Applicability of Wavelet Galerkin Method for Solving High Level Radioactive Waste Transport Model

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Abstract: - The relative merits of the wavelet-Galerkin solution of the nonlinear system of partial differential equations arising from a model formulation of migration of high-level radioactive waste (HLW) are quantitatively and qualitatively analyzed. Wavelet functions are generated by dilation and translation on a scaling function. The wavelet functions are localized in space and compactly supported, so these properties can be utilized to solve differential equations that have severe "**stiff**". A mathematical model for predicting the nuclide migration of (HLW) was formulated and compared with the results from the reference case. The model, which is wavelet-discretized model, is devised to be very reasonable and accurate by proper selection of wavelet order and dilation order pair.

Keywords: - geological disposal, wavelet Galerkin method, compactly supported, orthogonality, connection coefficients, repository system.

1. Introduction

When discussing geological disposal in Japan, it is necessary to consider the specific geological conditions of the archipelago, which is located in a tectonically active zone. As a result, Japan has a high frequency of earthquakes, fault movement and volcanic activity. Thus, the Engineered Barrier System (EBS) is selected with a high performance margin, which reduces the requirements on the barrier functions of the geosphere. A detailed calculation is required to analyze the system showing that such natural phenomena will not affect the safety of geological disposal. The complexity of this situation makes it difficult to obtain analytical solutions for the radionuclide transport in the near field of the repository. Standard numerical methods require a detailed discretization at the interfaces between the different regions where a step radionuclide concentrations gradient exists. Therefore, nuclide release calculations would be very time consuming. Recent developments in wavelet techniques [1] have made the wavelet Galerkin procedure a viable option for the numerical solution of ordinary and partial equations with encouraging results [2,3,4].

Newly developed techniques for nuclide transport calculations uses the advantage of the wavelet discretization with a proper selection of wavelet order and dilation order pair of the system of equations that describe the nuclide migration. Radioactive decay/ingrowth, nuclide sorption in the solid, and precipitation are included in the model. The discretization of the geometrical system is very coarse and rather simple. Since the scaling functions are compactly supported, only a finite number of the connection coefficients are nonzero [1]. The resultant matrix has a block diagonal structure, which can be inverted easily using any iterative technique, which gives reasonable time, compared to conventional methods.

This paper will show the applicability of wavelet Galerkin discretization to calculate the nuclide release from the nuclear waste repository based on Japanese concept. The calculations are made for various nuclides. Also, it presents a useful tool to calculate the transport of radionuclides in the near filed of the repository taking into account all the possible phenomena as diffusion, sorption and precipitation.

2. Problem Formulation

2.1 Engineered Barrier System Description

The EBS is designed with a high performance migration to cover the wide range of geological conditions found in Japan [5]. The EBS consists of the vitrified High Level radioactive Waste (HLW), a rigid vessel (overpack) for containment of the vitrified waste and a buffer that fills the gaps between the overpack and the surrounding rock mass, as shown in Figure 1.

2.2 Engineered Barrier System Modeling

The mathematical model for nuclide migration in EBS as shown in Fig. 1, consists of a series of equations describing various processes related to:

- Dissolution of vitrified waste to determine the inner boundary condition, dissolution of nuclides to a hypothetical region in the vicinity of the vitrified waste, precipitation/dissolution, decay/ingrowth and release to the buffer material
- Nuclide diffusion in the buffer material, sorption onto the buffer material, precipitation/dissolution and decay/ingrowth.
- Instantaneous mixing of nuclides reaching the boundary between the buffer and host rock. Ground water flow through the EDZ, and nuclide release from the EDZ to the surrounding host rock.

The governing equations are formulated in terms of the following variables: concentration of dissolved nuclide (C_{ij}), concentration of sorbed nuclide (S_{ij}) and concentration of precipitate nuclide (P_{ij}), where "ij" represents isotope j of element i. The four regions considered are the vitrified waste (*G*), a hypothetical water-filled region around the vitrified waste (*R*), the buffer material (*B*) and the Excavation Disturbed Zone EDZ (*M*).

