RADIONUCLIDE MIGRATION: A NUMERICAL STUDY

Claudia S. da Silveira, Zelmo R. de Lima, and Antonio C. M. Alvim

Programa de Engenharia Nuclear (COPPE/ UFRJ)
Caixa Postal 68509
21945-970, Rio de Janeiro, RJ
csilveira@con.ufrj.br, zelmolima@yahoo.com.br, aalvim@gmail.com

ABSTRACT

Crystalline rock has been considered as a potentially suitable matrix for high-level radioactive waste (HLW) repository because it is found in very stable geological formations and may have very low permeability. A common problem encountered in this context is the modeling of migration of radio nuclides in a fractured medium. Generally, this consists of a large main fracture, which is surrounded by a rock matrix. Transport in the main fracture is usually assumed to obey an advection-dispersion relation, while molecular diffusion is the assumed dominant mechanism of transport in the porous rock. In this work, a numerical study of the governing partial differential equations is done, to describe radionuclide movement in the fracture and within the rock matrix. The adopted physical system consists of the rock matrix containing a single planar fracture situated in water saturated porous rock. The initial radionuclide concentrations are assumed to be zero in both fractured and rock matrices. As inlet boundary condition, a kinetic solubility-limited dissolution model is used, in order to calculate the radionuclide concentration in the fracture. The solution of the governing partial differential equations was obtained by finite difference methods, namely: fully explicit, fully implicit and Crank-Nicolson discretization schemes. Note that the influence of the advective term was considered in the partial differential equation in the fracture, in such discretization schemes. It was shown that all numerical schemes are consistent and that the explicit method, in all configurations of the advective term, and the implicit methods and Crank-Nicolson, for the forward discretization in the advective term, presented stability conditions to be considered.

1. INTRODUCTION

This work analyzes some of the possible numerical methods that are applied to partial differential equations (PDE) describing the radionuclide transport inside a discrete fracture, based on a kinetic solubility-limited dissolution model at fracture entrance. The governing equations were discretized using finite difference techniques, where the following methods were adopted: Explicit Euler, Implicit Euler and Crank-Nicolson. And, for each one of these methods, the advective term was discretized with the following numerical schemes: backward differences, centered differences and forward differences.

Consistency of the numerical methods was analyzed by applying Taylor expansions to the discretized terms, in order to obtain the truncation error. Afterwards, the amplification factor was obtained via the Von Neumann stability analysis in order to derive stability conditions for the investigated methods. Finally, comparative plots are shown for all methods tested, as a function of the advective term discretization .

2. PROBLEM FORMULATION

The model used in this work is based on [1] and [2]. The geometry of the model can be observed in Fig. 1.
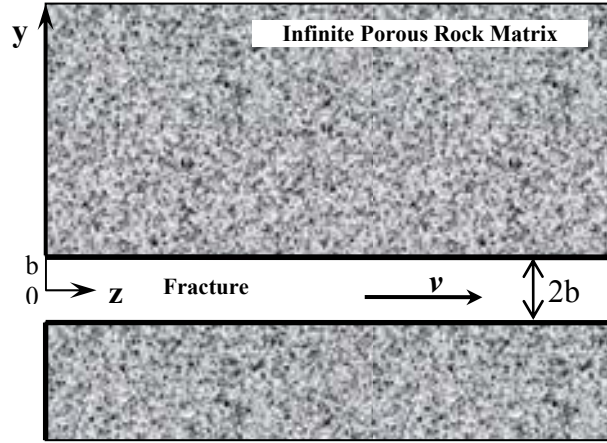
Figure 1. Geometry of the model with a single, planar, infinite fracture.

