BLT: A SOURCE TERM COMPUTER CODE FOR LOW-LEVEL WASTE SHALLOW LAND BURIAL*

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ABSTRACT

We developed a source term model for low-level waste shallow land burial facilities by separating the problem into four individual compartments: 1) water flow, 2) corrosion and subsequent breaching of containers, 3) leaching of the waste forms, and 4) solute transport. For the first and the last compartments, we adopted the existing codes, FEMWATER and FEMWASTE, respectively. We wrote two new modules for the other two compartments in the form of two separate Fortran subroutines - BREACH and LEACH. They were incorporated into a modified version of the transport code FEMWASTE. The resultant code, which contains all three modules of container breaching, waste form leaching, and solute transport, was renamed BLT (for Breach, Leach, and Transport). This paper summarizes the overall program structure and logistics, and presents two examples from the results of verification and sensitivity tests.

INTRODUCTION

In assessing the performance of low-level waste shallow land burial facilities, the source term for transport calculation is governed by the rate of radionuclide release from the waste forms into the surrounding soil in the vadose zone. We developed a quantitative model to predict the source term applicable under a range of conditions. This model consists of four individual compartments: 1) water flow through the structure (typically a trench), 2) corrosion and subsequent breaching of waste containers, 3) leaching of radionuclides from the waste form, and 4) solute transport to the boundary of the structure. The bases for modeling these four processes were discussed in detail in our earlier report (1), and summarized in one of our previous papers (2). Further developments of this model are described in our recent report (3), and in a separate paper presented at this meeting (4). This paper describes the computer implementation of the source term model by developing a Fortran computer code called BLT (which stands for Breach, Leach, and Transport).

COMPUTER CODE DEVELOPMENT

Computer code development is the most essential task of numerical modeling. For the two compartments of water flow and solute transport, the existing finite-element two-dimensional codes FEMWATER and FEMWASTE (5,6) had been already tested, adopted, and modified, where necessary. Our main effort was focused subsequently on developing subroutines for the remaining two of the four compartments, namely, the container breaching (corrosion) and the waste form leaching compartments.

The implementation of the remaining two model compartments was accomplished by writing two additional Fortran subroutines, one called BREACH and the other, LEACH. The BREACH subroutine calculates the time and the amount (in terms of breached area) of container corrosion based on parameters input by the user, and also the moisture content of the soil first calculated by FEMWATER. A number of default values were also built into the code, so that users can omit many input values when no better numbers are available. The LEACH subroutine calculates the radionuclide release rate from the waste using the waste form parameters provided by the user, hence, the released amount at a given time (time-step). The released amount is assumed zero, if container breaching has not occurred. These two subroutines were directly incorporated into a modified version of FEMWASTE, so that the computed released amounts feed back directly into the solute transport portion of the code as time dependent sources. Accordingly, a number of original subroutines in FEMWASTE were also modified. This newly modified and extended code, which has the capabilities of handling container breaching, waste form leaching as well as solute transport, is therefore abbreviated as BLT to distinguish it from the original FEMWASTE code.

Overall Program Structure of BLT

The original FEMWASTE code consists of a number of subroutines and the main program. The main program in FEMWASTE does very little other than dimensioning arrays and defining common blocks. Once the initialization is done, the main program passes control to the "general manager" subroutine called GM2DXZ, which handles all the logistics of the simulation program. GM2DXZ in turn calls the other subroutines directly or indirectly to carry out more specific or repetitive tasks, for example, input/output chores, solving the global matrix, and calculating mass fluxes at a specific time. Time is incremented by one time-step with each pass of the time loop in this subroutine.

The call structure among the subroutines in FEMWASTE had been described by Yeh (6). The extended

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subroutines in BLT, namely, BREACH, LEACH and TRACE, are all called directly from the subroutine GM2DXZ. The TRACE subroutine is an output routine which keeps records of concentration and/or flux as a function of time for any input specified nodes.

The BREACH and LEACH subroutines are called inside the time loop of GM2DXZ. Figure 1 shows a portion of the time loop within subroutine GM2DXZ, where time dependent element sources/sinks and other boundary conditions are determined.

In the original version of FEMWASTE, GM2DXZ obtains source/sink values, if any, by calling subroutine INTERP at every time step, i.e., every pass of the time loop, during transient simulation. INTERP calculates the values by interpolation in time between data points according to the input table specified by the user. The other boundary conditions, e.g. Dirichlet and Neumann boundary conditions, are also handled in a similar way.

In BLT, an alternate path was created in lieu of table look-up for the source/sink term (Fig. 1). When waste packages, represented by elements, are specified by the input, GM2DXZ calls subroutines BREACH and LEACH in sequence. LEACH then returns a source term for each breached waste container element for that particular time-step. These "waste sources" are subsequently manipulated in a similar way as the element source/sink values from table look-up.

Subroutine BREACH

The main purpose of subroutine BREACH is to compute the area of waste container that is breached based on the general corrosion rate and the pitting corrosion rate at a given time-step. The dependence of these rates had been discussed in detail in one of our previous reports (1).

In BLT, waste packages are represented by elements within the finite element scheme. Each waste element is assigned a specific container type by the user. The number of container types can be as many as the number of the designated waste elements. The corrosion and pitting parameters for each container type are input by the user. These input parameters are:

- Thickness of the container;
- Surface area of the container;
- Pitting parameter, *n*;
- Pitting parameter, *k*;
- Area scale factor, *a*;
- Number of pits per container;
- General corrosion rate;
- Clay content, *pH*, and aeration index of the soil.

Default values were built into the code for all of the above parameters, except for the thickness and the surface area of the container, which are required as input parameters.

The logistics of BREACH is shown in Fig. 2. After initialization, it examines every designated waste elements via an outer do-loop. First, if pitting parameter *n*, is omitted in the input, it calculates *n* from the clay and moisture contents of the soil using Eq. 2 in our complementary paper presented at this meeting (4). The moisture content is obtained from the output of FEMWATER. However, if clay content is not available, the value of *n* is defaulted to 0.26, 0.39, 0.44, or 0.59 according to the soil aeration index of good, fair, poor, or very poor, respectively. Second, if pitting parameter, *k*, is not known, the value of *k* is calculated based on the soil *pH* according to Eqns. 3, 4 and 5 (ibid.), but if the soil *pH* is also unavailable, it is assumed to be neutral. Third, default values are then assigned to the general corrosion rate, the scale factor *a*, and the number of pits, if not supplied as inputs by the user. Finally, for each waste element, pit depth and general corrosion depth are calculated according to Eqns. 1 and 7 (ibid.).

