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ANALYTICAL STUDY IN 1D NUCLEAR WASTE MIGRATION

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Abstract. *The simulation of the nuclear waste migration phenomena are governed mainly by diffusive–convective equation that includes the effects of hydrodynamic dispersion (mechanical dispersion and molecular diffusion), radioactive decay and chemical interaction. For some special problems (depending on the boundary conditions and when the domain is considered infinite or semi-infinite) an analytical solution may be obtained using classical analytical methods such as Laplace Transform or variable separation. The hybrid Generalized Integral Transform Technique (GITT) is a powerful tool that can be applied to solve diffusive-convective linear problems to obtain formal analytical solutions. The aim of this work is to illustrate that the GITT may be used to obtain an analytical formal solution for the study of migration of radioactive waste in saturated flow porous media. A case test considering* ^{241}*Am radionuclide is presented.*

Key-word: *Radionuclide Migration, Transport Equation, Integral Transform Methods*

1. INTRODUCTION

Health and environmental impacts that may result from disposal of radiocative and non-radioactive, toxic waste are growing concern on the last years for sustainable development of human society. Agenda 21, the action plan prepared as a result of the United Nations Conference on Environment and Development (UNCED) held in Rio de Janeiro in 1992, included three chapters specifically focussed on waste management. Minimizing the amount of waste generated and safe disposal of waste are the main topics to be discussed. A special attention should be given to radioactive waste as well as the heavy metals, because their prolonged risk effects. This article will take into consideration only the radioactive wastes.

The migration phenomena of radionuclides in geosphere may be modeled using the transport equation, e.g., a convective-diffusive partial differential equation. Traditionally this problem has been solved by Laplace Transform or by separation of variables methods, and for some simplified boundary conditions and for infinite or semi-infinite domains, a complete set of analytical solutions can be obtained. These solutions are very important as benchmarking, but of limited utility in the case of a realistic site. When it is very difficult to obtain an analytical inversion of Laplace Transform and when is not possible to applied separation of variables, the problem is usually solved using purely numerical methodology, such as finite differences.

On the other way, hybrid numerical-analytical methods such as the Generalized Integral Transform Method (GITT)have been growing in the last years. In particular, this method presents some important characteristics such as automatic error control and moderate computational cost, even when are analyzed multidimensional problems. A systematized description and application of the GITT method can be found in Cotta, 1993 and Cotta et al. (1997,1998). The main idea of this semi-analytic method, with spectral characteristic, is to represent the field as an eigenexpansion and using the orthogonality property of the eigenfunction to transform the partial differential equation into a coupled ordinary system, e.g., reducing the number of independent variables to only one, in the case of one dimensional problem. The ordinary differential system is then solved numerically, using scientific subroutine libraries with global error control procedure. Therefore, the numerical task is only the solution of the ordinary differential system.

Another interesting aspect of the GITT is that because of the hybrid conception, it is possible to insert properties or analytical strategies for those purely numerical methods would be incapable to adopt. In this sense, the aim of this paper is to show that the analytical structure of the GITT allows to find formal exact solutions for the linear dispersive-diffusive problem of the radioactive waste migration in the soil.

2. PROBLEM DESCRIPTION

The short, medium or long lived radioactive waste such as ^{137}Cs, ^{241}Am, ^{238}U, ^{14}C, etc, originated from industrial, medical and research applications of radionuclides as well as those originating from nuclear plants, should be disposed in a repository specially made for this objective, as represented in Fig. 1.

Usually, as a consequence of the engineering barrier failure, water infiltrates the system and leaches the radioactive waste causing migration to the soil. When the principal flow direction is known and transverse dispersion is small, compared to longitudinal dispersion, the one-dimensional advection-dispersion model is often sufficient to describe the transport (Klukas M.H. and Moltyaner G.L., 1995).

