

SOTEC: A SOURCE TERM CODE FOR HIGH-LEVEL NUCLEAR WASTE GEOLOGIC REPOSITORIES

USER'S MANUAL: VERSION 1.0

Iterative Performance Assessment — Phase 2

Prepared for

**Nuclear Regulatory Commission
Contract NRC-02-88-005**

Prepared by

**Center for Nuclear Waste Regulatory Analyses
San Antonio, Texas**

July 1992



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1 BACKGROUND

In accordance with the provisions of the Nuclear Waste Policy Act (NWPA), as amended of 1987, the Nuclear Regulatory Commission (NRC) has the responsibility of granting a license for the first (and subsequent if any) geologic repository for high-level nuclear waste (HLW). The Center for Nuclear Waste Regulatory Analyses (Center) at the Southwest Research Institute (SwRI) is a Federally Funded Research and Development Center (FFRDC) created to support the NRC in its mission of licensing the repository. To meet its licensing function, the NRC will review the application submitted by the Department of Energy (DOE). One of the critical sections of the license application will deal with the assessment of the future performance of the repository system which has to meet certain minimum standards established by regulations.

In order to develop capabilities to review the performance assessment in DOE's license application, the staffs of the NRC and the Center are engaged in developing and applying performance assessment methods and models to existing data. Later, at the time of license application review, these methods may be used to conduct independent performance assessment, if the NRC elects to do so.

Because of the large space and time scales involved in estimating repository performance, mathematical models encoded as computer codes are the chosen tools for performance assessment. The repository system consists of designed (or engineered) barriers embedded in the natural geologic setting. Estimating performance of the total system requires that the behavior of these components be projected under possible future conditions. This is obviously a complex task that requires a variety of calculations. The development of the executive module described in a previous report (Sagar and Janetzke, 1991) is a step towards performing these calculations in a systematic manner. The Source Term Code (*SOTEC*) described in this report will work along with other such modules under the control of the executive module.

The *SOTEC* code is designed to calculate a space- and time-dependent radionuclide source term from the engineered barriers. *SOTEC* considers the collection of all the waste packages emplaced in the repository as well as the host rock immediately surrounding each waste package.

1.1 REGULATORY BACKGROUND

As indicated above, the conduct of performance assessments has, in addition to its other functions, a very specific regulatory purpose—that of determining whether the geologic repository system satisfies the regulatory standards. This is done by comparing the estimated values of the regulatory performance measures with the minimum values of the same measures specified in the regulations. Thus, the performance assessment models must be designed to estimate the regulatory performance measures. In addition to the regulatory function, performance assessments will also be used to design (by DOE) and judge the adequacy of (by NRC) the site characterization program. Because of its interdisciplinary nature, performance

assessments are also used to integrate technical work across disciplines. Operational flexibility in the Total System Performance Assessment Model as well as in its constituent modules is necessary to meet these varied objectives.

The primary regulations applicable to the HLW geologic repository were promulgated by the NRC in 10 CFR Part 60—Disposal of High-Level Radioactive Wastes in Geologic Repositories. Two sections of 10 CFR Part 60 pertain specifically to post-closure performance. These are Section 60.112—Overall System Performance Objective for the geologic repository after permanent closure; and Section 60.113—Performance of particular barriers after permanent closure. Section 60.112 makes reference to satisfying the generally applicable environmental standards for radioactivity established by the Environmental Protection Agency (EPA). These environmental standards referred to in Section 60.112 were promulgated by the EPA in 40 CFR Part 191 in 1985. These EPA standards are currently under court ordered remand and may change. The Total System Code (TPA) and *SOTEC* will be modified when the final EPA standard is available.

Three different performance measures are used in 40 CFR Part 191. The first is the release of radioactivity over the entire (integrated over space) accessible environment boundary cumulated (integrated over time) over 10,000 year period after closure (Section 191.113—Containment Requirements). This cumulative release of radioactivity must not exceed specified limits at specified probability levels. The preferred method of representing this performance measure is through a Complimentary Cumulative (Probability) Distribution Function (CCDF). The second is that the dose to humans in the first 1,000 years after repository closure must not exceed specified limit (Section 191.15—Individual Protection Requirements). This requirement has no probability attached to it. Third is the concentration of alpha, beta and gamma emitting radionuclides must not exceed specified limits (Section 191.16—Ground Water Protection Requirements). There is no probability attached to this requirement also. While the first performance measure is to consider all future credible scenarios, the other two apply only to "undisturbed" performance. Source term will have to be calculated for each of these regulatory requirements.