If the calculated general corrosion depth is larger than the thickness of the container, then the breached area is taken to be the total area of the container. If not, the calculated pit depth is compared to the thickness. If the pit depth is less than the thickness, the container is not breached (i.e. breached area = 0). Otherwise, the breached area is calculated using Eq. 6 in our other paper (4), but its maximum value is limited by the total area of the container.

This process is repeated for each designated waste element. The subroutine BREACH passes back the breached areas for all waste elements to GM2DXZ, which also registers the initial time when each container is breached. It subsequently calls subroutine LEACH to compute radionuclide releases.

Subroutine LEACH

The purpose of subroutine LEACH is to calculate the radionuclide release rate, and hence, the amount released from each designated waste element as a function of time.

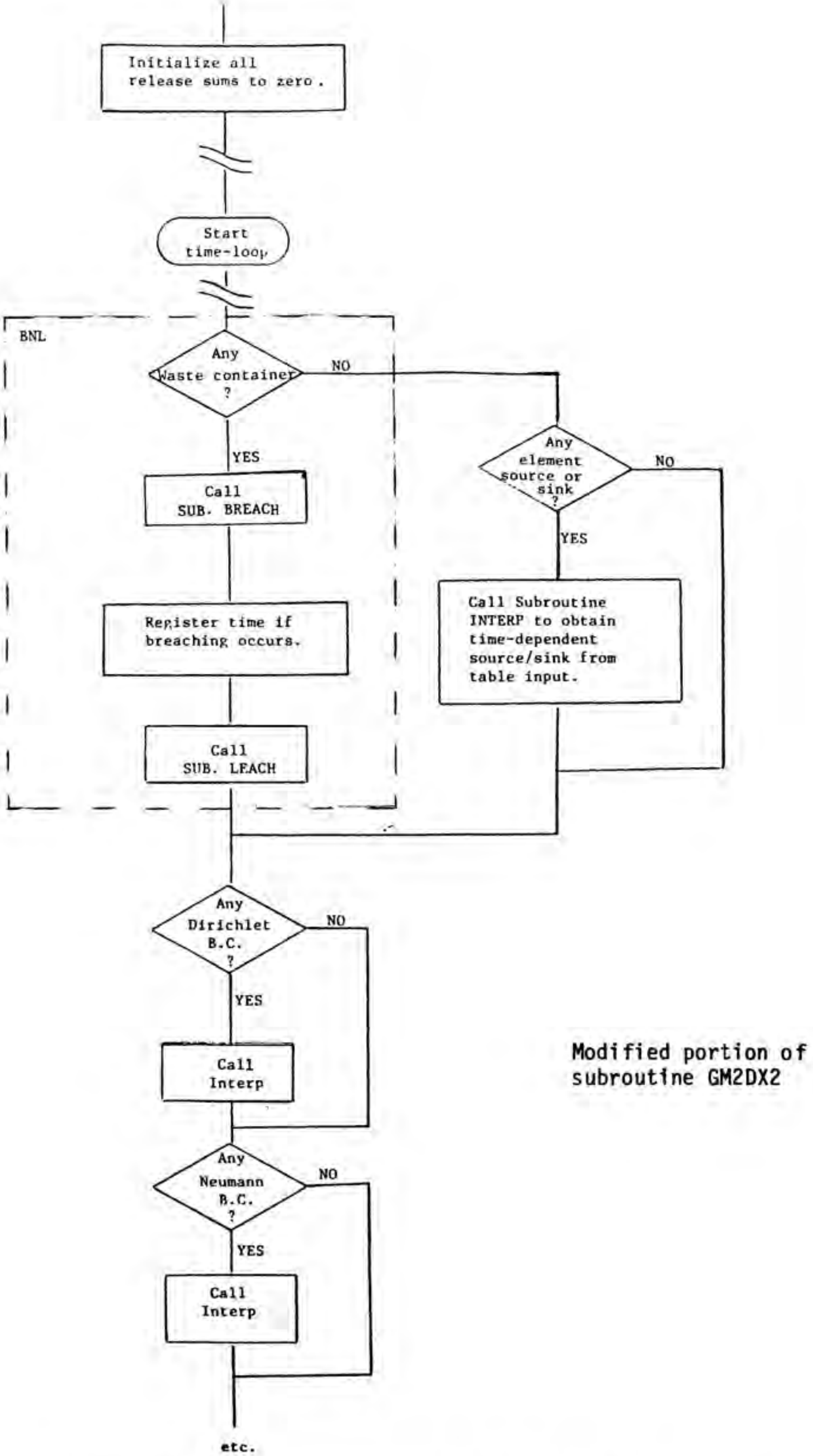

Fig. 1. Modified Portion of Subroutine GM2DX2 Where the BREACH and LEACH Subroutines are Called.