The partial differential equation that describes the movement of the radionuclide in a fracture is given by:

$$R_f \frac{\partial C_f(z,t)}{\partial t} - D_f \frac{\partial^2 C_f(z,t)}{\partial z^2} + v \frac{\partial C_f(z,t)}{\partial z} + R_f \lambda C_f(z,t) + \frac{q(z,t)}{b} = 0, \text{ for } z > 0, \ t > 0 \qquad (1)$$

where $C_f(z,t)$ is the concentration of the radionuclide in fracture water (Kg-nuclide/m³-water), R_f is the retardation factor in the fracture, K_f is the surface distribution coefficient of a solute per unit area of the fracture rock interface over the unit volume (m), D_f is the longitudinal dispersion coefficient (m² /yr), λ is the decay constant (1/yr), b is the fracture half width (m), $q(z,t)$ is the diffusive flux of a solute from a fracture into a rock matrix (Kg-nuclide / m²yr), z is the coordinate along the fracture (m) and, finally, t is the time in years. Similarly, the governing equation describing the movement of the radionuclide in a rock matrix is:

$$R_p \frac{\partial C_p(y,z,t)}{\partial t} - D_p \frac{\partial^2 C_p(y,z,t)}{\partial y^2} + R_p \lambda C_p(y,z,t) = 0, \text{ for } |y| \geq b, \ z > 0, \ t > 0 \qquad (2)$$

where R_p is the retardation coefficient in a rock matrix.

The initial conditions for the equations (1) and (2) are:

$$C_f(z,0) = 0, \text{ for } z > 0, \qquad (3)$$

$$C_p(y,z,0) = 0, \text{ for } |y| \geq b, \ z > 0 \qquad (4)$$

The boundary conditions in the fracture are given by:

$$-D_f \frac{\partial C_f(z,t)}{\partial z} + v C_f(z,t) = k[C_0 - C_f(z,t)], \text{ for } z = 0, \ t > 0 \qquad (5)$$

$$C_f(\infty, t) = 0 \text{, for, } t > 0 \tag{6}$$

where k is a constant rate (m/yr) and C_0 is the solubility of solute (Kg/m^3).

In equation (5), the terms on the left side represent advection and dispersion fluxes, while the term on the right side represents kinetic solubility-limited dissolution rate. The boundary conditions in the rock matrix are:

$$C_p(b, z, t) = C_f(z, t) \text{, for } z = 0 \text{, } t > 0, \tag{7}$$

$$C_p(\infty, z, t) = 0 \text{, for } z > 0 \text{, } t > 0, \tag{8}$$

The loss rate of diffusive flux crossing the fracture-rock interface is expressed by Fick's first law as:

$$q(z,t) = -\theta D_p \left. \frac{\partial C_p}{\partial y} \right|_{y=b} \text{, for } z > 0 \text{, } t > 0 \tag{9}$$

θ is the porosity of the rock, $C_p(z, y, t)$ is the concentration of the radionuclide in rock matrix water (Kg-nuclide/m^3-water), D_p is the pore diffusion coefficient in a rock matrix (m^2/yr) and y the coordinate perpendicular to the fracture axis (m). The equations (7) and (9) provide a coupling between equations (1) and (2).

3. NUMERICAL SOLUTION

The governing equations presented in the previous section were numerically approximated by finite difference techniques. For both equations (1) and (2) the terms with second order spatial derivatives were discretized with centered difference schemes and derivatives in time were discretized with the well known theta method. Besides, the advective term present only in eq. (1), was discretized with backward, centered and also forward differences. With these, considerations, eq. (1) can be rewritten as:

$$\frac{C_{f,i}^{n+1} - C_{f,i}^{n}}{\Delta t} = (1 - \Theta) \left[\frac{D_f}{R_f} \left(\frac{C_{f,i-1}^{n} - 2C_{f,i}^{n} + C_{f,i+1}^{n}}{\Delta z^2} \right) - \frac{v}{R_f} \left(\frac{C_{f,i+r}^{n} - C_{f,i-s}^{n}}{(r+s)\Delta z} \right) - \lambda C_{f,i}^{n} - \frac{1}{R_f b} q_i^{n} \right] +$$

$$+ \Theta \left[\frac{D_f}{R_f} \left(\frac{C_{f,i-1}^{n+1} - 2C_{f,i}^{n+1} + C_{f,i+1}^{n+1}}{\Delta z^2} \right) - \frac{v}{R_f} \left(\frac{C_{f,i+1}^{n+1} - C_{f,i-s}^{n+1}}{(r+s)\Delta z} \right) - \lambda C_{f,i}^{n+1} - \frac{1}{R_f b} q_i^{n+1} \right] \tag{10}$$

