2

CONF-940553--21

UCRL-JC-114699
PREPRINT

Performance Assessment Model of a Single Waste Package

William J. O'Connell
Tzou-Shin Ueng
Lynn C. Lewis

This paper was prepared for submittal to the
American Nuclear Society
International High Level Radioactive Waste Management Conference
Las Vegas, NV
May 22-26, 1994

January, 1994

Lawrence Livermore National Laboratory

PERFORMANCE ASSESSMENT MODEL OF A SINGLE WASTE PACKAGE*

William J. O'Connell, Tzou-Shin Ueng, and Lynn C. Lewis
Lawrence Livermore National Laboratory
P.O. Box 808
Livermore, CA 94551
(510)422-8789, 423-9681, 422-8949

ABSTRACT

PANDORA-1.1 is a system model for the mobilization and release of radionuclides from a spent nuclear fuel disposal package. Earlier processes affecting release are represented by input tables. Several groundwater contact alternatives and spent fuel constituents lead to different release-rate behaviors and controlling parameters. Rate control is provided by a product of parameters from hydrology, design, and/or geochemistry/waste form interaction parameters. The program is designed to accommodate evolving requirements such as a wider range of hydrological input values. A computerized configuration management system automates much of the change control process.

INTRODUCTION

This paper describes a system model of some core features of the performance of waste packages for the permanent disposal of spent nuclear fuel. The paper also shows examples of the range of release-rate performances of the waste package system produced by these core features under a range of waste package environments and radionuclide types. The model is realized in the prototype computer program[1] PANDORA-1.1. The PANDORA system model links processes leading to possible release of radionuclides from a single waste

* Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

package. The process models are summaries based on data or on more detailed models. The current version, PANDORA-1.1, models the last steps in the sequence of processes leading to release of radionuclides into rock. Earlier steps, such as the hydrologic flux outside the waste package and the breach of the container, are now taken as inputs. They can be modeled in later versions of PANDORA, or incorporated with other performance assessment models, e.g., YMIM[2].

MODEL FEATURES

The waste package features and processes treated in PANDORA-1.1 are summarized in Table 1. The program is organized at the top level by the two process categories of mobilization (called the alteration model because that is the major process for mobilization) and transport. For each process category the effects of bathtub and flowthrough water contact alternatives and air contact are modeled. The air-mediated processes take place alone or in parallel if there is also water contact. The effects in these process conditions are treated for the radionuclide types of gas, high-solubility (mobilization limited by matrix alteration rate) and low-solubility (mobilization limited by solubility). For each type, the different locations within the spent fuel are treated.

Many of the features and processes of the waste packages and their immediate environments have been identified as needed inputs and hence potentially important to performance. The topical areas of the inputs for waste form alteration and waste release are: (1) Container breach, (2) Near-field hydrology, (3) Waste form characteristics, and (4) Geochemistry/waste form interaction.

Table 1. Features and Processes Treated in PANDORA-1.1

In-package hydrology[a] and mobilization process	Air Water contact – bathtub Water contact – flowthrough
Transport process[a]	Gaseous diffusion Advection – bathtub Advection – flowthrough
Chemical types[b]	Gas High solubility Low solubility Secular equilibrium
Locations of radionuclides[c]	Surface of cladding Fuel/cladding gap Spent fuel pellet matrix

Note: (a) Wet diffusive contact is treated separately by Ueng and O'Connell[3,4].
(b) For a radionuclide with a given solubility, the program determines whether the low or high solubility process controls its release rate.
(c) Cladding and hardware solids can be treated in a future version.

The main features that comprise the substance of the calculation are:

1. Availability for release: the interplay of the in-package hydrology and the waste form properties and alteration.
2. Release: the interplay of availability, solubility, inventory, and release processes.