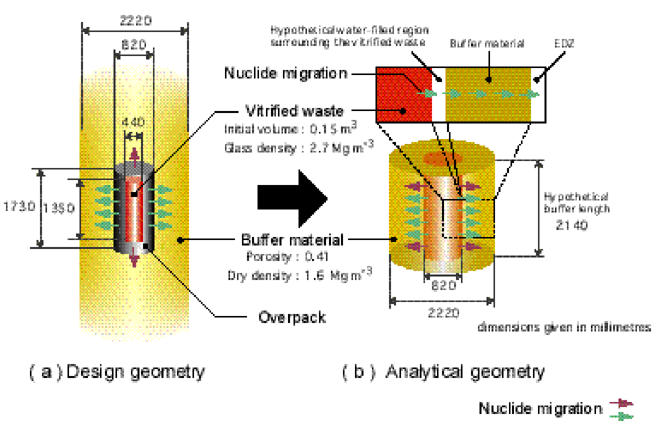

Fig. 1. Component and Geometry of EBS

Dissolution of glass vitrified waste, which, expressed in terms of long-term glass dissolution rate. The concentration in vicinity of the vitrified waste are described by the mass balance equation as follows:

$$V^R \frac{\partial A_{ij}^R}{\partial t} = 2pr_{in} Le^B D_{Pi} \frac{\partial C_{ij}^B}{\partial r}\Big|_{r=r_{in}} + M_{ij}^G g_{Si} - V^R l_{ij} A_{ij}^R + V^R l_{IJ} A_{IJ}^R \quad (1)$$

where V^R volume of the hypothetical water-filled region [m^3], M_{ij}^G inventory of isotope *j* of element *i* in the vitrified waste [mole], g_{si} fractional rate of decrease of the glass volume [y^{-1}], D_{pi} diffusion coefficient in the porewater [$m^2 y^{-1}$], e^B porosity of the buffer material, r_{in} inner radius of the buffer material [*m*], l_{ij} nuclide decay constant [y^{-1}]. The total amount of nuclide in a unit volume of the hypothetical water-filled region around the vitrified waste A_{ij}^R and the fractional rate of decrease of the glass volume are given by the following equations:

$$A_{ij}^R = C_{ij}^R + P_{ij}^R$$

$$g_{si} = \frac{a^G}{r^G V^G} k$$

where, a^G surface area of the vitrified waste [m^2], r^G density of the vitrified waste [kg/m^3], V^G volume of the vitrified waste [m^3], and *k* glass dissolution rate [$kg/m^2 y$].

Nuclide migration in the buffer material region, which represents nuclide diffusion, sorption, precipitation and decay/ingrowth, is expressed:

$$\frac{\partial A_{ij}^B}{\partial t} = e^B D_{pi} \frac{1}{r}\frac{\partial}{\partial r}\left(r \frac{\partial C_{ij}^B}{\partial r}\right) - l_{ij} A_{ij}^B + l_{IJ} A_{IJ}^B \quad (2)$$

where A_{ij}^B represents the total amount of nuclide in a unit volume of buffer material , which can be expressed as follows:

$$A_{ij}^B = e^B C_{ij}^B + (1 - e^B) S_{ij}^B + P_{ij}^B$$

The concentration of sorbed nuclide in a unit volume of the solid part of the buffer material S_{ij}^B is given by,

$$S_{ij}^B = r^B kd_i C_{ij}^B,$$

where r^B is the density of the buffer material, and kd_i is the distribution coefficient of the element $i[m^3/kg]$. Precipitation occurs when

$$e^B C_i^* + (1 - e^B) r^B kd_i C_i^* < \sum_j A_{ij}$$

where C_i^* is the elemental solubility $[mol/m^3]$. The concentration of precipitate in a unit volume of the buffer material, P_{ij}^B, is derived by,

$$P_{ij}^B = A_{ij}^B - (e^B C_i^* + (1 - e^B) r^B kd_i C_i^*) \frac{A_{ij}^B}{\sum_j A_{ij}^B}$$

where the concentration of dissolved nuclides in a unit volume of porewater in the buffer material , C_{ij}^{B} , are given by,

$$C_{ij}^{B} = C_i^{*} \frac{A_{ij}^{B}}{\sum_j A_{ij}^{B}}$$

This equation accounts for the limited elemental solubility (C_i^{*}), which is partitioned among stable and radioactive isotopes.