where, for $r = s = 1$ the advective term indicates centered differences, for $r = 0$ and $s = 1$ backward differences (upwind) and $r = 1$ e $s = 0$ corresponds to forward differences. For diffusion in the pores (the rock matrix), eq. (2) is rewritten in finite difference form as:

$$\frac{C_{p,i,j}^{n+1} - C_{p,i,j}^n}{\Delta t} = (1-\Theta)\left[\frac{D_p}{R_p}\left(\frac{C_{p,i,j-1}^n - 2C_{p,i,j}^n + C_{p,i,j+1}^n}{\Delta y^2} - \lambda C_{p,i,j}^n\right)\right] +$$

$$+ \Theta\left[\frac{D_p}{R_p}\left(\frac{C_{p,i,j-1}^{n+1} - 2C_{p,i,j}^{n+1} + C_{p,i,j+1}^{n+1}}{\Delta y^2} - \lambda C_{p,i,j}^{n+1}\right)\right]$$

(11)

In equations (10) and (11), subscripts i and j denote spatial nodes in z and y directions, respectively and index n denotes discretization in time. The parameter Θ is a real constant that varies between 0 and 1 [3]. For $\Theta = 0$ and $\Theta = 1$, the Explicit and Implicit Euler methods, respectively, are obtained. For $\Theta = 1/2$, the Crank-Nicolson semi-implicit method is obtained.

4. NUMERICAL ANALYSIS

For the numerical analysis of equations describing the radionuclide migration study of a consistence and stability study was done. As follows [4].

4.1. Consistency of Numerical Methods

To verify consistency, expand the terms of the discretization in Taylor series and make $\Delta z, \Delta y, \Delta t \to 0$. If the local truncation error (LTE) tends to zero, the discretization is consistent with the PDE. Making use of these expansions, we derived the following expressions for the truncation error associated with equations (10) and (11):

$$TLE = A\frac{\partial C_f}{\partial t}\bigg|_i^n + B\frac{\partial^2 C_f}{\partial t^2}\bigg|_i^n + C\frac{\partial^3 C_f}{\partial t^3}\bigg|_i^n + D\frac{\partial^2 C_f}{\partial z\partial t}\bigg|_i^n + E\frac{\partial^3 C_f}{\partial t\partial z^2}\bigg|_i^n + F\frac{\partial^3 C_f}{\partial z\partial t^2}\bigg|_i^n +$$

(12)

$$+ G\frac{\partial^2 C_f}{\partial z^2}\bigg|_i^n + H\frac{\partial^3 C_f}{\partial z^3}\bigg|_i^n + O\left[(\Delta z)^3, (\Delta z)^2\Delta t, \Delta z(\Delta t)^2, (\Delta t)^3\right],$$

$$TLE = A\frac{\partial C_p}{\partial t}\bigg|_{i,j}^n + B\frac{\partial^2 C_p}{\partial t^2}\bigg|_{i,j}^n + C\frac{\partial^3 C_p}{\partial t^3}\bigg|_{i,j}^n + J\frac{\partial^3 C_p}{\partial y^2\partial t}\bigg|_{i,j}^n + O\left[(\Delta t)^3, \Delta t(\Delta y)^2, (\Delta y)^3\right]$$

(13)

where:

$$A = -\Delta t\frac{\lambda}{2}, \quad B = -\lambda\frac{\Delta t^2}{4} - \frac{\Delta t}{2}, \quad C = -\frac{\Delta t^2}{6} - \lambda\frac{\Delta t^3}{12}, \quad D = -\frac{v}{R_f}\frac{\Delta t}{2}, F = -\frac{v}{4}\frac{\Delta t^2}{R_f},$$

$$G = \frac{v}{R_f}\frac{\Delta z}{2}, \quad H = -\frac{v}{R_f}\frac{\Delta z^2}{6}, \quad I = \frac{\Delta t}{2}\frac{D_p}{R_p}, J = \frac{v}{4}\frac{\Delta z\Delta t}{R_f}, \quad L = \frac{\Delta t}{2}\frac{D_f}{R_f},$$

(14)

Where $E = J + L$ corresponds to backward differences in the advective term, $E = L$ and $G = 0$ corresponds to centered differences and $E = L - J$ to forward differences. In view of the expressions (12), (13) and (14) and letting $\Delta z, \Delta y, \Delta t \rightarrow 0$ it can be observed that all the numerical schemes are consistent.