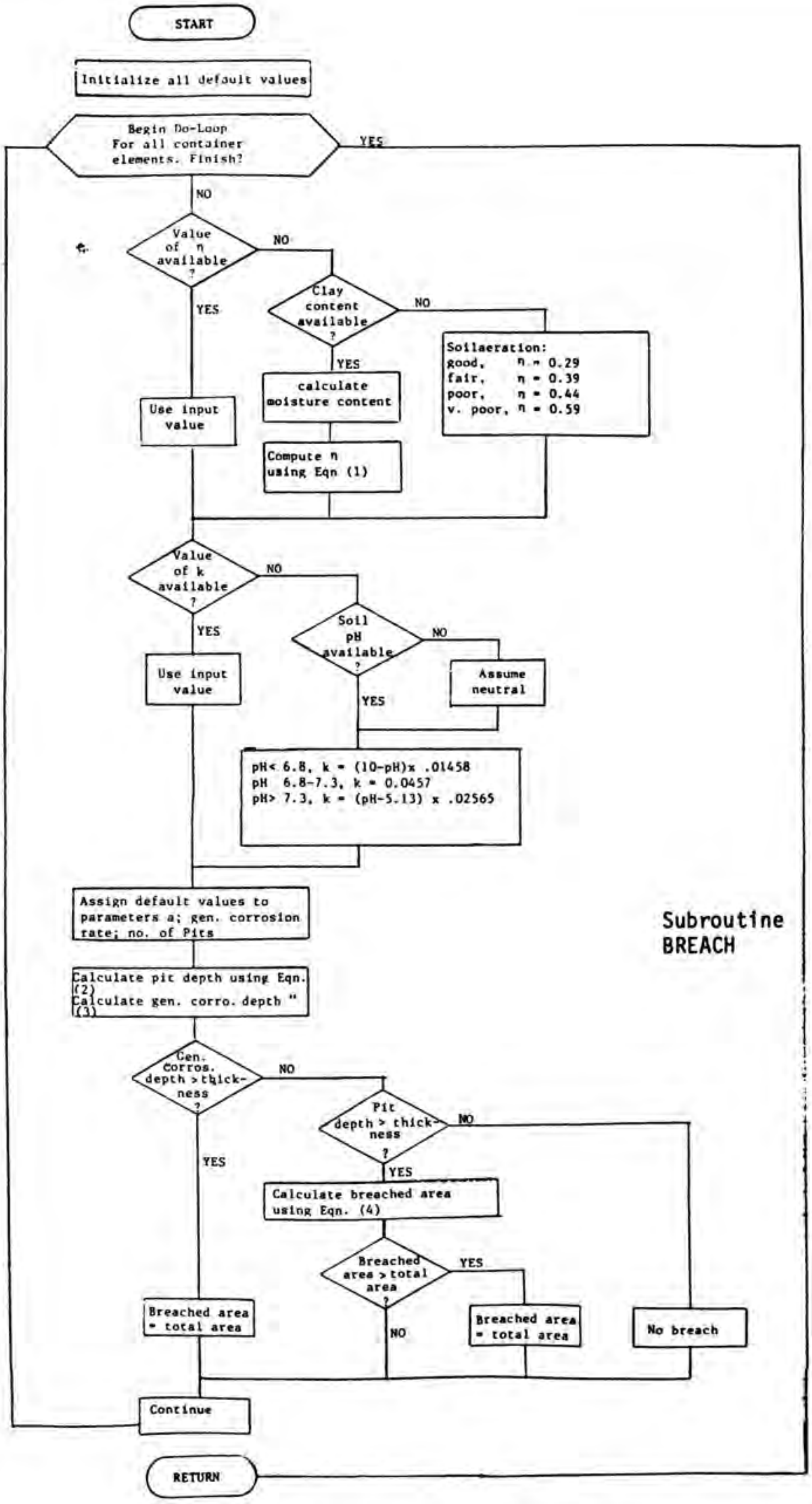

Fig. 2. Flowchart of Subroutine BREACH.

The incremental releases are summed at the end of each time-step to obtain the total amount of release.

Three parallel processes are modeled: 1) The surface rinse process, which releases all the radionuclides residing on the surface of the waste form, limited only by the solubility. 2) The diffusion process, which accounts for the diffusional transport of radionuclides through the pore water within the waste form, such as cement. 3) The dissolution process, which frees the radionuclides in the bulk solid by dissolving the solid phase. This would be the major release mechanism for wastes such as activated metals. The details of modeling these three processes are described in our other paper (4). When the container is not yet breached, the release is taken to be zero.

Subroutine LEACH treats all three processes independently from each other. The user can choose the relative contribution of each release mechanism by distributing fractions of the total mass available for each of the three processes. To decide the distribution, the type of waste form should be taken into considerations. For example, 100% of the total mass may be assigned to dissolution if the waste type is activated metal.

Similar to the container type in BREACH, a waste type is assigned to each of the designated waste elements. The number of waste types can be as many as the number of waste elements, and the waste type assignment can be independent of the container type. The release properties of each waste type are input by the user. They include:

- Total amount (in mass, or optional activity) of radionuclide available for release initially;
- Fractional distributions of the total mass for each of the three release mechanisms;
- Effective diffusion coefficient;
- Effective pore length;
- Effective water column length;
- Dissolution rate of the waste form.

The atomic weight and the solubility limit of the particular radionuclide under consideration are also required as inputs without reference to the waste type.

The logistics of subroutine LEACH is shown in Fig. 3. As in BREACH, the program considers each of the designated waste elements one by one via an outer do-loop. If the container for the element under consideration is not yet breached, the program bypasses all three release process calculations and assumes a release rate of zero. However, it still accounts for the loss in available mass of radionuclides due to radioactive decay, and updates the total mass available for release for the next time-step.

When the container is breached, the program first calculates the average moisture content, the magnitude of specific discharge (Darcy velocity), and the cross-sectional area of the element, based on the data from the auxiliary output file first created by FEMWATER. Then it proceeds to calculate releases from the three mechanisms discussed, in the order of rinse, diffusion and dissolution.

In all three modules, if the fractional distribution of the available mass is zero for that mechanism, the release from that mechanism is automatically put to zero, bypassing the calculations. (The first conditional branching (Block IF) encountered in all three modules. See Fig. 3).

The rinse and the dissolution release calculations are handled similarly, except for the rinse release, the radionuclide concentration in the leachate must first be calculated using Eq. 19 in our other paper (4), but limited by the solubility. This limit is set by another conditional branch (Logical IF). The release rate is then calculated using Equation 20 (ibid.). For the dissolution release rate, Equation 16 (ibid.) is used. At the end of both calculations, mass balance accounting is kept by calculating the released mass, and summing the cumulated release to date (to the current time-step). The actual released mass is again limited by the available mass. Finally, the available mass is updated for the next time-step by subtracting the released mass and the mass lost by radioactive decay during the current time-step. It should be noted that this treatment assumes no decay within a time-step when calculating releases. It accounts for decay only at the end of each time-step. This approach is conservative, because it over-estimates the amount of radioactive element actually available for the release processes.

