximum interstitial fluid velocity along the fracture centerline in the x-direction (Lt^{-1})
x	coordinate location (L)
$\mathbf{X}$	three-dimensional position vector (L)
y	coordinate location (L)
z	coordinate location (L)
Δz	specified spatial step, equal to $z^m - z^{m-1}$ (L)
$Z(0,1)$	standard normally distributed random number (dimensionless)
$Z(\mu,\sigma^2)$	normally distributed random number with mean μ and variance σ^2 (dimensionless)
$\mathbf{Z}(0,1)$	three-dimensional vector of standard normally distributed random numbers (dimensionless)

Greek letters

α	slope of the least-squares fit (dimensionless)
β	y-intercept of the least-squares fit (dimensionless)
η	dynamic viscosity of the solvent ($ML^{-1}t^{-1}$)

μ_τ mean of the log-travel times (dimensionless)
σ_τ standard deviation of the log-travel times (dimensionless)
τ log-travel time for a particle, equal to $\ln \sum \Delta t$ (dimensionless)

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