Other features included in the PANDORA-1.1 calculation are:

1. In-package hydrology,
2. Timing of releases and spreading of releases over time,
3. Treatment of the different locations of radionuclides in spent fuel,
4. Tracking of inventories due to buildup and decay,
5. Exhaustion of inventories by transport and decay, and
6. Ratio of release rates to NRC limits.

Careful attention was given to making the program robust over a wide range of water flux, matrix chemical alteration rate, and solubility parameter values. The average groundwater flux is expected to be low, but in some scenarios, the water flux could be episodic flux in a few fractures, and hence one or two waste packages could experience high water flux (see example application below). PANDORA-1.1 checks whether the solubility-limited radionuclides are even more limited by the matrix alteration rate. This limit could be the control in cases of very high water flux. On the other hand, PANDORA-1.1 does not now accommodate solubility limits for the present matrix-limited category. Such a limit could dominate in very low water flux cases. The model can be revised for this in a future version.

Figure 1 provides a high-level schematic of the flow of control and data in the PANDORA-1.1 model. The calculation starts at the container breach time which is specified by the user. The system model selects the size of the next time step based on system state changes, using small time steps when release rates may be changing rapidly. The environment submodel determines if there is groundwater flowing into the waste package and computes the quantity of groundwater flow according to the input data. The inventories of the tracked radionuclides in Curies per metric ton initial heavy metal (MTIHM) at various years out-of-reactor are given in external data files. PANDORA-1.1 determines the inventories at a desired year by an interpolation scheme. Based on the user-specified groundwater exposure scenarios, the waste form alteration submodel determines the amount of gaseous radionuclides mobilized and the concentration of waste radionuclides present in liquid groundwater inside the waste package. The radionuclide transport submodel then determines the release rate for each radionuclide.

This series of calculations is repeated at each time step until a specified post-closure time is reached, at which point the assessment is complete. The release rates of these radionuclides are checked against the NRC criteria given in the Code of Federal Regulations in Section 10 CFR 60. The total cumulative release of each radionuclide is also computed for the user-specified time period of analysis.

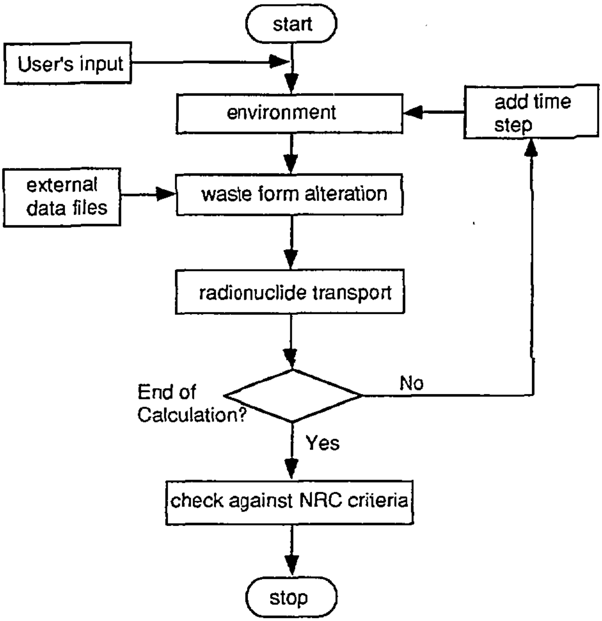

Figure 1. System Model of PANDORA-1.1

SOFTWARE ENGINEERING

Software engineering provides for effective development of software and eventual qualification for its intended use. PANDORA-1.1 is graded as a preliminary non-quality-affecting code. Appropriate procedures were selected at the time of QA grading for the activity. The practical requirements at this stage are configuration management; change control; sufficient documentation to explain the code's models, input-output data, and some of the implementation structure; and foundations for extended development.

Changing requirements are a fact of life. In the PANDORA development thus far, extensions to longer time periods of analysis and to unanticipated scenarios of high groundwater influx were required. These then required more detailed consideration of inventory depletion, alternation between dissolution-rate and solubility controls on release rate, and timing of water-related system state changes. Control of the changing software products is required to verify implementation of the changes and retention of unchanged requirements.