The diffusional release calculation is slightly different from the others. Instead of obtaining the release rate (in mass per unit volume per unit time), it uses analytical solutions for mass flux as a continuous function of time (measured from the moment of first breach). The computed release is obtained by using numerical approximates, depending on the user's choice of boundary conditions. Four choices are provided:

1) One-dimensional semi-infinite medium;
2) One-dimensional finite medium with zero flux boundary;
3) One-dimensional finite medium with zero concentration boundary;
4) Three-dimensional finite cylinder with zero concentration boundary.

Equation 8 in our other paper (4) (approximate solution A in Fig. 3) is used for choice 1 above. Equations 8, 9, and 10 (ibid.) (approximate solutions A, B, and C in Fig. 3, respectively) are used for choice 2, and Eqns. 8, 11, and 12 (ibid.) (approximate solutions A, D, and E in Fig. 3, respectively) are used for choice 3. The automatic selection is according to a computed parameter TMP (TMP = Dt/ℓ^2).

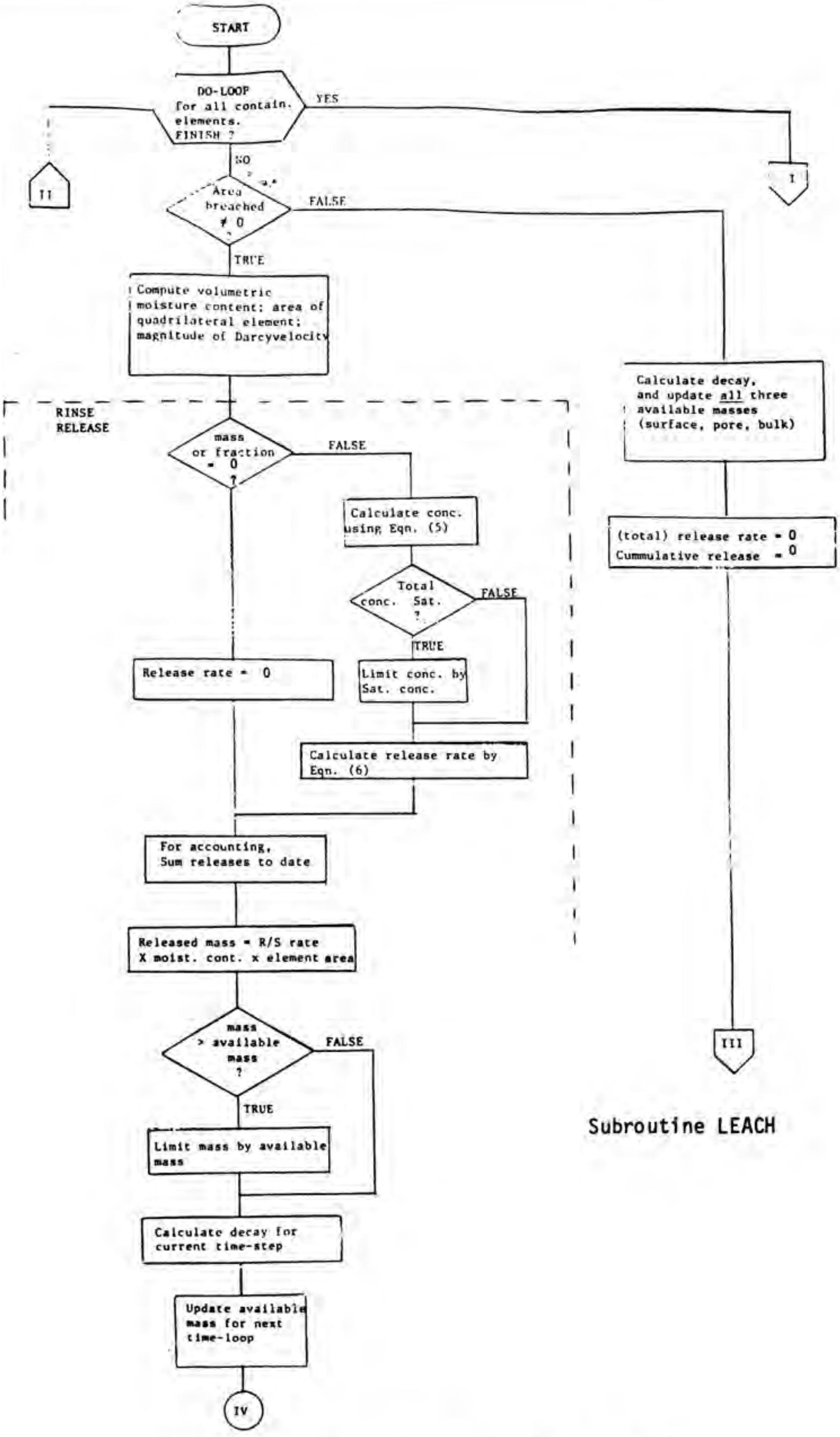

Fig. 3. Flowchart of Subroutine LEACH. (Page 1 of 3)

