MATHEMATICAL MODELING OF THE ANOMALOUS TRANSPORT OF RADIOACTIVE MATERIALS IN A POROUS MEDIUM

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ABSTRACT

The subsurface nuclear waste repositories have several engineered and natural barriers that isolate the radioactive material from the human's environment until the radio-toxicity of the waste decays to insignificance. One of the major natural insulating barriers is rock formation. If due to the various reasons the leakage of the radioactive waste would take place, the groundwater reservoirs in the vicinity of the repository can be seriously contaminated by the radioactive elements transferred through the cracks and fractures within the insulating barriers. Aquifer contamination by contaminants radioactive elements is an actual environmental problem for all developed countries. Analysis of mass transport in a complex environment shows that the conventional diffusion equation based on Fick's law fails to model the anomalous character of the diffusive mass transport observed in the field and laboratory experiments. Two regimes of anomalous diffusion are identified. One regime, which is called sub-diffusion, is characterized by the slower propagation of the concentration front, so that the squared distance of the front passage requires longer time than in the case of the classical Fickian diffusion. The second regime (called super-diffusion) is characterized by the higher diffusion rate. Both regimes can be modelled by non-local diffusion equation with temporal and spatial fractional derivatives. In the present paper fractional differential equations are used for modeling the transport of radioactive materials in fractured porous medium. New form of fractional equation for modeling migration of the radioactive elements is proposed and justified. Solutions of particular boundary value problems for these equations were found by application of the Laplace transform method. As an example, a mathematical model of the radioactive contaminant transport in a confined, porous, fractured aquifer is derived and analysed. Through the use of fractional derivatives, the model accounts for contaminant exchange between fissures and randomly distributed porous blocks of fractal geometry and non-local character of radioactive decay of the contaminant trapped by the porous medium. For the case of an arbitrary time-dependent source of radioactive contamination located at the inlet of the aquifer, closed-form solutions for solute concentration in the aquifer and in the confining rock is obtained.

INTRODUCTION

Aquifer contamination by radioactive elements is a widespread environmental problem. In many countries it is common to use fractured bedrock aquifers as water supply and contamination of these aquifers is becoming a serious problem (Keller et al., 1995). The fractured porous aquifers are formed by porous rock matrixes of nonzero porosity dissected by a fractal-type network of fissures of high hydraulic conductivity (Fomin et al, 2003). For fractured porous medium, it can be said that fluid is stored in the porous elements and transported along the fissures. Water flow and solute transport by seeping groundwater are relatively slow, and it is not possible to carry out experiments over thousands of years and hundreds of meters of interest. Instead, one has to rely on models that describe the processes and mechanisms that would be dominant over long periods of time. It is therefore essential to understand the key processes well enough that credible predictions can be made using models based on well-established laws of nature. The lack of measurement data makes it difficult to build models that account for all the processes. Simplications are therefore made in an attempt to bring out the dominant processes; Recently, Fomin et al. (2005,2010, 2011) suggested a relatively simple model, with fractal retention times, capable of simulating the anomalous character of solute concentration distributions for the flows in fractured porous media of fractal geometry. In the field experiments carried out by Becker and Shapiro (2000), Haggerty et al. (2000) and Reimus et al. (2003) for the solute transport in highly heterogeneous media, the solute concentration profiles exhibited faster-than-Fickian growth rates, skewness, and sharp leading edges. These effects cannot be predicted by the conventional mass transport equations. It was demonstrated in a number of publications (e.g. Benson et al., 2000, 2001; Baeumer, 2001; Schumer et al., 2003; Meerschaert et al., 1999 and Herrick et al., 2002, Fomin et al 2005) that fractional differential equations can simulate the anomalous character of solute transport in highly heterogeneous media. Suggesting inclusion a fractional-in-time derivative into the mass transport equation (in addition to the conventional derivative with respect to time), Schumer et al. (2003) referred to the conceptual model of multi-rate diffusion into immobile zones that had been described by Cunningham et al. (1997), Haggerty and Gorelick (1995), Carrera et al. (1998) and Haggerty et al. (2000).
In the present study a mathematical model of the radioactive contaminant transport in a fracture within the fractured porous medium is derived and analyzed. Through the use of fractional derivatives, the model accounts for the effects of contaminant transport retardation caused by non-Fickian diffusion into the porous blocks within the aquifer and into the confining rock. For the case of an arbitrary time-dependent source of contamination located at the inlet of the fracture, closed-form solutions for radioactive solute concentration in the fracture and in the surrounding rock are obtained.