The nuclides reaching the boundary between buffer and host rock are mixed instantaneously with the ground water flowing through the Excavation Distributed Zone (EDZ). The nuclide concentrations in the EDZ region are described by the following equation, which includes solubility limits with precipitation/dissolution, diffusion from the buffer, sorption and radioactive decay/ingrowth:

$$V^{M} \frac{\partial A_{ij}^{M}}{\partial t} = -2pr_{out} Le^{B} D_{pi} \left. \frac{\partial C_{ij}^{B}}{\partial r} \right|_{r=r_{out}} - V^{M} l_{ij} A_{ij}^{M} + V^{M} l_{IJ} A_{IJ}^{M} - QC_{ij}^{M} \quad (3)$$

where V^{M} is the volume of the EDZ [m^3], Q is the groundwater flow rate [m^3 / y], and A_{ij}^{M} represents the total amount of nuclide in a unit volume of EDZ.

Nuclide migration in the EBS is evaluated by solving the above equations and the release rate of nuclide into the surrounding host rock $f_{ij}^{buffer \to hostrock}$ is derived by,

$$f_{ij}^{buffer \to hostrock} = QC_{ij}^{M}$$

Application of the leap-frog scheme to equations (1), (2), and (3) yields the semi-discrete system

$$[A_{ij}^{R}]^{n+1} = [A_{ij}^{R}]^{n-1} + \frac{2\Delta t}{V^{R}} \left[2pr_{in} Le^{B} D_{Pi} (C_{ij}^{B})_r \Big|_{r=r_{in}} + M_{ij}^{G} g_{Si} - V^{R} l_{ij} A_{ij}^{R} + V^{R} l_{IJ} A_{ij}^{R} \right]^{n} \quad (4)$$

$$[A_{ij}^{B}]^{n+1} = [A_{ij}^{B}]^{n-1} + 2\Delta t \left[e^{B} D_{Pi} (C_{ij}^{B})_{rr} + \frac{e^{B} D_{Pi}}{r} (C_{ij}^{B})_r - l_{ij} A_{ij}^{B} + l_{IJ} A_{IJ}^{B} \right]^{n} \quad (5)$$

$$[A_{ij}^{M}]^{n+1} = [A_{ij}^{M}]^{n-1} - \frac{2\Delta t}{V^{M}} \left[2pr_{out} Le^{B} D_{Pi} (C_{ij}^{B})_r \Big|_{r=r_{out}} + V^{M} l_{ij} A_{ij}^{M} - V^{M} l_{IJ} A_{ij}^{M} + QC_{ij}^{M} \right]^{n} \quad (6)$$

2.3 Wavelet Galerkin Discretization

Our Galerkin procedure uses a class of compactly supported scaling functions introduced by Daubechies [6]. The scaling function are determined by a wavelet order $2N$ and a set of scaling parameters $\{c_k : 0 \le k \le 2N\}$ that define a generator function $f(x)$ through the scaling relation

$$f(x) = \sum_{k=0}^{N-1} a_k f(2x - k)$$

For each $0 \le j$ we set

$$f_k^{j}(x) = 2^{j/2} f(2^{j} x - k), \text{ for } 0 \le k \le 2^{j}$$

If one sets $V^{j} = span\{f_k^{j} : 0 \le k \le 2^{j}\}$, it is shown that $\{f_k\}$ can be periodized and made to form an orthonormal basis for $V^{j} \in L^2[0,1]$, with $\overline{\bigcup V^{j}} = L^2[0,1]$ and $\bigcap V^{j} = 0$. Moreover the subspaces V^{j} are nested, so that $V^{j} \subset V^{j+1}$. If one lets W^{j} denote the orthogonal complement of V^{j} in V^{j+1}, it is shown that W^{j} is spanned by an orthonormal set of wavelet functions $y_k^{j} = 2^{j/2} y(2^{j} x - k)$, where the generator wavelet $y(x)$ defined by

$$y(x) = \sum_{k=-1}^{N-2} (-1)^k c_{k+1} f(2x + k)$$

The base generators $f(x)$ and $y(x)$ have support $[0, N-1]$ and every polynomial of degree $K \le N/2$ lies in the space V^0, which is equivalent to $y(x)$ having N/2 vanishing moments. The Daubechies class is distinguished by having this interpolation property and the smallest possible support. Thus, from interpolation property, we see that $f(x)$ has at least N/2 continuous derivatives.