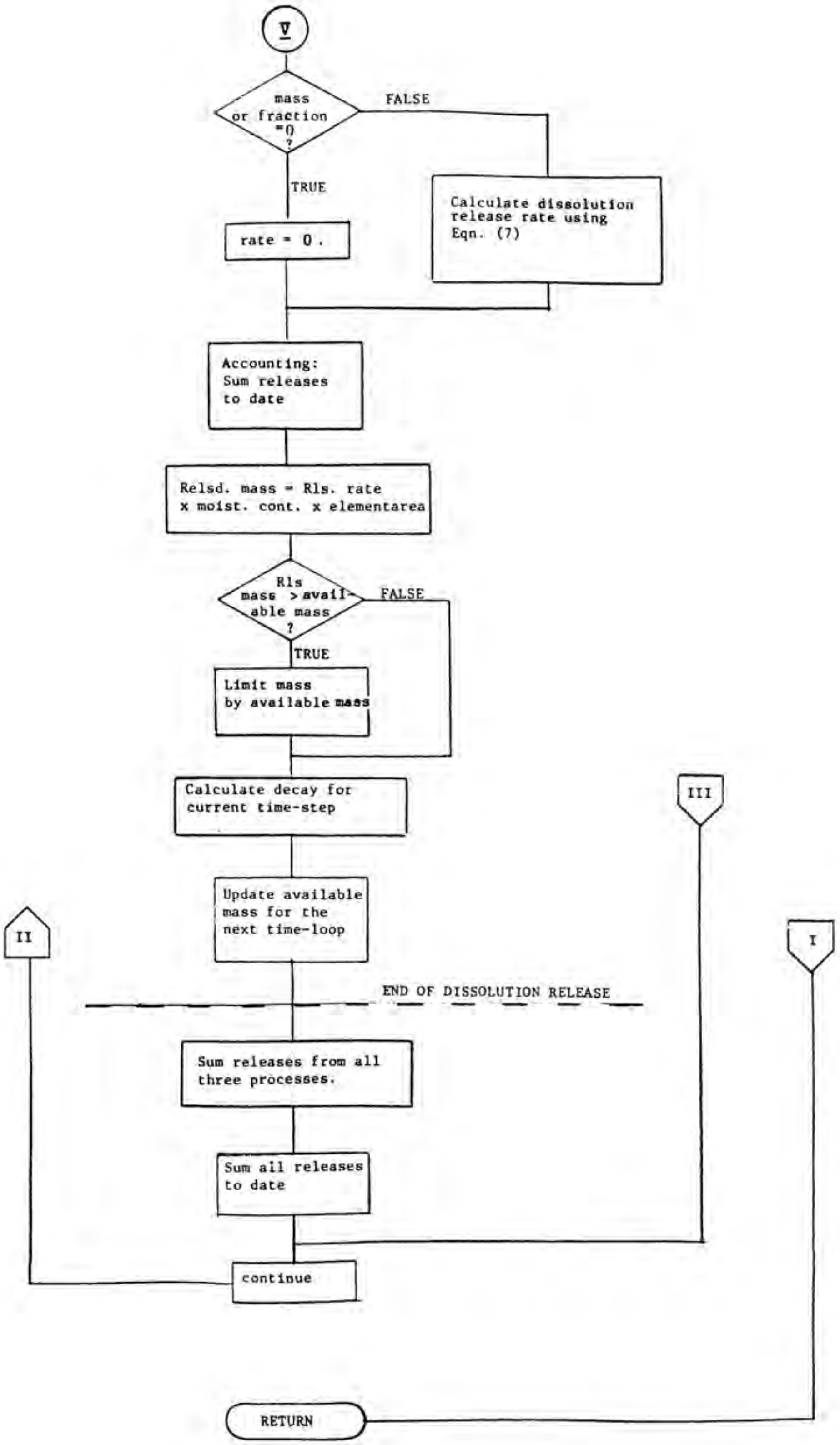

Fig. 3. (Page 2 of 3).

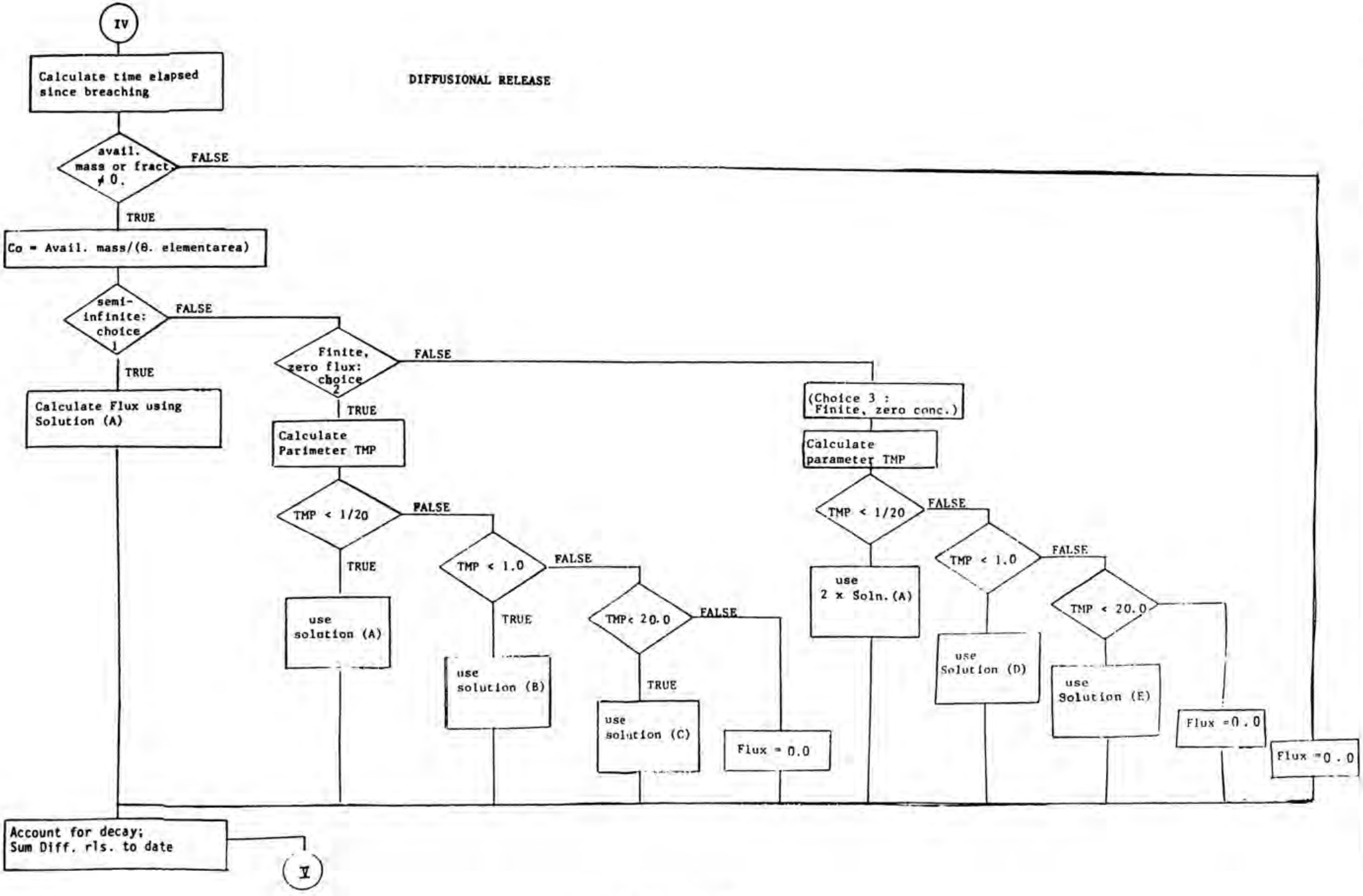

Fig. 3. (Page 3 of 3).