4.2. Stability of Numerical Methods

In this work one of the most employed techniques, known as von Neumann stability analysis [5], will be used in the study of the stability of finite differences equations. Starting from the Fourier expansion of the solutions, in this case, $C_{f,i}^n$ for the fracture and, $C_{p,i,j}^n$ for rock matrix we have:

$$C_{f,i}^n = \Phi^n e^{iQz_i} \quad \text{and} \quad C_{p,i,j}^n = \Phi^n e^{iQz_i} e^{iRy_j} \tag{15}$$

where $i = \sqrt{-1}$ and, Φ^n is the amplitude in instant n , Q and R are the wave numbers. For the terms in $n+1$, $i \pm 1$ and $j \pm 1$, similar expressions to equations (15) are obtained. The condition of stability to be satisfied is:

$$\left| \Phi^{n+1} / \Phi^n \right| \leq 1 \tag{16}$$

where $\left| \Phi^{n+1} / \Phi^n \right|$ is known as the amplification factor. Substituting these relationships into (15) and their similar ones for $n+1$, $i \pm 1$ and $j \pm 1$, equations (10) and (11), and imposing condition (16), one arrives at the conditions for stability shown in Tables 1, 2 and 3, for the equations in the fracture, and in Table 4, for the rock matrix.

Table 1. Stability for Equation in the Fracture
Case of Backward Differences in the Advective Term

Scheme	Stability Condition
Forward Euler ($\Theta = 0$)	$\dfrac{D_f \Delta t}{R_f (\Delta z)^2} + \dfrac{v \Delta t}{2 R_f \Delta z} + \dfrac{\lambda \Delta t}{4} \leq \dfrac{1}{2}$ $\left(\dfrac{v \Delta t}{R_f \Delta z} \right)^2 - \left[1 - \left(\dfrac{\lambda \Delta t}{2} \right)^2 \right] \leq 0$
Backward Euler ($\Theta = 1$)	Unconditionally Stable
Crank-Nicolson ($\Theta = 1/2$)	Unconditionally Stable

Table 2. Stability for Equation in the Fracture
Case of Centered Differences in the Advective Term

Scheme	Stability Condition
Forward Euler ($\Theta = 0$)	$$\frac{D_f \Delta t}{R_f (\Delta z)^2} + \frac{\lambda \Delta t}{4} \leq \frac{1}{2}$$ $$\left(\frac{v \Delta t}{R_f \Delta z} \right)^2 - \left[1 - \left(\frac{\lambda \Delta t}{2} \right)^2 \right] \leq 0$$
Backward Euler ($\Theta = 1$)	Unconditionally Stable
Crank-Nicolson ($\Theta = 1/2$)	Unconditionally Stable

Table 3. Stability for Equation in the Fracture
Case of Forward de Differences in the Advective Term

Scheme	Stability Condition
Forward Euler ($\Theta = 0$)	$$0 \leq \frac{2D_f \Delta t}{R_f (\Delta z)^2} - \frac{v \Delta t}{R_f \Delta z} + \frac{\lambda \Delta t}{2} \leq 1$$ $$\left(1 - \frac{\lambda \Delta t}{2} \right) \left(-\frac{2D_f \Delta t}{R_f (\Delta z)^2} + \frac{v \Delta t}{R_f \Delta z} - \lambda \Delta t \right) + \left(\frac{v \Delta t}{R_f \Delta z} \right)^2 \leq 0$$
Backward Euler ($\Theta = 1$)	$$\frac{2D_f \Delta t}{R_f (\Delta z)^2} - \frac{v \Delta t}{R_f \Delta z} + \frac{\lambda \Delta t}{2} \geq 1$$
Crank-Nicolson ($\Theta = 1/2$)	$$\frac{2D_f \Delta t}{R_f (\Delta z)^2} - \frac{v \Delta t}{R_f \Delta z} + \frac{\lambda \Delta t}{2} \geq 0$$