SYSTEM MODEL AND ANALYSIS

In the A schematic sketch of the fracture within the porous rock is presented in Fig. 1. Cartesian coordinates (x, y) are chosen in such a manner that fluid in the fracture flows in the x-direction and that the coordinate y is directed upward. Within the fracture the certain portion of the solute adsorbs on its walls and the other portion diffuses into surrounding porous matrix, where also takes part the adsorption on the walls of the pores. The radioactive decay of the radioactive contaminant takes place both in fracture and porous matrix.

\[ c = \frac{1}{h} \int_{h}^{y} c \, dy \] , equations that model radioactive mass transport in the fracture and porous medium can be presented in the following form (Tang et al., 1981):

\[
\frac{\partial c}{\partial \tau} + v \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial x^2} - \frac{1}{h} \frac{\partial s}{\partial \tau} - \lambda c - \frac{s}{h} + \frac{q}{h} \quad (0 < x < \infty) \quad (1)
\]

\[
\frac{\partial c_1}{\partial \tau} + \frac{\partial c_1}{\partial x} = D_1 \frac{\partial^2 c_1}{\partial x^2} - \lambda_1 c_1 - \frac{s_1}{h} \quad (-h < y < \infty) \quad (2)
\]

where \( v \) is the average velocity of the solution in the fracture, \( \lambda \) is a radioactive decay constant, \( \tau \) is time, \( s \) is the mass of the solute adsorbed on the walls of the fracture, \( q \) is the diffusive mass flux on the wall of the fracture, \( D \) and \( D_1 \) are the effective diffusivities in the fracture and in the porous medium, which account for dispersion and molecular diffusion in the fracture and porous medium, \( s_1 \) is the mass of the contaminant within the matrix, which is adsorbed on the walls of the pores, \( \rho_m \) is the density of the rock matrix, \( \theta \) is the matrix porosity, and \( \frac{\partial^\alpha c_1}{\partial y^\alpha} \) is the fractional derivative of order \( \alpha \), \( 0 < \alpha \leq 1 \), that can be defined by means of Laplace transformation \( L \), from the equation

\[
L \left[ \frac{\partial^\alpha c_1}{\partial y^\alpha} \right] = p^{\alpha-1} (pL[c_1] - c(\tau,0)) \],
\]

which is equivalent to the Caputo definition (Samko et al., 1993),

\[
\frac{\partial^\alpha c_1}{\partial y^\alpha} = \left[ \frac{(y-\xi)^{\alpha-1}}{\Gamma(1-\alpha)} \frac{dc_1}{d\xi} \right]_{\xi=y}.
\]

Inclusion of the fractional derivative into equation (2) is attributed to the fact that pore geometry and distribution in the rocks is very complex and often resembles the fractal structure. For these type of media the conventional mass Fickian mass flux can be generalized by introducing the fractional derivative, so that it will take the following form, \( j = -\theta D_1 \frac{\partial^\alpha c_1}{\partial y^\alpha} \). Apparently, the mass flux \( q \) on the wall of the fracture, \( y=0 \), should have the same form, i.e.

\[
q = -\theta D_1 \frac{\partial^\alpha c_1}{\partial y^\alpha} \bigg|_{y=0} \quad (3)
\]

The system of equations (1)-(3) will be closed when the relationships between \( c \) and \( s \) along with \( c_1 \) and \( s_1 \) are known. With the good degree of accuracy it can be assumed (Tang et al., 1981) that

\[
S = K_c c, \quad S_1 = K_c c_1 \quad (4)
\]

\[
S_1 = K_m c_1 \quad (5)
\]