Performance assessment software validation can be started now and can establish some of the work products leading to overall validation[5]. Model validation is a longer-term challenge due to the broad model scope and presently limited data. Software validation is[6] "the process of evaluating a [software] system or component during or at the end of the development process to determine whether it satisfies specified requirements." It is helpful to have the requirements and their implementation structure described at several levels of perspective and summarization. Early work on the software validation products also helps minimize programming errors and implicit assumptions.

PANDORA development uses these software engineering tools:

a) For traceability to requirements, we maintain an extended work breakdown structure (WBS) database of the activity and correlation of software product elements to the extended WBS elements.

b) We assess compliance with requirements by partial implementation of walkthroughs or reviews, and track all identified exceptions through the extended WBS database to ensure closure.

c) We developed a computerized configuration management system based on UNIX® utilities and shell scripts. It manages source code modules, test setups, test results, test configurations, and software releases. Its programmed functions include:

i) capture of the revision-number relationships among all these elements and reference to the extended WBS database elements;

ii) check-out/check-in of source code, test setup, and test result modules to document revision versions and to prevent concurrent modifications;

iii) automated regression testing of new software configurations. This runs a test suite, compares to stored test results, reports differences, and archives the products.

EXAMPLE APPLICATIONS

Different release-rate behaviors occur and different sensitive parameters are controlling, depending on the radionuclide type and on the base-case parameter values. The examples illustrate some of these diverse possible situations.

For the bathtub water contact/transport mode and for the highly soluble radionuclide Tc-99, Fig. 2 illustrates release rate results for a range of water influx values. The example assumes a container breach near the top at 1700 years after emplacement, and a waste matrix alteration rate of 1.2×10^{-3} fraction/year. Breached cladding of all the spent fuel rods is assumed. The incoming water gradually fills the container, contacting more spent fuel as the standing water level rises, and then starts to spill over. The cladding-gap component of the Tc-99 is assumed to be readily dissolved when wetted. The matrix alters and releases Tc-99 into the water at a steady rate after being wetted. Mixing of new incoming water with the standing water is assumed.

For this process mode and radionuclide type, the release rate is controlled by the interplay of the waste alteration process and the turnover of the standing water. The turnover rate is the inflow rate divided by the void volume up to the breach level, and is thus a combination of hydrological and design parameters. The plateau value of the release rate is determined by the waste matrix alteration rate for the 0.05 and 0.01 m^3/y water influx cases, and by the stored-water turnover rate for the 0.001 m^3/y water influx case.

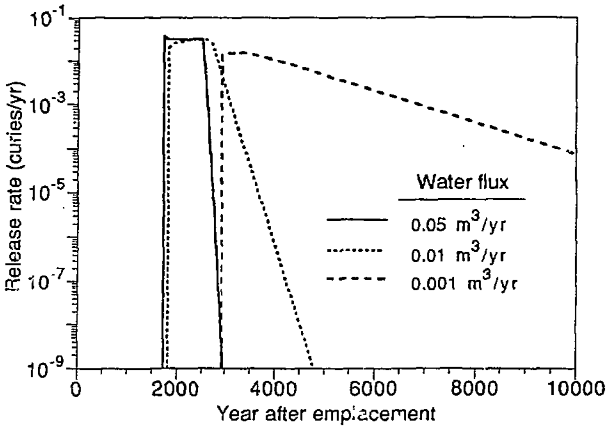

Fig. 2 Release rate of Tc-99 from a single waste package for the bathtub water contact mode.

The cladding-gap component of Tc-99 makes a discernable contribution to the release-rate curve only in cases where the water-fill time is short compared to the waste matrix alteration time. This can be seen in Fig. 2 only in the curve for the case of 0.05 m^3/y water influx.