In extreme cases, flux is set to zero. (See Fig. 3). For choice 4, i.e. finite cylinder, Eqns. 13, 14, and 15 (ibid.) are used. (The branch for choice 4 is not shown in Fig. 3). Similar to the rinse and dissolution release calculations, mass balance accounting is kept after the release calculation is completed.

Releases from all three mechanisms (rinse, dissolution and diffusion) are added to obtain the total release from the element for the time-step. Cumulative release is then obtained by summing releases of all the time-steps. Then the do-loop is repeated until each waste element is accounted for. Subroutine LEACH then passes back to GM2DXZ an array containing release rates (mass/time/vol) for all designated waste elements.

IMPLEMENTATION

BLT was coded in standard Fortran-77, compiled, debugged and executed with an IBM-AT microcomputer using Microsoft Fortran Compiler version 4.0. In order to use BLT, a corresponding version of FEMWATER must first be executed with the pertinent geometry, hydraulic properties, and boundary conditions. The resulting information in water flow velocity and moisture content is written into an auxiliary file, which is then used by BLT to assess corrosion rate, leaching rate, and solute transport. BLT also reads an input file containing corrosion parameters, waste form leaching parameters, and transport properties of the soils. Figure 4 is a system procedural road map showing the input/output relationship of the two codes.

SENSITIVITY TESTING AND VERIFICATION

To verify the code and examine the overall sensitivities with different input values, we performed extensive tests on different portions of the completed BLT code in a two-dimensional vertical mode. For our initial tests, we used a finite simple element mesh of 100 (10 X 10) square elements and 121 nodes with uniform properties (Fig. 5). A steady-state simulation of water flow was first carried out with the code FEMWATER. The top boundary had a prescribed flux condition equivalent to a net rainfall of 5 cm per year, and the bottom boundary was the water table (Dirichlet boundary condition). The two sides were zero flux boundaries.

A number of transient simulations were performed using the code BLT, with 25 time-steps and equal time intervals of one year each. Zero flux conditions were applied to all the sides except the bottom where the concentration was zero at all times (Dirichlet boundary condition). Five elements (No. 18, 38, 58, 78, and 98) were designated as waste elements where the sources were located. Waste container parameters and waste type parameters were varied systematically for a number of test runs. Initially, a base case (PR-1) was set up and executed. Based on this base case, flags were turned on and off, and parameters were varied to test the different features and segments of the BLT code. The parameters for this base case are summarized in Table I. Only two simple examples of the test results are presented herein.

Base Case with and without Radioactive Decay

The upper curve in Figure 6 shows the total cumulative release without radioactive decay (from all three release processes: rinse, diffusion and dissolution) as a function of time, up to 24 years. The discontinuity at time = 10 yrs. is due to the fact that the container disappears abruptly as a result of general corrosion. Before year 10, the container is breached by pitting, and the release is scaled by the ratio of pit area to total area. Because the dissolution rate is slow (4×10^{-10} cm/sec), most of the release up to 24 years have been from the rinse and diffusion processes. This is the reason why only approximately 2 grams out of a total input of 3 grams have been released. After year 24, the release will be mostly from dissolution, and the release curve is expected to increase linearly approaching a limit of 3 grams.

Keeping all other parameters the same, the decay constant was set to 4.4×10^{-9} sec^{-1} (equivalent to a half-life of about 5 years), to see the effect of radioactive decay. The resulting release is represented by the lower curve in Fig. 6. Comparing the two curves, the difference in cumulative releases is small at early times, but the two curves diverges as time progresses due to decay. The curve with decay flattens considerably after 15 years, about three half-lives.

Effect of Solubility Limits on Rinse Release

To test rinse release, we "turned off" diffusion by setting the effective diffusion coefficient (D) to a very low value (1×10^{-14} $\text{cm}^2\,\text{sec}^{-1}$). The saturated concentration (CSAT) was set at three different values: 1.0 g/cm^3 (i.e. no limit), 1.0×10^{-4} g/cm^3 and 1.0×10^{-5} g/cm^3. The cumulative rinse releases from all three test cases are plotted in Fig. 7. For the case of CSAT = 1.0 g/cm^3 (top curve in Fig. 7), there is no rate limiting effect. The curve is limited only by the available mass of 1 gram. For the other two curves (middle and bottom curves in Fig. 7), the release rate is limited by the value of CSAT. They are approximately linear with time, and their slope, i.e. the rate of rinse release, is proportional to CSAT. This is expected, because for a constant water flow rate, at every time-step the amount released is approximately equal to a constant amount dissolved at saturation. The released amount will vary if the water flow rate is changed.

Several other tests on corrosion rate and mechanisms, diffusion rate and models, as well as dissolution rates were