Table 4. Stability for Equation in the Pore

Scheme	Stability Condition
Forward Euler ($\Theta = 0$)	$$\frac{D_p \Delta t}{R_p (\Delta y)^2} + \frac{\lambda \Delta t}{4} \leq \frac{1}{2}$$
Backward Euler ($\Theta = 1$)	Unconditionally Stable
Crank-Nicolson ($\Theta = 1/2$)	Unconditionally Stable

5. NUMERICAL TESTS

To evaluate the numerical methods, a code was implemented in Fortran 90 language. The physical parameters for fracture entrance and rock matrix are shown in the Table (5), where the half-life time corresponds to radionuclide ^{237}Np. The choice of the discretized mesh was based on the stability conditions derived in Section 4.2. In all cases a spatial grid $\Delta z = 1.0$ m and $\Delta y = 0.2$ m was adopted, where the maximum lengths in the z and y-directions were 400 m and 80 m respectively. To satisfy Table 1 through 4 restrictions, we choosed $\Delta t = 1.0$(yr) for the implicit methods and Crank-Nicolson, in all configurations for the advective term, while for the explicit method, $\Delta t = 0.5$(yr) for forward differencing, $\Delta t = 0.3$(yr) for centered differences and $\Delta t = 0.25$(yr) for backward discretization of the advective term.

The comparative graphs of the methods with the profiles of the concentrations in the fracture and the rock matrix are plotted in the intervals of 10, 10^2, 10^3 and 10^4 years, see Figs. 2, 3 and 4, where Fig. 2 corresponds the backward discretization in the advective term, Fig. 3 the centered discretization and Fig. 4 the forward discretization.

In the solution of the tri-diagonal systems that appear in the implicit and Crank-Nicolson methods, LU decomposition was used with forward and backward substitution procedures. In Table (6) CPU times corresponding to implicit and Crank-Nicolson methods relative to the explicit method in each type of advective term discretization, registered in 10^4 years are shown.
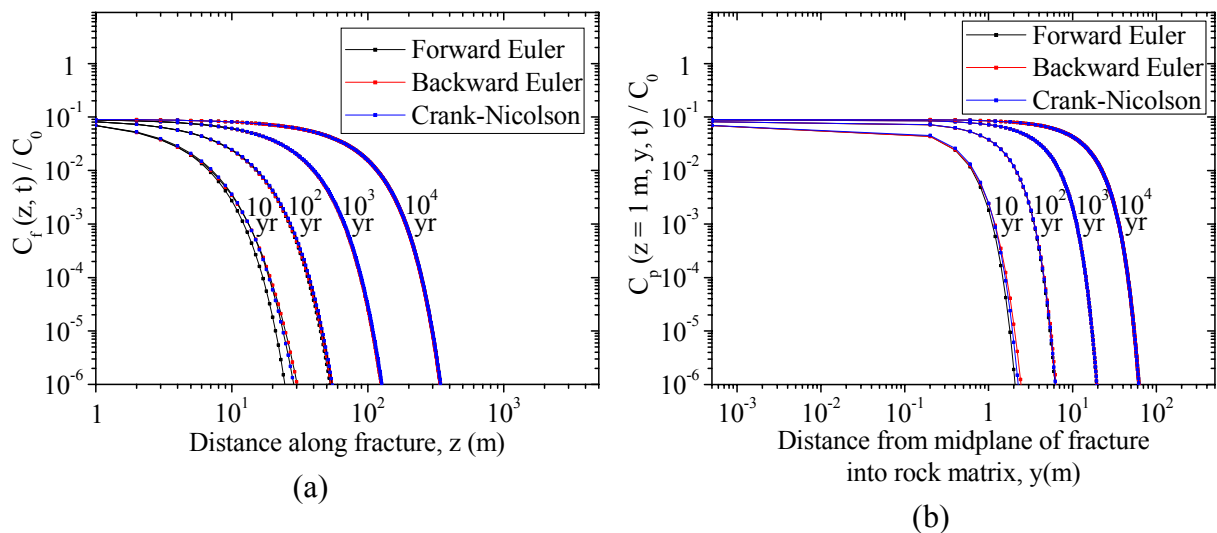