Where \( K_f \) and \( K_m \) are given constants. Substituting correlations

\[
(3)-(5) \text{ into equations (1) and (2) yields}
\]

\[
R \frac{\partial c}{\partial \tau} + v \frac{\partial c}{\partial x} - D \frac{\partial^2 c}{\partial x^2} \frac{1}{h} \frac{\partial s}{\partial \tau} + R \lambda c = -\theta D_1 \frac{\partial^\alpha c_1}{\partial y^\alpha} \bigg|_{y=0} \quad (6)
\]

\[
R \frac{\partial c_1}{\partial \tau} - D_1 \frac{\partial^2 c_1}{\partial x^2} \frac{\partial c_1}{\partial y^\alpha} - R \lambda c_1 = 0 \quad (-h < y < \infty) \quad (7)
\]

where \( R = (1 + \frac{\rho_m K_f}{\theta}) \) and \( R = (1 + \frac{K_m}{h}) \) are retardation coefficients. The order of the fractional derivative in equations (6) and (7), \( \alpha \), is close to 0 for highly heterogeneous media and
increases for more homogeneous substances; \( \alpha = 1 \) for homogenous materials. In general, concentration \( c_1 \) is a function of both spatial coordinates, \( x \) and \( y \): \( c_1 = c_1(t, x, y) \). However, in the equation (2) the derivative of \( c_1 \) with respect to \( x \) is ignored. Dependence of \( c_1 \) on \( x \) is a consequence of the boundary conditions on the rock-fracture interface \( (y = 0) \), which couples \( c_1 \) with the mean concentration in the fracture, \( c = c(t, x) \). In order to convert equations (6) and (7) to the non-dimensional form, the proper characteristic scales must be defined. The scale for time, \( \tau_0 = h^a v R / D \), represents the characteristic time for contaminant penetration in the rock matrix to the distance \( h \). The scale for the variable \( x \) along the fracture, \( l = v \tau_0 / R = (h^a v R)/RD \), is the characteristic distance of contaminant migration during the characteristic time \( (0, \tau_0) \). The scale for the \( y \)-coordinate is defined by the half-thickness of the aquifer, \( h \). The initial concentration of solute at the inlet of the fracture, \( c_0(0) \), where the source of contamination is located, can be used as the scale for solute concentration. Based on these scales, non-dimensional variables can be introduced as follows:

\[
C_1 = \frac{c_1}{c_0(0)}; \quad C = \frac{c}{c_0(0)}; \quad \bar{c}_1 = \frac{c_1}{c_0(t)}; \quad Pe = \frac{v l}{D};
\]

\( t = \frac{\tau}{\tau_0}; \quad X = \frac{x}{h}; \quad Y = \frac{y}{h}; \quad \Lambda = \frac{\lambda R h^a}{D} \) \hspace{1cm} (8)

Substituting the non-dimensional variables given by (8) into equations (6) and (7) yields the following:

\[
\frac{\partial C}{\partial t} - \frac{1}{Pe} \frac{\partial C}{\partial X} + \frac{\partial \bar{c}_1}{\partial X} + \Lambda C = \theta \frac{\partial c_1}{\partial Y}; \quad \text{at } t = 0, \ C = C_1 = 0; \quad \text{at } X = 0, \ C = C_1(T); \quad \text{at } Y = \infty, \ C \to 0; \quad \text{at } Y = 0, \ C_1 = C; \quad \text{at } \bar{c}_{1} = \Lambda C_1; \quad 0 < Y < \infty; \quad t > 0 .
\]

(10)

The following boundary and initial conditions can be imposed in order to close the mathematical model of the radioactive contaminant transport within the single fracture and surrounding porous medium:

\[
t = 0, \ C = C_1 = 0 ; \quad X = 0, \ C = C_1(t); \quad X \to \infty, \ C \to 0; \quad Y \to \infty, \ C_1 \to 0; \quad Y = 0, \ C_1 = C; \quad \text{where } C_0(t) \text{ is the non-dimensional concentration at the inlet of the fracture. Note that assuming the concentration } C_1 \text{ on the rock-aquifer interface } Y = 0 \text{ to be equal to the mean concentration of solute in the aquifer, } C, \text{ (boundary condition (15)) we slightly overestimate the value of concentration in the surrounding rocks. The boundary condition (15) is approximation of the more general mass transfer equation (Welty, 2001): } \text{Sh}(C_1 - C) \text{ at } Y = 0, \text{ where the Sherwood number } \text{Sh} = kh^a / D \text{ is greater than 10 for the regimes characterized by high Peclet numbers (due to the high values of convective mass transfer coefficient } k \text{, which is much greater than the effective diffusivity of the rock matrix (Guo et al., 1999; McGuire et al., 2004)).}
\]