Figure 3 illustrates release rates for the flowthrough water contact mode and Tc-99. The results show release rates with a closer congruence to the waste mobilization rate and a much smaller delay time after water contact to the spent fuel than in the bathtub mode of Fig. 2. The flowthrough mode is associated with a container with multiple through-wall breaches. A fraction of the spent fuel is wetted by the process; the fraction is an input parameter. We assume that as the wetted spent fuel is altered, the wetting progresses to maintain a constant wetted surface area. We assume that the cladding-gap component of the initially wetted fraction is available for rapid solubilization, and that the cladding-gap component of the remaining spent fuel is released gradually as that spent fuel becomes wet. With water influxes in the anticipated range, the time delays to provide enough water mass even for just a wetting film can be a noticeable part of the overall release process.

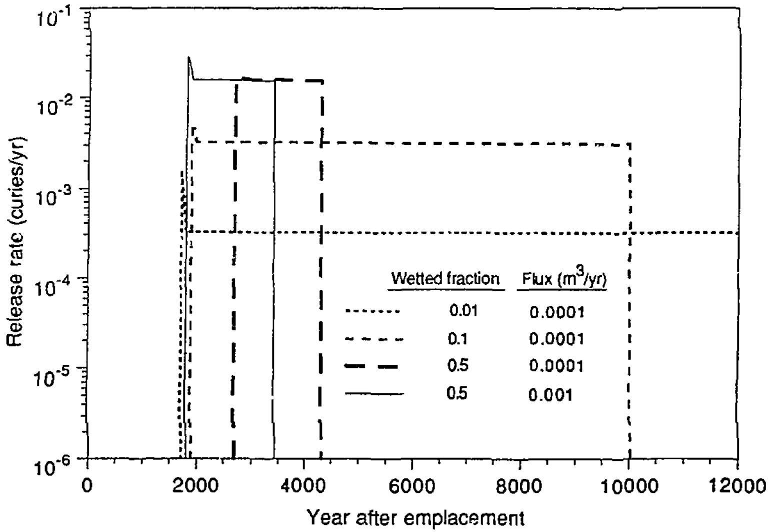

Figure 3 Release rate of Tc-99 in the flowthrough mode.

With this process model, the plateau value of the rate depends on the product of the wetted surface area times the waste matrix alteration rate. Fig. 3 shows results for cases with several values of spent fuel wetted fraction and water influx. The peak rate of the short-duration release from the wetted cladding-gap component, added onto the plateau, depends on the wetted surface's water film volume, the water inflow rate, and the cladding-gap inventory.

For a radionuclide, the choice between solubility and matrix alteration as the release-rate controlling process depends on a case's solubility, water influx, and matrix alteration rate parameter values. The rationale for the modeled choice illustrated by the cases of Fig. 4 follows. Solubilities can vary by orders of magnitude depending on the geochemical conditions arising from the incoming geochemistry and its interaction with the waste form materials. The water influx can also vary by many orders of magnitude if we include unanticipated conditions in the scope of the model. Hence it is convenient to make the choice between solubility and matrix alteration within the model rather than by hand calculation prior to data input.

The water influx varies by up to seven orders of magnitude in the situations considered in the model scope. Water influx rates of $1x10^{-5}$ to $1x10^{-3}$ m^3/y or less might be anticipated based on the low groundwater percolation rates. Flint et al.[7] project a net infiltration rate into the mountain of up to 0.02 mm/y during wet climates, and a flux at the potential repository level holding steady at the long-term average infiltration of 0.005 mm/y. They also considered alternate cases with up to a factor of ten higher infiltration near the ground surface. Long and Childs[8] model the present-day infiltration as about 1 mm/y, and the long-term future values averaging 2.4 mm/y. First taking the flux at depth to be 0.005 mm/y and the cross-sectional area intercepting the waste package for water collection to be about 1 m^3, the available water for inflow is $5x10^{-6}$ m^3/y. A flux of 1 mm/y would give an inflow of $1x10^{-3}$ m^3/y. This cross-section method was used in earlier analyses[9]. Only a fraction if any of this nearby water would enter the waste package in most cases. The larger water flux values shown in Fig. 2 are for hypothetical cases where much of the infiltration water is channeled in a few fractures, intersecting only a handful of waste packages. Gauthier et al.[10] examined hypothetical fluxes in 1-m-long fractures, with a mid-range case at a flux of 1.92 m^3/y, and a hypothetical range from 0.004 m^3/y up to 157 m^3/y.