Figure 2. Profiles of Normalized Concentration in the Fracture (a) and Rock Matrix (b). Backward Discretization in Advective Term
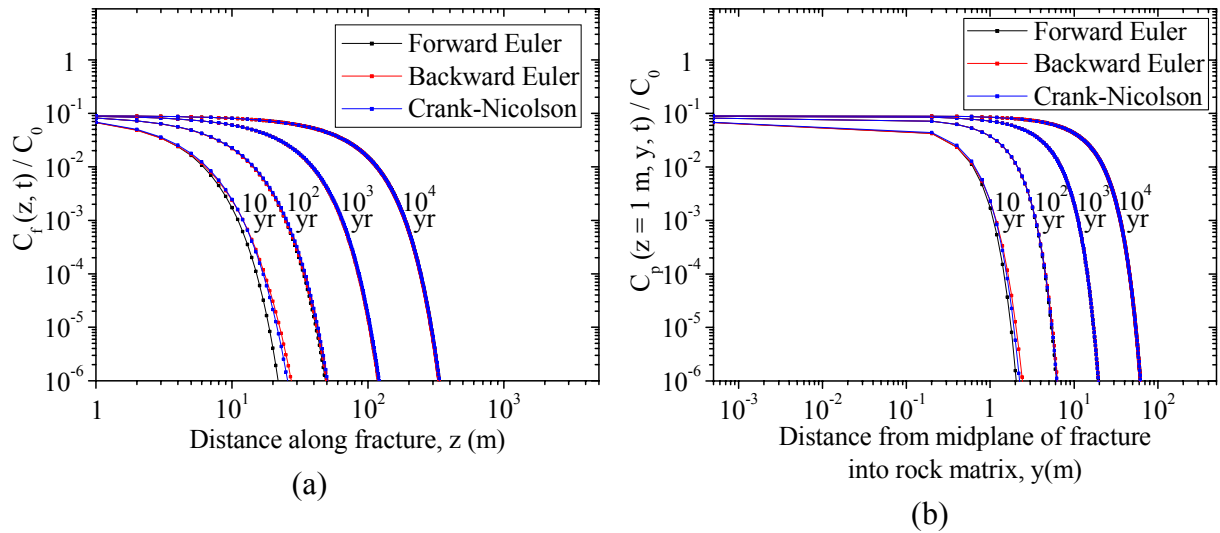
Figure 3. Profiles of Normalized Concentration in the Fracture (a) and Rock Matrix (b). Centered Discretization in Advective Term
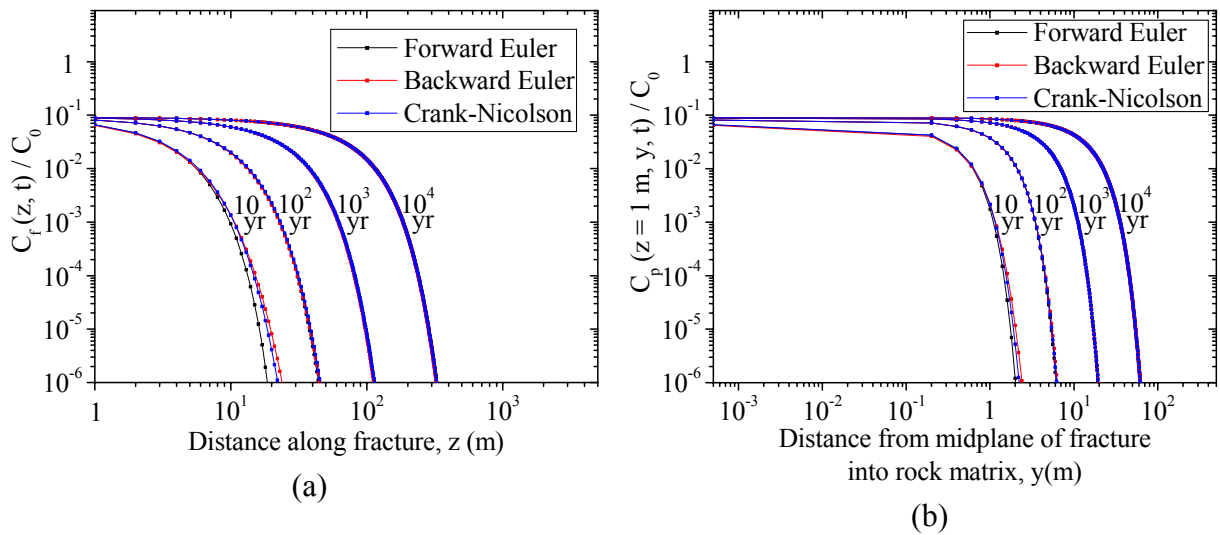