In this work is assumed a saturated and homogeneous media, no radionuclide chain, constant soil properties, adsortion and one-dimensional approach. Therefore, the dimensional mass transport equation (Bear, 1972 and De Wiest, 1972) is given by:

$$R_t \frac{\partial C_1}{\partial t^*} = D_1 \frac{\partial^2 C_1}{\partial Z^2} - V_1 \frac{\partial C_1}{\partial Z} - \lambda_1 R_t C_1 \tag{1.a}$$

and the dimensional boundary conditions:

$$C_1(0,t^*) = F_1^*(t^*) \qquad ; \qquad D_1 \frac{\partial C_1(L_1,t^*)}{\partial Z} + h_1 C_1(L_1,t^*) = 0 \tag{1.b,c}$$

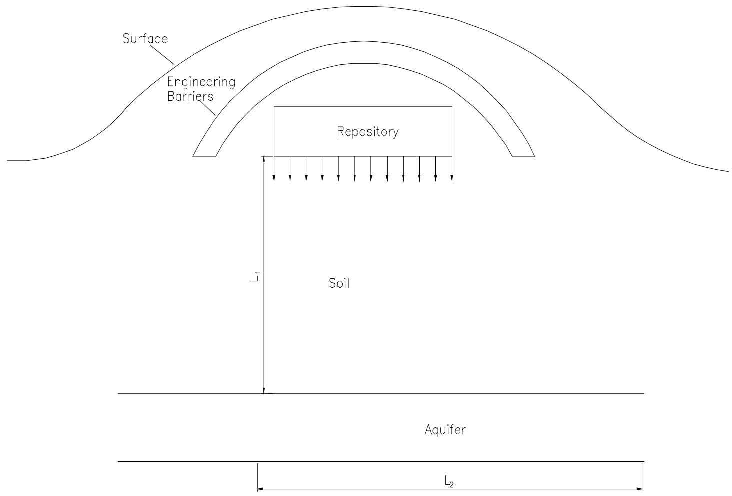

Figure 1. Repository Model

where R_t is the retention factor in the satured media for the radionuclide,
D_1, dispersion coefficient (m^2/y),
λ_1, decay constant (y^{-1}),
V_1, velocity of radionuclide in the water (pore velocity) (m/y),
Z, dimensional coordinate (m),
C_1, concentration (Ci/cm^3, or Bq/cm^3; 1Ci = $3{,}7 \times 10^{10}$ Bq),
t^* dimensional time (y),
$F_1^*(t^*)$, injection function in the soil (Ci/m^3, or Bq/m^3)

The problem is completed considering null concentration in the soil:

$$C_1(Z,0) = 0 \tag{1.d}$$

The problem is turn non-dimensional defining the following parameters:

$$\Delta_1 = \frac{1}{R_t} \quad ; \quad \sigma_1 = \frac{L_1 V_1}{D_1 R_t} \quad ; \quad \gamma_1 = \frac{\lambda_1 L_1^2}{D_1} \tag{3.a-c}$$

$$z = \frac{Z_1}{L_1} \quad ; \quad c_1 = \frac{C_1}{C_0} \quad ; \quad t = \frac{D_1 t^*}{L_1^2} \tag{3.d-f}$$

$$Bi_1 = \frac{h_1 L_1}{D_1} \tag{3.g}$$

Therefore, the non-dimensional version of the problem is given by:

$$\frac{\partial c_1}{\partial t} = \Delta_1 \frac{\partial^2 c}{\partial z^2} - \sigma_1 \frac{\partial c_1}{\partial z} - \gamma_1 c_1 \tag{4.a}$$

with the following boundary conditions:

$$c_1(0,t) = F_1\ (t\ ) = a\ \ e^{-bt}\ ; \qquad \frac{\partial c_1(1,t)}{\partial z} + Bi_1 c_1(1,t) = 0 \tag{4.b,c}$$

The injection function adopted is an exponential decaying type, simulating a first order leaching type of the waste. Finally, the initial condition is given by:

$$c_1(z,0) = 0 \tag{4.d}$$

3. INTEGRAL TRANSFORM SOLUTION

For a better application of the GITT methodology, is convenient to homogenize the boundary conditions, because the boundary conditions of eigenfunctions are homogeneous (Vidakovic Romani, 1996). Then, the concentration field is split in two parts:

$$c_1(z,t) = H_1(z,t) + P_1(z;t) \tag{7}$$

where $H_1(z,t)$ is the unknown function to be calculated.