**SOLUTION OF THE PROBLEM (9)-(15)**

In the present problem, two processes can be singled out: mass transport in a fracture and mass transport in a porous medium. Apparently, the first process is described by equation (9), which contains \( C \) and \( C_1 \). So, provided that correlation between \( C \) and \( C_1 \) is known, equation (9) with boundary conditions (11)-(13) constitutes the closed boundary value problem that can be solved separately from equation (10). This correlation can be obtained by integrating equation (10) with boundary conditions (14), (15) and initial condition (11). It can be readily shown that solution for concentration, \( C_1 \), can be coupled with the concentration in the fracture by the following equation:

\[
C_1(t, X, Y) = e^{-\omega t} \frac{\partial}{\partial Y} \int e^{\epsilon Y} C(r, X) u_0(t - r, Y) dY , \quad (16)
\]

which analogous to the Duhamel’s theorem (Carslaw and Jaeger, 1959). Function \( u \) in equation (16) is solution of the auxiliary problem:

\[
\frac{\partial u_0}{\partial t} = \frac{\partial}{\partial Y} u_0; \quad 0 < Y < \infty; \quad t > 0 ,
\]

\( t = 0, \ u_0 = 0; \quad Y = 0, \ u_0 = 1; \quad Y \to \infty, \ u_0 \to 0 ; \quad (17) \quad (18) \quad (19)
\]

Equation (16), in terms of mass fluxes, can be converted into the following form:

\[
Q = \frac{\partial C_1}{\partial Y} = -\frac{\partial}{\partial t} \int C(t - r, X) Q_1(t - r, 0) dr \quad (21)
\]

where

\[
Q_1(t) = e^{-\omega t} \frac{\partial}{\partial Y} \int e^{\epsilon Y} Q_0(t - r) dr \quad \text{and } \quad Q_0(t) = \frac{\partial u_0}{\partial Y} . \quad (22)
\]

Substituting formula (21) into equation (9) leads to the following boundary value problem for \( C \)

\[
\frac{\partial C}{\partial t} - \frac{1}{Pe} \frac{\partial C}{\partial X} + \frac{\partial \bar{c}_1}{\partial X} + \Lambda C = -\theta \frac{\partial}{\partial t} \int C(r, X) Q_1(t - r, 0) dr
\]

\( t = 0, \ C = 0 ; \quad X = 0, \ C = C_1(t); \quad X \to \infty, \ C \to 0 \quad (23) \quad (24) \quad (25) \quad (26)

Applying the technique of group Lie analysis (Akcenov et al. (1994)) it can be readily shown that solution of the above formulated boundary-value problem (17)-(20), for function \( u_0 \), can be presented in the following form (Fomin et al., 2010):

\[
u_0(\eta) = \sum_{m=0}^{\infty} \frac{(-1)^m}{m!} \int \frac{\alpha + m}{\Gamma(\alpha + 1)} \eta^m \Gamma(\alpha + m + 1) + \eta^{\alpha + M(m + 1)} \quad (27)
\]

where \( \eta = Y^\gamma \) and \( \Gamma(z) \) is Gamma function. From the formula (27) it follows that

\[
\frac{\partial^\nu u_0}{\partial Y^\nu} = -\frac{1}{\Gamma(1 - \beta) t^\beta} \quad (28)
\]
where $\beta = \alpha / (1 + \alpha)$. Accounting for the formula (28), formulae (22) lead to the following expression for $Q_1$, which represents the mass flux on the wall of the fracture when concentration $C_1$ on this wall is set to be equal to 1:

$$Q_1 = \frac{e^{-\Lambda t}t^{-\beta}}{\Gamma(1-\beta)} + \frac{\Lambda}{\Gamma(1-\beta)}[\Gamma(1-\beta) - \gamma(1-\beta, \Lambda t)]$$  \hspace{1cm} (29)

where $\gamma(a, z)$ is Incomplete Gamma function. Thus, formulae (21) and (29) define the actual mass flux on the wall of the fracture. In two particular cases that can be easily considered.