Consider a set $\{f_k^{p}\}$ that spans the space $V^{p}[0,1] \subset L^2[0,1]$. A multiresolution is effected by noting that the space $V^{p} \supset V^{p-1} ..... \supset V^1 \supset V^0$. For the Galerkin approximation of the problem, the field variables are projected into the space of trail functions belonging to V^{p} . When we use test functions from the same space, a system of differential equations in time for the coefficients of the field variable results when the inner products $\langle .,. \rangle$ are evaluated and orthogonality among the elements of V^{p} is used. In this work the evolution equations are solved at scale p determined by the resolution of the space V^{p} . If, at any time a multiresolution is desired, this can be performed as a post-processing step or as an adjunct calculation.

In what follows, we project the semi-discrete real variable to V^{p} so that

$$f^{n}(x_j) = \sum_{l=0}^{N-1} f_l^{n} f_l(x_j)$$

The weak formulation of the semi-discrete system is obtained by substituting Equation (4) into Equations (4), (5), and (6) multiplying by the test function $f_k \in V^{p}$, and integrating:

$$\left\langle [A_{ij}^{R}]^{n+1}, f_k \right\rangle = \left\langle [A_{ij}^{R}]^{n-1}, f_k \right\rangle + 2\Delta t \begin{bmatrix} \frac{a_{in} D_e}{V^{R}} \left\langle [C_{ij}^{B}]_r^{n} \Big|_{r=r_{in}}, f_k \right\rangle + \frac{g_{si}}{V^{R}} \left\langle [M_{ij}^{G}]^{n}, f_k \right\rangle \\ - l_{ij} \left\langle [A_{ij}^{R}]^{n}, f_k \right\rangle + l_{IJ} \left\langle [A_{IJ}^{R}]^{n}, f_k \right\rangle \end{bmatrix} \quad (7)$$

$$\left\langle \left[A_{ij}^{B}\right]^{n+1}, f_k \right\rangle = \left\langle \left[A_{ij}^{B}\right]^{n-1}, f_k \right\rangle + 2\Delta t \begin{bmatrix} D_e \left\langle \left[C_{ij}^{B}\right]_{rr}^{n}, f_k \right\rangle + \dfrac{D_e}{r} \left\langle \left[C_{ij}^{B}\right]_{r}^{n}, f_k \right\rangle - \\ l_{ij} \left\langle \left[A_{ij}^{B}\right]^{n}, f_k \right\rangle + l_{IJ} \left\langle \left[A_{IJ}^{B}\right]^{n}, f_k \right\rangle \end{bmatrix} \quad (8)$$

$$\left\langle \left[A_{ij}^{M}\right]^{n+1}, f_k \right\rangle = \left\langle \left[A_{ij}^{M}\right]^{n-1}, f_k \right\rangle - 2\Delta t \begin{bmatrix} \dfrac{a_{out} D_e}{V^M} \left\langle \left[C_{ij}^{B}\right]_{r}^{n}\Big|_{r=r_{in}}, f_k \right\rangle + \dfrac{Q}{V^M} \left\langle \left[C_{ij}^{M}\right]^{n}, f_k \right\rangle - \\ l_{ij} \left\langle \left[A_{ij}^{M}\right]^{n}, f_k \right\rangle + l_{IJ} \left\langle \left[A_{IJ}^{M}\right]^{n}, f_k \right\rangle \end{bmatrix} \quad (9)$$

Following the convention in [1], we refer to the inner products as connection coefficients:

$$\Omega_{k,l}^{0,1} = \left\langle f_k, f_l{}' \right\rangle$$

$$\Omega_{k,l}^{1,1} = \left\langle f_k{}', f_l{}' \right\rangle$$

$$\Omega_{k,j,l}^{0,1,1} = \left\langle f_k, f_j{}' f_l{}' \right\rangle$$

$$\Omega_{k,j,l}^{1,0,0} = \left\langle f_k^{/}, f_j f_l \right\rangle$$

$$\Omega_{k,j,l}^{1,0,1} = \left\langle f_k^{/}, f_j f_l^{/} \right\rangle$$

The most expedient strategy available for the evaluation of these connection coefficients is given in [1]. The connection coefficients should be precomputed. The resulting tables are then read in the time marching procedure.

The dimension of the matrix T in the heterogeneous case is $2\times(I(2^j + N) - I + 1)$, I is the number of nodes. Because the scaling function is compact supported, only a finite number Ω are nonzero. Then, the resultant matrix T becomes $2*(I(N-2)+1)$. The matrix T has the block diagonal structure. Such matrix can be inverted easily using any efficient matrix inversion method.