The filter $P_1(z;t)$ is determined from the solution of the following quasi-permanent problem:

$$\frac{d^2 P_1}{dz^2} - \left(\frac{\sigma_1}{\Delta_1}\right)\frac{dP_1}{dz} - \left(\frac{\gamma_1}{\Delta_1}\right)P_1 = 0 \tag{8.a}$$

$$P_1(0;t) = F_1(t)\ ; \qquad \frac{dP_1(1;t)}{dz} + Bi_1 P_1(1,t) = 0 \tag{8.b,c}$$

The problem above has an analytical solution, given by:

$$P_1(z;t) = \frac{(Bi_1 + dw)e^{dw - sw + sw\ z} - \ e^{dw\ z}(Bi_1 + sw)}{-Bi_1 + (Bi_1 + dw)e^{dw - sw} - sw} F(t) \tag{9.a}$$

$$sw = \frac{R_1 + \sqrt{R_1^2 + 4S_1}}{2}\ ; \qquad dw = \frac{R_1 - \sqrt{R_1^2 + 4S_1}}{2} \tag{9.b-c}$$

$$R_1 = \frac{\sigma_1}{\Delta_1}\ ; \qquad S_1 = \frac{\gamma_1}{\Delta_1} \tag{9.d-e}$$

Problem (4) can then be re-written in terms of H_1 e P_1:

$$\frac{\partial H_1}{\partial t} = \Delta_1 \frac{\partial^2 H_1}{\partial z^2} - \sigma_1 \frac{\partial H_1}{\partial z} - \gamma_1 H_1 - \frac{\partial P_1}{\partial t} \tag{10.a}$$

$$H_1(0,t)=0 \qquad ; \qquad \frac{\partial H_1(1,t)}{\partial z}+Bi_1 H_1(1,t)=0 \tag{10.b,c}$$

$$H_1(z,0)=-P_1(z;0) \tag{10.d}$$

3.1 Eigenvalue Problem and integral transform of partial differential equation

A Sturm-Liouville eigenvalue problem was choose, with the boundary conditions equivalent to equation (10)

$$\frac{d^2\tilde{\varphi}_i}{dz^2}+\beta_i^2\,\tilde{\varphi}_i=0 \tag{11.a}$$

$$\tilde{\varphi}_i(0)=0 \qquad ; \qquad \frac{d\tilde{\varphi}_i(1)}{dz}+Bi_1\tilde{\varphi}_i(1)=0 \tag{11.b,c}$$

The eigenfunction and the transcendental equation for obtain the eigenvalues are given by:

$$\varphi_i=\sin(\beta_i z) \qquad \beta_i=-Bi_1\tan(\beta_i) \tag{12.a,b}$$

The orthogonality property for the eigenfunctions spectrun is given by:

$$\int_0^1 \tilde{\varphi}_i\tilde{\varphi}_j dz=\delta_{ij} \tag{12.c}$$

where δ_{ij} is the delta function from Kronecker, and $\tilde{\varphi}_i$ is the normalized eigenfunction:

$$\tilde{\varphi}_i=\frac{\varphi_i}{N_i^{1/2}} \qquad ; \qquad N_i=\frac{1}{2}\left(\frac{(\beta_i^2+Bi_1^2)+Bi_1}{\beta_i^2+Bi_1^2}\right) \tag{12.d-e}$$

The transform-inverse pair is defined by:

$$\overline{H}_{1i}(t)=\int_0^1\tilde{\varphi}_i(z)\;\; H_1(z,t)\;\; dz \quad \text{(Transform)} \tag{13.a}$$

$$H_1(z,t)=\sum_{i=1}^{\infty}\tilde{\varphi}_i(z)\overline{H}_{1i}(t) \qquad \text{(Inverse)} \tag{13.b}$$