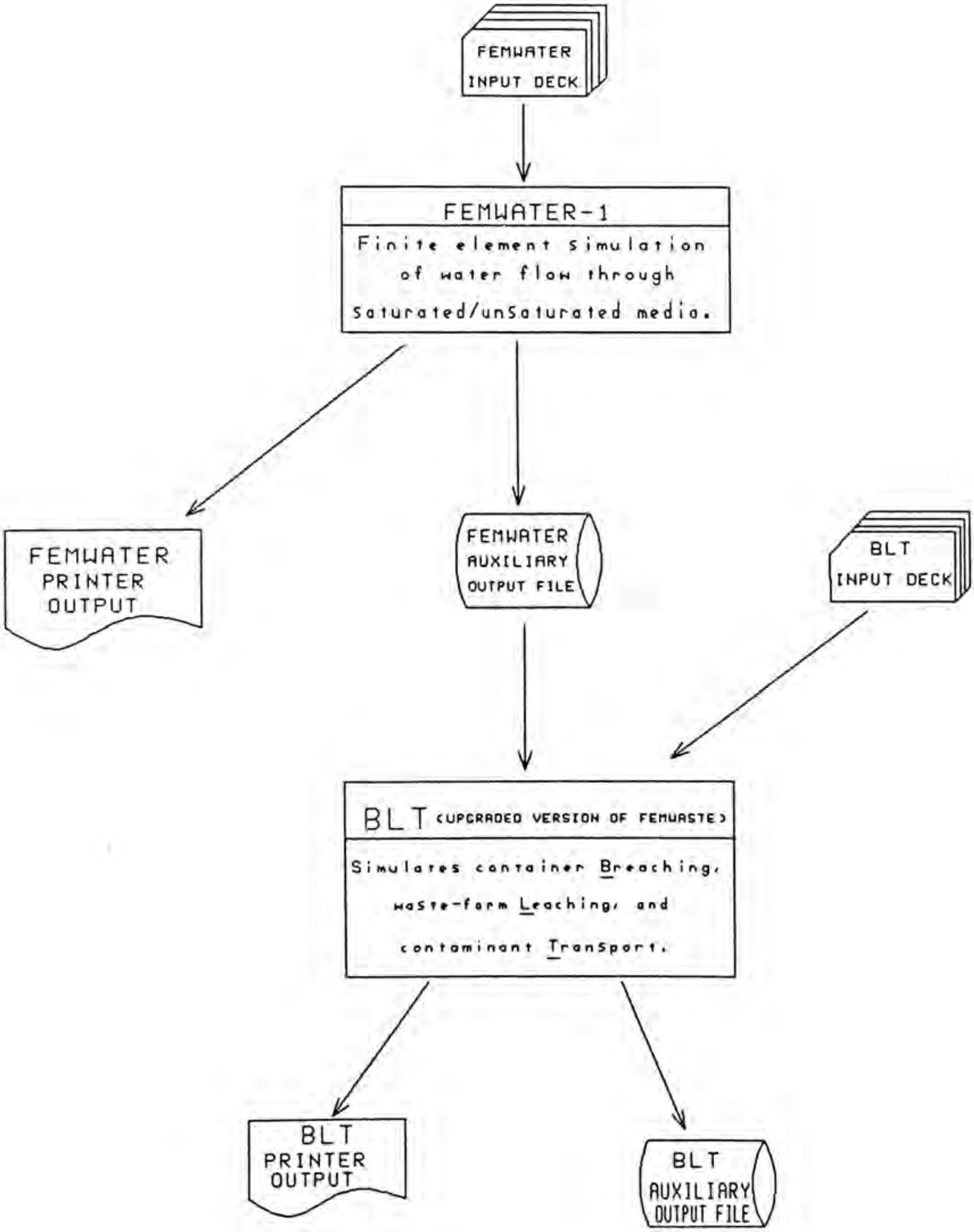

Fig. 4. Procedural Road Maps of FEMWATER/BLT Codes.

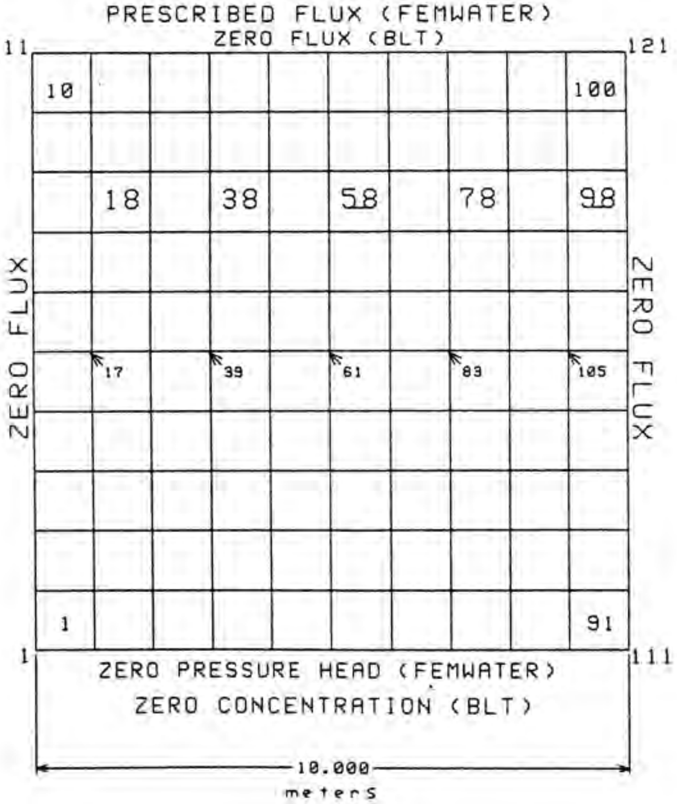

Fig. 5. Simple Finite Element Mesh for BLT Test Runs.

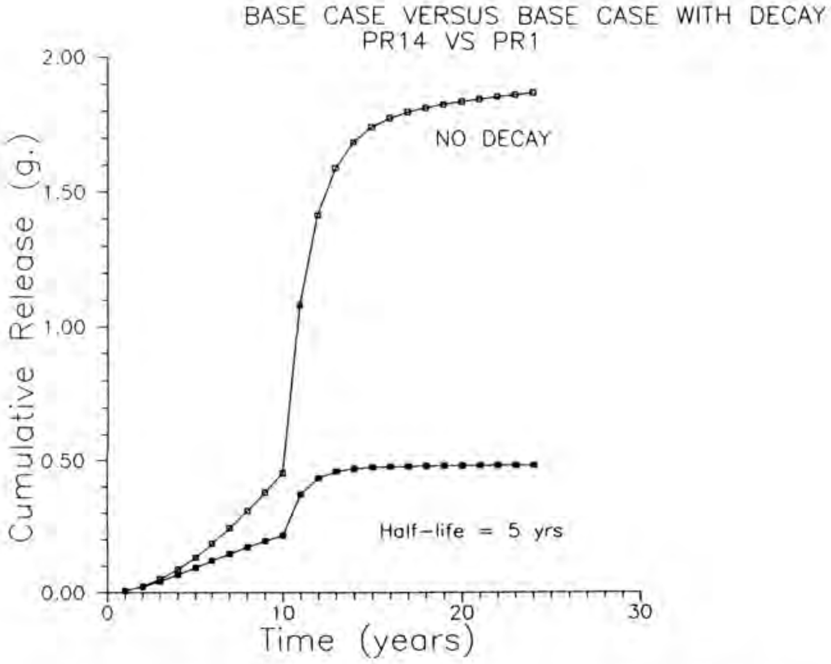

Fig. 6. Total Cumulative Release vs. Time, Comparing the Base Case and a Case with Radioactive Decay (half-life = 5 years).

also performed with reasonable results. These results are not included here, but will be discussed elsewhere.