3. Problem Solution

The radionuclides to be considered in the nuclide migration analysis were selected by the following procedure:

- Nuclide whose ratio of calculated concentration in well water to maximum permissible concentration in the water is greater than 10^{-3} are include in the safety assessment
- Daughter nuclides with half-lives of less than one year are excluded from the nuclide migration calculation on the assumption that they are in the equilibrium with their parent nuclides.

Table 1, shows the safety relevant radionuclides considered in the analysis of nuclide migration in the Engineered Barrier System [7].

To give a picture of the capability of WGM to calculate the radionuclide transport, release calculations for several fission nuclides and radioactive chains are discussed. Results obtained running Sn-126 isotope case are shown in Fig. 2 for several wavelet-dilation orders pair. At long times, a good accuracy is found regardless of the choice of wavelet-dilation order pair. The choice of wavelet-order pair is only important at early time. Better accuracy is obtained by increasing wavelet order and dilation order. The results from our WGM and MESHNOTE a finite difference code [8] are compared. The comparison shows a well agreement between both of them. The running time using WGM was less than the one using FD code with fine discretization. The wavelet-based system is fast compared to the detailed finite difference code. Computational time for all decay chains takes 2.54/13000. Min. for each time step compared to 20/83 Min. for detailed finite difference MAYCIES code [9].

Nuclides released from the waste form precipitate when their concentrations in the porewater result in their elemental solubility limits the release rates to the surrounding rock as shown in Fig. 3 for Np-237 isotope.

The following observations are made regarding the time-dependent nuclide release rates from the EBS:

During the period of 5×10^5 years following the overpack failure, Cs-135 has the highest release rate for any nuclide, in terms of Bq per year as shown in Fig. 4. This is due to its high solubility, relatively small distribution coefficient and relatively long half-life. After $7.\times10^4$ years, the release rate of Cs-135 decreases sharply due to the glass having completely dissolved by this time. Nb-93m shows the highest release rate after 5×10^5 years. Nb-93m is in radioactive equilibrium with its parent (Zr-93), due to its relatively short (several decades) half-life. Nb-93m is mobile in the EBS due to its relatively low distribution coefficient and its release rate is higher than that of Zr-93. Precipitation of a nuclide in the decay series under radioactive equilibrium may result in a release rate for the nuclide different from that of its parent and daughters (e.g. Pb-210, Ra-226 and Th-230) as well as a difference in sorption behavior as shown in Figures, 5, 6, 7, and 8. Nuclides with large inventories and low solubility, e.g. Np-237, and Tc-99, precipitate in the vicinity of the vitrified waste. This results in a low and pseudo-steady-state release rate for a prolonged period. Relatively short-lived (half-life shorter than several tens of thousands of years) of highly sportive elements, e.g. Pu-240 and Am-241 decay significantly within the buffer material and their peak release rate extremely small.

Table 1. Safety-relevant Radionuclides

Fission Products	Sm-151, Cs-135, Sn-126, Pd-107, Tc-99, Nb-94, Zr-93→ Nb-93m, Se-79
4N Series	Pu-240→U-236→Th-232
4N+1 Series	Cm-245→Pu-241→Am-241→Np-237→U-233 →Th-229
4N+2 Series	Cm-246→Pu-242→U-238→U-234→Th-230 →Ra-226→Pb-210
4N+3 Series	Am-243→Pu-239→U-235→Pa-231 →Ac-227

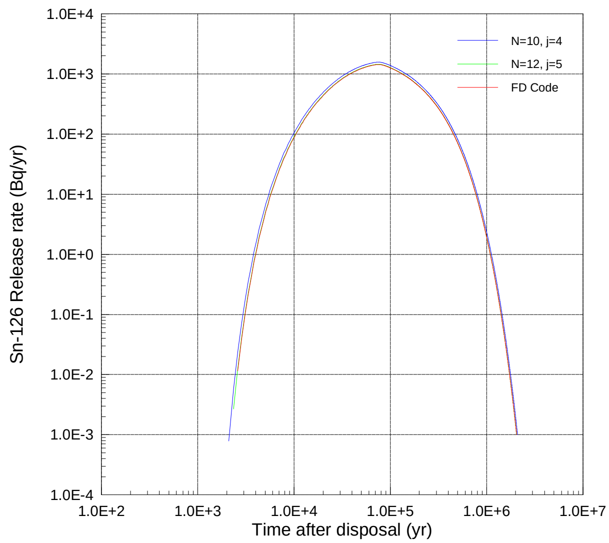

Fig. 2 Sn-126 release rate using various wavelet orders-dilation orders pairs.