System (10) can now be transformed in a differential system from the application of the operator $\int_0^1\tilde{\varphi}_i[\,]dz$ and considering the transform-inverse pair. Thereby, it results in a coupled ordinary differential system as follows:

$$\frac{d\overline{H}_{1i}}{dt}=\sum_{j=1}^{\infty}A1_{ij}\,\overline{H}_{1j}+g_{1i}(t) \tag{14.a}$$

$$A1_{ij} = -(\Delta_1 \beta_i^2 + \gamma_1)\delta_{ij} - \sigma_1 F1_{ij} \ , \qquad\qquad F1_{ij} = \int_0^1 \tilde{\varphi}_i \frac{d\tilde{\varphi}_j}{dz} dz \qquad (14.b,c)$$

and the source term is calculated analytically from:

$$g_{1i}(t) = -\int_0^1 \tilde{\varphi}_i \ \frac{\partial P_1}{\partial t} dz \qquad (14.d)$$

The integral transformation for the initial conditions gives:

$$\overline{H}_{1_i}(0) = h_{0_i} = -\int_0^1 \tilde{\varphi}_i \, P_1(z;0) dz \qquad (14.e)$$

3.2 Analytical solution of the ordinary differential system

The coupled ordinary differential system (14) present stiff characteristic that is more critic for higher truncation order. Typically in this stage of GITT application, the system will be solved numerically, making use of subroutines with automatic error target, appropriated for stiff cases. If the problem is a linear one, it is possible to use the method of diagonalization about solutions of non-homogeneous linear system (Kreyszig, 1993). Through of this alternative is possible to find formal analytical solution for the problem.

The initial value problem (14) may be represented in a compact way as:

$$\{\overline{H}_1\}' = [A]\{\overline{H}_1\} + \{g_1(t)\} \qquad ; \qquad \{\overline{H}_1(0)\} = \{h_0\} \qquad (15.a,b)$$

where the symbol "{ }" identifies vector and "[ ]" identifies matrix.

Due the problem to be coupled and non-homogeneous, it is appropriate to consider the method of the diagonalization. So, it is considered that $\{H_1(t)\}$ may be represented in terms of the modal eigenvectors matrix, $[X]$,associated to coefficient matrix $[A]$:

$$\{\overline{H}_1(t)\} = [X]\{W(t)\} \qquad (16)$$

Thus, system (15) may be rewritten as:

$$[X]\{W(t)\}' = [A][X]\{W(t)\} + \{g_1(t)\} \qquad (17)$$

or making an inversion matrix, the new decouple system is given by:

$$\{W(t)\}' = [X]^{-1}[A][X]\{W(t)\} + [X]^{-1}\{g_1(t)\} \qquad (18.a)$$

$$\{W_j(0)\} = [X]^{-1}\{h_0\} \qquad (18.b)$$

The first term from the right hand side of eq. (18.a) is a diagonal matrix, whose main diagonal are the eigenvalues λ_j, associated to the matrix $[A]$. It is evident that eq.[18] is an uncoupled ordinary differential system whose analytic solution is given by:

$$W_j(t) = \varsigma_j e^{\lambda_j t} + e^{\lambda_j t}\int e^{-\lambda_j t} h_j(t) \quad dt \tag{19.a}$$

where,

$$\{h\} = [X]^{-1}\{g_1(t)\} \tag{19.b}$$

The integral coefficient a_j is obtained from the initial condition (18.b):

$$\varsigma_j = h_{0\,j} - \left[\int e^{-\lambda_j t} h_j(t) \quad dt\right]_{t=0} \tag{19.c}$$

An analytic explicit solution for $\{W\}$ may be obtained as long as the integral in eq.(19.a) exist. When it is not possible the integral will be calculated numerically. Another alternative in this last case is to solve directly the ordinary differential system (18), using for example Runge Kutta or Adams Bashford's methods, but there is no need to use a more specialized method as the Gear, because the system is not stiff.