SUMMARY

To predict the source term for low-level waste shallow land burial, we developed two computer modules to model the container breaching and the waste form leaching processes. These two modules are in the form of two separate subroutines - BREACH and LEACH, which were incorporated into a modified version of the existing computer code FEMWASTE. This resultant code, which contains all three modules of container breaching, waste form leaching and solute transport, was renamed BLT (Breach, Leach, and Transport). We successfully verified the BREACH and LEACH modules of the BLT code, and also carried out a number of sensitivity tests.

To use the BLT code, a corresponding version of FEMWATER must first be run. Starting with the input deck

TABLE I

Input Parameters for the Base Case.

BASE CASE (PR1)

IDIFF = 0 (semi-infinite medium)
CSAT = 1 (no solubility limit)
LAMBDA = 0 (no decay)

5 waste elements;
1 type of container;
1 type of waste.

Container: THICK = 0.127 (n/c)
n = 0.39; k = 0.074
No. of pits = 5×10^3
Gen. corros. rate = 4×10^{-10}
(i.e. through in 10 yrs.)
Area = 2.1×10^4 (n/c)
Ascale = 0.2 (n/c)
Clay = 0.2; pH = 6; Iaer = 2
(not used)

Waste: Mass distribution
(Rinse, diff., diss.)
1/3, 1/3, 1/3, total = 3 g (n/c)

Surf. Area = 1×10^5, Vol = 1×10^6
SA/Vol = 1/10 (n/c)

D = 1×10^{-6}
Diss Rate = 4×10^{-10}

Pore L = 10;
Water col L = 100 (not used)

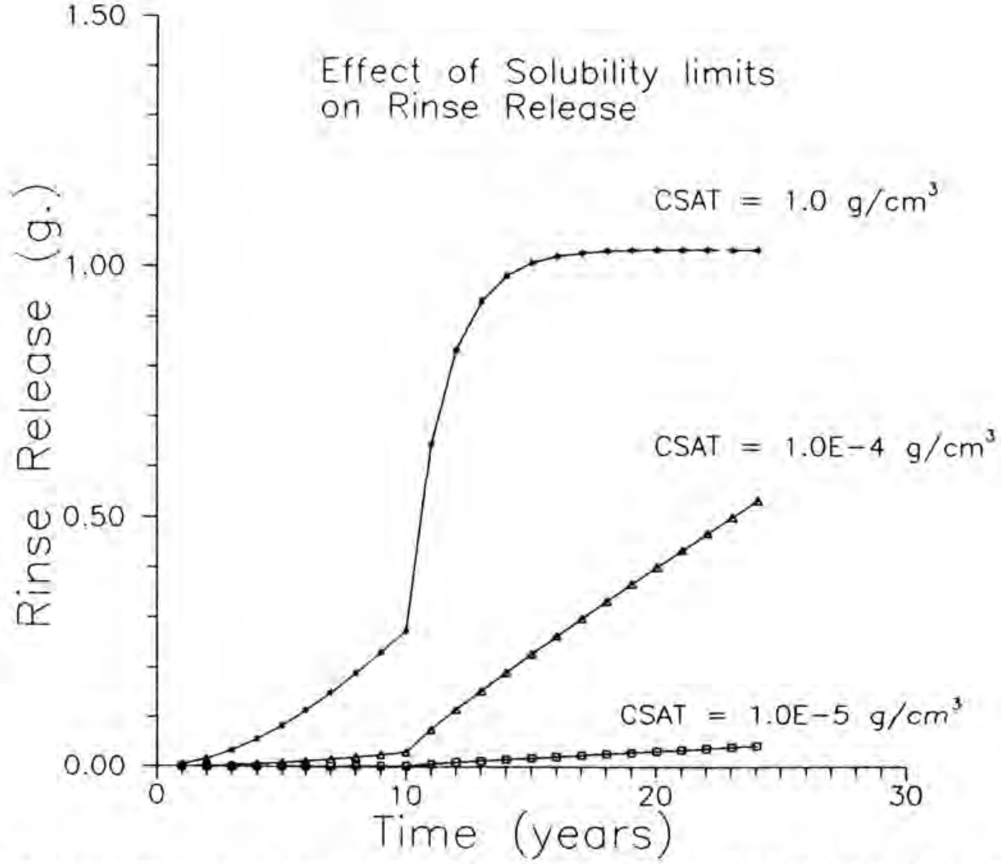

Fig. 7. Cumulative Rinse Release vs. Time, Showing the Effect of Solubility Limits on Rinse Release.

(file) of FEMWATER, the input data include:

- I.D. number & basic control parameters;
- Geometry;
- Soil properties (for water flow);
- Initial/boundary conditions.

The output files of FEMWATER contains the following information needed by BLT:

- Time (transient simulation only);
- Pressure head;
- Total head;
- Moisture content;
- Specific discharge (Darcy velocity).

To execute BLT, the additional input requirements are:

- I.D. number & basic control parameters;
- Soil properties (for transport);
- Container locations and properties;
- Waste form properties;
- Initial/boundary conditions.

The final results calculated by BLT include:

- Time;
- Time of breach of each container;
- Concentration;
- Flux;
- Releases (release rate & cumulative amounts).

BLT can be compiled and executed using a desk-top microcomputer, such as the IBM-AT. Because it was written in standard Fortran-77, it can also be transportable to other computer systems without any difficulties.

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