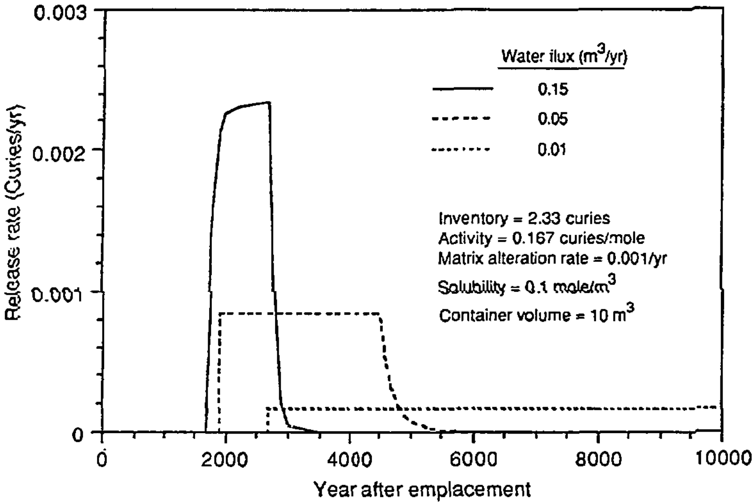

Figure 4 Effect of water flux on release rate of a solubility-limited radionuclide in the bathtub mode, for an example radionuclide with the parameter values shown.

Either solubility or the matrix alteration rate may control the availability of a radionuclide for release under different conditions. PANDORA-1.1 compares the release rates from either process to select the applicable, more limiting, process. The radionuclide solubility is part of an element solubility; it is calculated taking account of the various isotopes present including stable isotopes of an element. For high water flux cases, a solubility-limited radionuclide could be released eventually to the extent that its inventory becomes exhausted; hence this is also checked and modeled. In the last stage of such a release history, the radionuclide is below its solubility limit and becomes controlled by other factors. Figure 4 illustrates these modeled behaviors for a hypothetical radionuclide with selected parameter values. For the two lower water inflow rates shown, the release rate is controlled by solubility times water outflow. For the highest water inflow rate shown, the release rate is controlled by the matrix alteration rate.

PROJECT APPLICATIONS

PANDORA-1.1, with subsequent summation of releases over different waste packages at different local conditions, has been used to provide source terms[9,11] for total system performance assessments[12]. The program has also been used to provide guidance for the source term for further total system performance assessment[13] and for unanticipated high water flux cases[10]. Some of the modeling concepts of PANDORA were incorporated in a streamlined form in YMIM[2], which examines the interaction of a larger number of processes.

SUMMARY

The present model, although concentrating only on waste mobilization and release processes for several different types of radionuclides, shows a range of release-rate behavior and different sensitive parameters under different environmental and design conditions. Model extension to include processes rather than input tables for geochemistry-waste form interaction, container, and near-field environment would add to the range of resulting performance types. Our automated configuration management system eases development control and has been adopted also for the related YMIM program. Further development will likely merge approaches and features of the LLNL programs PANDORA, YMIM, and our diffusion-mediated release program.[3,4]

REFERENCES

1. W. J. O'Connell, T.-S. Ueng, and L. C. Lewis, *Post-Closure Performance Assessment of Waste Packages for the Yucca Mountain Project*, UCRL-ID-111979, Lawrence Livermore National Laboratory, Livermore CA, 1993.

2. A. Lamont and J. Gansemer, "Integrated Modelling of Near Field and Engineered Barrier System Processes," this conference.