When it is considered an exponential injection variable function as given in eq. (4.b), the potential W_j are found to be:

$$W_j(t) = \frac{1}{b+\lambda_j}\left\{b \quad e^{t\lambda_j} r_j + a \quad b(- \quad e^{t\lambda_j} + \quad e^{-b\,t})\sum_{k=1}^{\infty} cte_k \quad \Omega_{jk} + e^{t\lambda_j} r_j \lambda_j\right\} \tag{20.a}$$

Where $[\Omega]=[X]^{-1}$ (20.b)

The vector $\{cte\}$ was determined using the symbolic manipulator MATHEMATICA (1996), whose expression in FORTRAN language is:

cteng1(k)=betk**3*bi1 + betk**3*sw + betk*bi1*sw**2 + betk*sw**3 +

- betk**3*dw*dcos(betk)*dexp(dw) +
- betk*bi1*dw**2*dcos(betk)*dexp(dw) +
- betk*dw**3*dcos(betk)*dexp(dw) -betk**3*sw*dcos(betk)*dexp(dw) -
- betk*bi1*sw**2*dcos(betk)*dexp(dw) -
- betk*sw**3*dcos(betk)*dexp(dw) -
- betk**3*bi1*dexp(dw - sw) - betk**3*dw*dexp(dw - sw) -
- betk*bi1*dw**2*dexp(dw - sw) - betk*dw**3*dexp(dw - sw) +
- betk**2*bi1*dw*dexp(dw)*dsin(betk) -
- betk**2*bi1*sw*dexp(dw)*dsin(betk) -
- bi1*dw**2*sw*dexp(dw)*dsin(betk) -
- dw**3*sw*dexp(dw)*dsin(betk) +
- bi1*dw*sw**2*dexp(dw)*dsin(betk) + dw*sw**3*dexp(dw)*dsin(betk) (21.a)

deng1k(k)=xnork*(-(betk**4*bi1) - betk**2*bi1*dw**2 -

- betk**4*sw - betk**2*dw**2*sw - betk**2*bi1*sw**2 -
- bi1*dw**2*sw**2 - betk**2*sw**3 - dw**2*sw**3 +
- betk**4*bi1*dexp(dw - sw) + betk**4*dw*dexp(dw - sw) +
- betk**2*bi1*dw**2*dexp(dw - sw) +
- betk**2*dw**3*dexp(dw - sw) +
- betk**2*bi1*sw**2*dexp(dw - sw) +

- betk**2*dw*sw**2*dexp(dw - sw) +
- bi1*dw**2*sw**2*dexp(dw - sw) +
- dw**3*sw**2*dexp(dw - sw)) (21.b)

cte(k)=cteng1/deng1k (21.c)

where the FORTRAN symbols are defined by: betk=β_k, Bi1=Bi_1 , dcos( )=cos( ), dsin( )=sin( ), dexp( )=Exp( ), xnork=N_k

4. RESULTS

The migration of ^{241}Am radionuclide was chosen as test case. This radionuclide present a radiotoxicity very high and was used in the past in lighting conductors, its also being used in smoke detectors and in some industrial gauges (SINAER, 1996).

The following parameter values were used in the simulation case test:

- Initial concentration (Bq/cm^3)=1.233
- Darcy velocity (m/y)=4
- porosity=0.2
- Distribution Coefficient (m3/kg)= 20.d-3
- Molecular Diffusion (m2/ano)=3153.6d-6
- Dispersivity (m)=0.03
- Half Life (y)=432
- Bulk Density meio 1 (kg/m3)=1800
- L_1 (m)=50
- Bi_1=20

Table 1. Convergence behavior of the concentration field

(a) Time=50 years

N	*Z*=1m	*Z*=2m	*Z*=4m	*Z*=6m
100	1,222680	1,206631	1,168250	2,587912E-1
200	1,214636	1,197787	1,160527	2,524101E-1
300	1,213260	1,196326	1,159202	2,513791E-1
400	1,212954	1,196004	1,158920	2,510562E-1
500	1,212873	1,195920	1,158852	2,509255E-1
600	1,212848	1,195894	1,158833	2,508633E-1
700	1,212840	1,195885	1,158829	2,508302E-1