Figure 4. Profiles of Normalized Concentration in the Fracture (a) and Rock Matrix (b). Forward Discretization in Advective Term

Table 5. Physical Parameters for the Fracture and Rock Matrix

$D_f \ (m^2/yr)$	R_f	$D_p \ (m^2/yr)$	R_p	$v \ (m/yr)$	$b \ (m)$	θ	$k \ (m/yr)$	$T_{1/2}(yr)$
1.0	1.0	0.01	1.0	1.0	0.0005	0.01	0.1	2.14×10^6

Table 6. Relative Times of CPU for
Explicit Method for 10^4 years

Scheme	Relative Time of CPU		
	Backward	Centered	Forward
Forward Euler	1	1	1
Backward Euler	0,29	0,37	0,61
Crank-Nicolson	0,29	0,37	0,61

6. CONCLUSIONS

This work has focused on development of a numerical consistency and stability analysis of the equations that describe the migration of the radionuclides. It was demonstrated that all the methods (Explicit, Implicit and Crank-Nicolson) togheter with all possible forms of advective term discretization (Backward, Centered and Forward) are consistent with the original PDE. In the stability analysis, with aid of the von Neumann technique, it was shown that the explicit method, for all configurations of the advective term, and the implicit and Crank-Nicolson methods with forward discretization in the advective term, presented stability conditions to be considered.

In view of the profiles of normalized concentrations in the fracture and in the rock matrix, the results showed a very similar behavior for any method analyzed, mainly for very long times, 10^3 and 10^4 years. With regard to processing time , the implicit and Crank-Nicolson methods, in spite of requiring a larger number of operations for every time step (greater computation expense is required by them), need much smaller total computing time than the explicit method since one can use a significantly larger time step as compared to explicit methods. The implicit schemes have same computing times, due to the same amount of computer operations involved both in totally implicit and Crank-Nicolson methods.

REFERENCES

1. S. H. Li and S. L. Chiou, "Radionuclide Migration in Fractured Porous Rock: Analytical Solution for a Kinetic Solubility-Limited Dissolution Model," *Nuclear Technology*, **104**, pp. 258-271 (1993).
2. E. O. Frind and E. A. Sudicky, "Contaminant Transport in Fractured Porous Media: Analytical Solution for a Single Fracture," *Water Resources Research*, **17**, pp. 555-564 (1981).
3. A.O. Fortuna, *Técnicas Computacionais para Dinâmica dos Fluidos*, Edusp, São Paulo, (2000).
4. C.Hirsch*, Numerical Computation of Internal and External Flows, Volume 1: Fundamentals of Numerical Discretization*, John Wiley & Sons, New York, (1992).
5. D. A. Anderson, J. C. Tannehill, *Computational Fluid Mechanics and Heat Transfer,* Hemisphere, New York, (1984).