In the case when there is no radioactive decay and diffusion is Fickian (i.e. $\Lambda = 0$ and $\beta = 1/2$), $Q_1 = t^{-\beta} / \Gamma(1-\beta)$ and $Q = \frac{\partial Q}{\partial t}$. In the case when there is no radioactive decay but diffusion is described by the generalized Fick’s law (3), which corresponds to $\Lambda = 0$ and $0 < \beta < 1/2$, $Q_1 = t^{-\beta} / \Gamma(1-\beta)$ and $Q = \frac{\partial Q}{\partial t}$. It can be readily shown that for small $\Lambda$ and finite time $t=O(1)$ formula (29) admits the following asymptotic representation:

$$Q_1 = \frac{t^{-\beta}}{\Gamma(1-\beta)} + \frac{\Lambda t^{-\beta}}{\Gamma(1-\beta)} \left[ 1 - \frac{\Gamma(1-\beta)}{\Gamma(2-\beta)} \right] + O(\Lambda^2 t^{-\beta})$$  \hspace{1cm} (30)

where $\beta = \alpha / (1 + \alpha)$. The accuracy of this asymptotic formula can be easily verified by simple numerical computations.

In these new variables, $t$ can be presented as $t = \frac{\tau}{\Lambda}$, and according to (31), $t = \frac{\tau}{\Lambda}$ is small in (30) and $\beta \geq 0.1$ the discrepancy between the values of $Q_1$ computed by the formulae (29) and (30) is negligibly small and, therefore, formula can be used as a good approximation for $Q_1$, which exact value is given by equation (29). Using formula (30), the mass flux $Q$ defined by equation (21) can be presented through the fractional derivatives:

$$Q = \frac{\partial^\beta C}{\partial t^\beta} - \Lambda - \frac{\Gamma(1-\beta)}{\Gamma(2-\beta)} \left[ 1 - \frac{\Gamma(1-\beta)}{\Gamma(2-\beta)} \right] + O(\Lambda^2 t^{-\beta})$$  \hspace{1cm} (31)

Let us turn to solution of the boundary value problem (23)-(26) where function $Q$ in the right hand side of equation (23) is defined by the formula (31). Considering the same time period as above, i.e. from 0 to 1/\Lambda, it is convenient to rescale equation (23) assigning 1/\Lambda as a scale for time, so that new time variable $T = \Lambda t$. In this case, as it follows from the scale analysis of equation (23), the scale $l$ for the spatial variable $X$ should be converted to a new scale $X_0 = 1 / (\Lambda + \theta x^\theta)$ and the new spatial variable will be defined as $\bar{X} = X(\Lambda + \theta x^\theta)$. In these new non-dimensional variables equation (23) can be presented in the following form

$$w_1 \frac{\partial C}{\partial T} + \frac{\partial C}{\partial \bar{X}} - \frac{\partial^2 C}{\partial \bar{X}^2} + C = -w_2 \frac{\partial}{\partial T} \int C(T - \bar{X}) \Psi_\beta(r) d\bar{X}$$  \hspace{1cm} (32)

where $w_1 = \frac{\Lambda}{\Lambda + \theta x^\theta}$, $w_2 = \frac{\theta x^\theta}{\Lambda + \theta x^\theta}$, $\epsilon = \frac{\Lambda + \theta x^\theta}{Pe}$ and $\Psi_\beta = Q_1 \Lambda^{-\beta} - 1$. Note that accounting for the equation (29), expression for $\Psi_\beta$, can be presented as $\Psi_\beta = \left[ \Gamma^{1-\beta} e^{-\tau} - \gamma(1-\beta, T) \right] / \Gamma(1-\beta)$. Parameter $\epsilon$ in equation (32) is rather small ($\epsilon < 1$) because $\Lambda < 1$ and $Pe = O(1)$. Hence, for the moments of time of the order $1/\Lambda$, in major cases, the effects of the diffusive transport in overall mass transport within the fracture are negligible small (mass transport is predominantly determined by an advection mechanism) and, therefore, equation (32) can be rewritten as follows

$$w_1 \frac{\partial C}{\partial T} + \frac{\partial C}{\partial \bar{X}} + C = -w_2 \frac{\partial}{\partial T} \int C(T - \tau, \bar{X}) \Psi_\beta(r) d\tau$$  \hspace{1cm} (33)