3. T.-S. Ueng and W. J. O'Connell, "Diffusive Barrier Simplified Analysis--Design and Sensitivity Applications," *Proceedings, Nuclear Waste Packaging, FOCUS '91*, Las Vegas, NV, Sept. 29 - Oct. 2, 1991, p. 277, American Nuclear Society, LaGrange Park IL; and UCRL-JC-104913, Lawrence Livermore National Laboratory, Livermore CA, 1991.

4. T.-S. Ueng and W. J. O'Connell, *Diffusion Releases Through One and Two Finite Planar Zones from a Nuclear Waste Package*, UCRL-ID-109215, Lawrence Livermore National Laboratory, Livermore CA, 1992.

5. L. C. Lewis, J. J. Dronkers, B. Pitsker, "Control of Research Oriented Software Development", In *ASQC Quality Congress Transactions*, Anaheim, CA, 1986, American Society for Quality Control, Milwaukee WI, 1986.

6. *Quality Assurance Requirements and Description for the Civilian Radioactive Waste Management Program*, DOE/RW-0333P, Rev. 0, U.S. Department of Energy, Office of Civilian Radioactive Waste Management, Washington, DC, December 18, 1992.

7. A. L. Flint, L. E. Flint, and J. A. Hevesi, "The Influence of Long Term Climate Change on Net Infiltration at Yucca Mountain, Nevada," *Proceedings, 4th International High-Level Radioactive Waste Management Conference*, Las Vegas, NV, April 26-30, 1993, p. 152, American Nuclear Society, LaGrange Park IL, 1993.

8. A. Long and S. W. Childs, Rainfall and Net Infiltration Probabilities for Future Climate Conditions at Yucca Mountain," *Proceedings, 4th International High-Level Radioactive Waste Management Conference*, Las Vegas, NV, April 26-30, 1993, p. 112, American Nuclear Society, LaGrange Park IL, 1993.

9. M. J. Apted, W. J. O'Connell, K. H. Lee, A. T. MacIntyre, T.-S. Ueng, T. H. Pigford and W. W.-L. Lee, "Preliminary Calculations of Release Rates From Spent Fuel in a Tuff Repository," *Proceedings, 2nd International High-Level Radioactive Waste Management Conference*, Las Vegas, NV, April 28 - May 3, 1991, p. 1080, American Nuclear Society, LaGrange Park IL, 1991.

10. J. H. Gauthier, M. L. Wilson, and F. C. Lauffer, "Estimating the Consequences of Significant Fracture Flow at Yucca Mountain," *Proceedings, 3rd International High-Level Radioactive Waste Management Conference,* Las Vegas, NV, April 12-16, 1992 p. 891, American Nuclear Society, LaGrange Park IL, 1992.

11. M.J. Apted, W.J. O'Connell, K.H. Lee, A.T. MacIntyre, T.-S. Ueng, W. W.-L. Lee, and T.H. Pigford, *Preliminary Calculations of Release Rates of Tc-99, I-129, Cs-135, and Np-237 from Spent Fuel in a Potential Repository in Tuff,* LBL-31069, Lawrence Berkeley Laboratory, Berkeley, CA. 1990.

12. R. W. Barnard and H. A. Dockery, Editors, "*Technical Summary of the Performance Assessment Calculational Exercises for 1990 (PACE-90), Volume 1: "Nominal Configuration" Hydrogeologic Parameters and Calculational Results,*" SAND90-2726, Sandia National Laboratories, Albuquerque, NM, 1991.

13. R. W. Barnard, M. L. Wilson, H. A. Dockery, J. H. Gauthier, P. G. Kaplan, R. R. Eaton, F. W. Bingham, and T. H. Robey, "*TSPA 1991: An Initial Total-System Performance Assessment for Yucca Mountain,*" SAND91-2795, Sandia National Laboratories, Albuquerque, NM, 1992.