(b) Time=200 years

N	*Z*=5m	*Z*=10m	*Z*=15m	*Z*=20m
100	1,149231	1,071460	9,987232E-1	9,001519E-1
200	1,140082	1,062635	9,904164E-1	8,940148E-1
300	1,138604	1,061221	9,890910E-1	8,929815E-1
400	1,138281	1,060914	9,888031E-1	8,927645E-1
500	1,138196	1,060833	9,887281E-1	8,927128E-1
600	1,138171	1,060809	9,887054E-1	8,926998E-1
700	1,138162	1,060801	9,886976E-1	8,926967E-1

(c) Time=1000 years (continuation)

N	*Z*=10m	*Z*=20m	*Z*=30m	*Z*=40m
100	1,020214	8,862712E-1	7,698889E-1	6,687968E-1
200	1,020580	8,855578E-1	7,701352E-1	6,690027E-1
300	1,020617	8,865886E-1	7,701614E-1	6,690240E-1
400	1,020624	8,865947E-1	7,701666E-1	6,690280E-1
500	1,020626	8,865963E-1	7,701679E-1	6,690290E-1
600	1,020626	8,865968E-1	7,701682E-1	6,690292E-1
700	1,020626	8,865969E-1	7,701684E-1	6,690293E-1

For Computational purposes the associated eigenvalue and eigenvetor of the matrix [A] were determined using the subroutine EVCRG of the IMSL library (1995).

The first step was to determine the behavior of the convergence for different times and various positions inside the soil. The results are shown in table 1.

The convergence with six decimal digits was allowed with N=700 terms in the series in some case . It is evident that the concentration values were converged with three or more digits with N=300 terms in the series. The computer processing time is about 15 sec. when used a Pentium II-266 as platform. It is clear that the numeric effort exists only to obtain the eigenvectors and eigenvalues of the matrix [A1] (14). So the CPU time are showed in table 2.

Table 2. Computational Cost

N	CPU time (Sec)
100	0,6
200	4
300	15
400	42
500	82
600	148
700	236

Accurately values of the concentration for this one-dimensional approach may be obtained very fast with the present alternative.

Also, it is important to remark that the present analytic solution is exact for any truncated system. A full numeric solution of truncated ordinary differential equation (15) would be very expensive computationally if compared with the present approach.

Figure 2 illustrates the typical advance of radioactive waste when is considered a decay exponential injection. In this case the level of concentration is reduced along the time as expected.

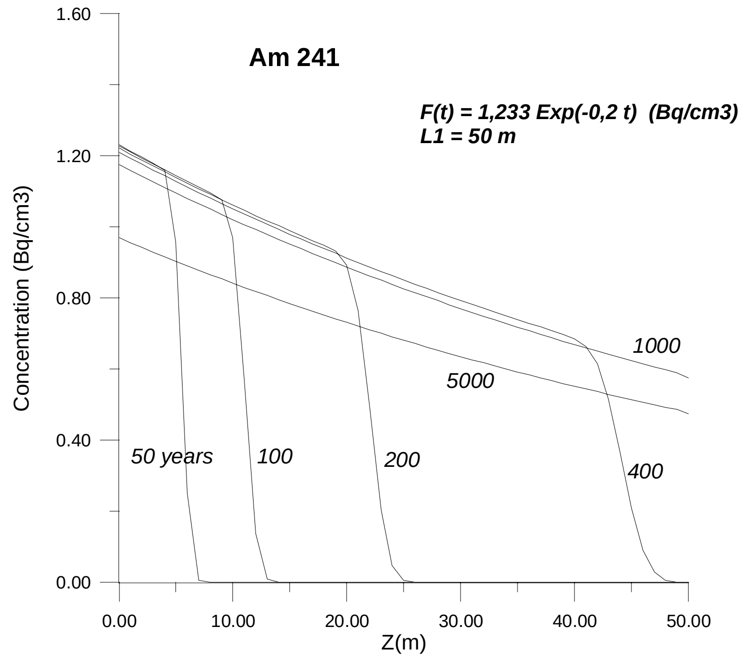

Figure 2. Concentration in the soil along the time

5. CONCLUSION

The present work showed that when the generalized integral transform technique, is used, it is possible to get formal analytical solutions in linear problems not transformable using the method of the diagonalization. The computational cost is minimized and the results may be used for benchmark or engineering purposes.

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