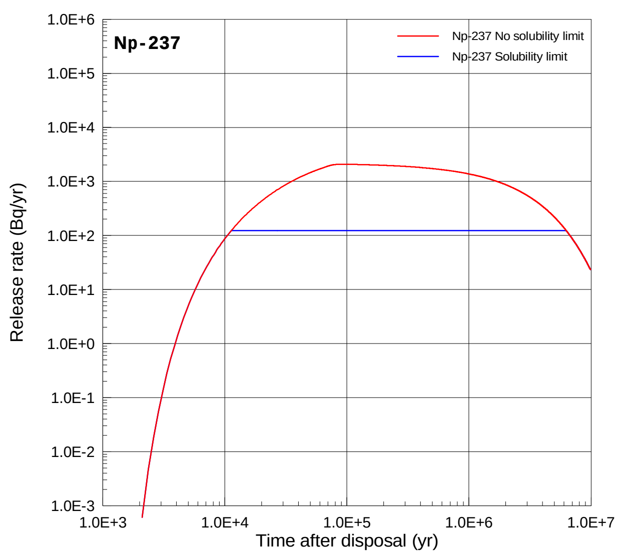

Fig. 3 Np-237 Precipitation calculation and its effect on nuclide release

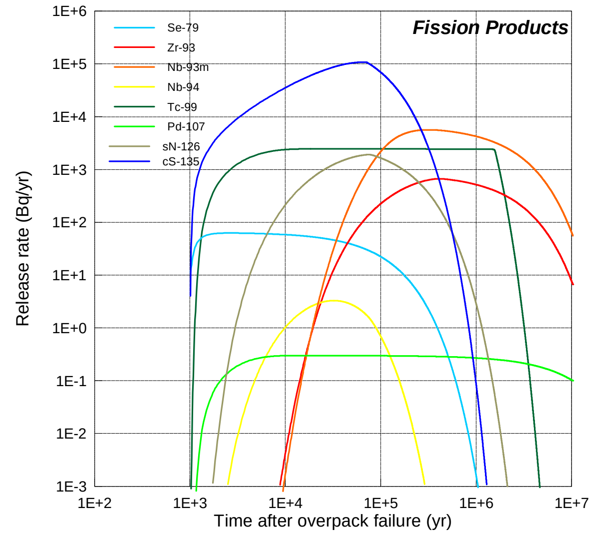

Fig. 4 Release rate of fission products

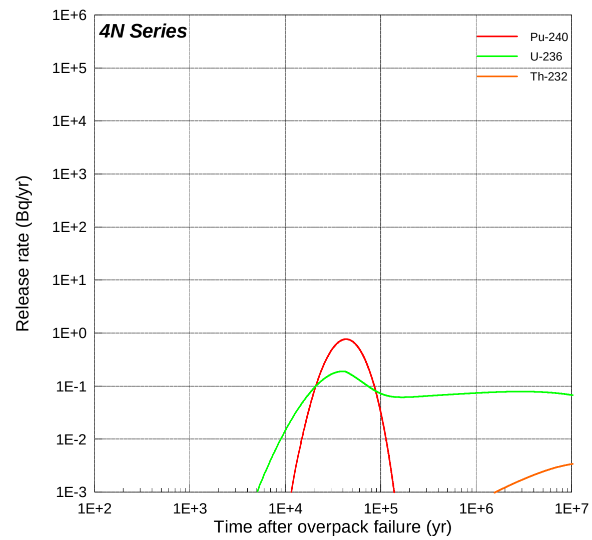

Fig. 5 Release rate of 4N series

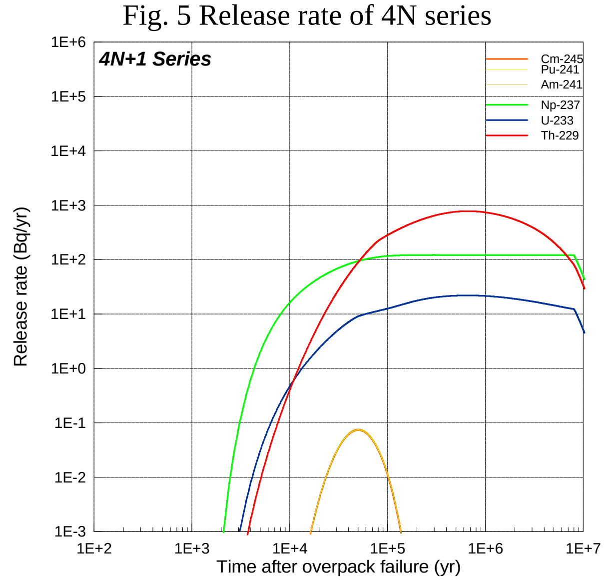

Fig. 6 Release rate of 4N+1 series

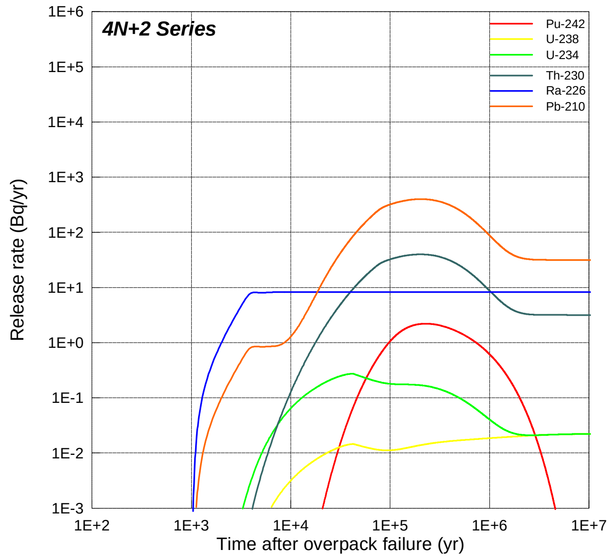

Fig. 7. Release rate of 4N+2 series

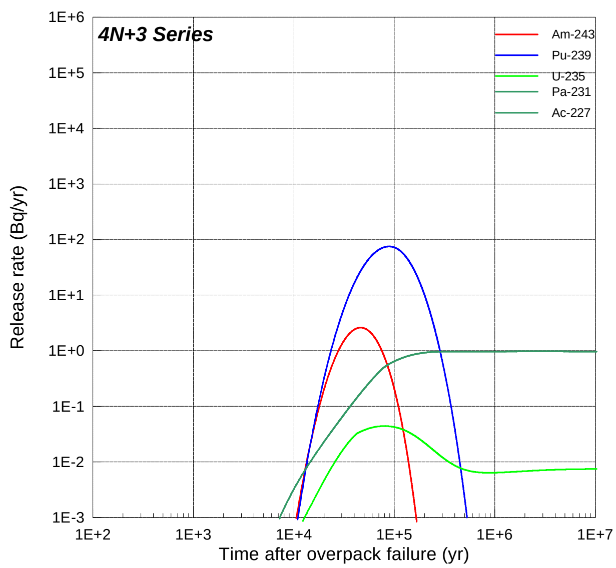

Fig. 8 Release rate of 4N+3 series

4. Conclusion

The capability of WGM to calculate radionuclide transport was shown by applying the model to the very complex transport situation at the Japanese repository.

The wavelet Galerkin method has been shown to be a powerful tool for the fast and accurate solution of the nonlinear system of partial differential equations arising from a model formulation of migration of high-level radioactive waste (HLW). A large Daubechies order gives more smoothness of the scaling function, and a large dilation order provides finer solution in the node, so wavelet function has strong ability to treat such a kind of stiff problems.

In order to verify our approach, we compared the results obtained using WGM with the results obtained by other codes. The results were very accurate and matched well with the results obtained with the conventional methods like finite difference (MESHNOTE and MAYCIES). The accuracy improves as the dilation order j increase for a Daubechies order N chosen large enough to ensure wavelet function smoothness. The selection of a proper value of dilation order j becomes very important especially near the boundaries for a highly soluble element like Cs-135.

Finally, this technique gives us the opportunity to overcome many problems encountered. The analytic solution of the connection coefficients makes the system with any Daubechies' order N and dilation order j unconditionally stable. The computation time is reasonably good compared to the other numerical methods for two reasons. First, those connection coefficients are computed once and stored for later use. Second, the matrix arising from the WGM can be inverted easily.

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