Applying Laplace transformation $L$ with respect to variable $T$ to the equation (33), and accounting for boundary condition (24)-(26), gives

$$\frac{d\bar{C}}{d\bar{X}}(x) + \left( \frac{s w_1}{s} + 1 + sw_2 \Psi_\beta \right) \bar{C} = 0$$  \hspace{1cm} (34)

$$\bar{X} = 0, \quad \bar{C} = \bar{C}_0,$$  \hspace{1cm} (35)

where $\bar{C} = L[C] = \int \left[ \int C \exp(-sT) dT \right] d\tau$ and $\Psi_\beta = L[\Psi_\beta]$. Substituting $\Psi_\beta = L[\Psi_\beta] = [(s + 1)^\beta - 1] / s$ into equation (34) and integrating it accounting for the boundary condition (35) yields

$$\bar{C} = \bar{C}_0 \exp(-w_2 x \beta) \int e^{-w_2 x \beta \psi(r) + \psi(r \beta)} \left[ \int \frac{d\bar{X}}{(\bar{X} + 1)} \right]$$  \hspace{1cm} (36)

When at the entrance of the fracture concentration is constant, i.e. $C_0 = 1$ and $\bar{C}_0 = 1 / s$, denoting concentration in the fracture for this case by $C_f$ and calculating the inverse Laplace transformation $L^{-1}$ leads to the following expression:

$$C_f(T, \bar{X}) = e^{-w_2 x \beta} \int \left[ \int C_f(T - \tau, \bar{X}) G_f(r, \bar{X}) d\tau \right]$$  \hspace{1cm} (37)

where $H(x)$ is a Heaviside Step function and

If concentration in the inlet is an arbitrary function of $T$, $C_0(T)$, then concentration in the fracture can be obtained by utilizing the Duhamel’s theorem (Carslaw and Jaeger, 1959):

$$C_f(T, \bar{X}) = \int \left[ \int C_0(T - \tau) C_f(\tau, \bar{X}) d\tau \right]$$  \hspace{1cm} (38)

where $C_f$ is defined by equation (37). If $C_0$ varies exponentially, $C_0 = e^{-\tau}$, i.e. when the radioactivity decays at the entrance point, then solution $C$ can be obtained by the inverse Laplace transform directly from the equation (36), where $\bar{C}_0 = 1 / (s + 1)$.

As a result of this inversion

$$C_f(\alpha, T) = e^{-\left(1-\psi(r) + \psi(x) \beta \right)} H(T - \psi(x) \beta) \bar{C}_f(\psi(x) \beta, T - \psi(x) \beta),$$  \hspace{1cm} (39)

where $G_\beta(\alpha, T) = L^{-1} \left[ 1 / s \right] e^{-\alpha T}$

$$= \int \left[ \int \frac{d\bar{X}}{(\bar{X} + 1)} \right] e^{-\alpha \psi(x) \beta \cos(\pi x)} \sin(\alpha \psi(x) \beta \sin(\pi x)) d\bar{X}.$$  \hspace{1cm} (40)
Thus, solutions of the boundary value problem (23)-(26), which models the radioactive mass transport in a single fracture, is obtained analytically. In general case, this solution is given by equations (38) and (37). In particular case, when the solute concentration in the inlet of the fracture is constant or decays exponentially, then concentration in the fracture is given by expressions (37) and (39), respectively. When the concentration distribution within the fracture C is found, concentration of the solute in the surrounding porous matrix can be obtained from the formula (16), where $u_0$ is defined by the series (27).

References