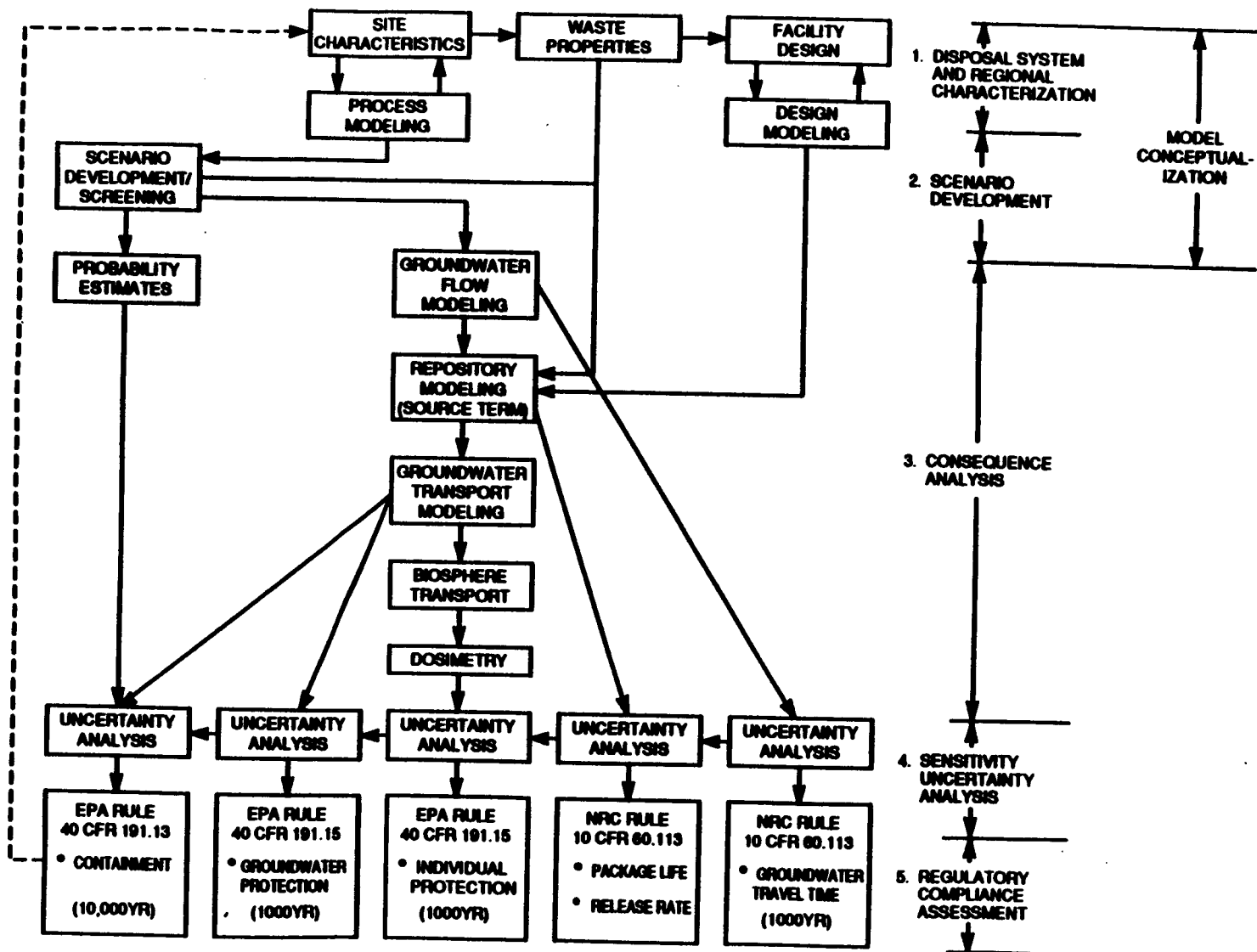
In addition, three other performance measures are used in Section 60.113 to define performance of individual barriers (in contrast to total system). These are: The life of the waste package must exceed specified limits (Section 60.113(1)(A)—Substantially Complete Containment Requirement). Second, release from engineered barriers must be less than specified limits (Section 60.113(1)(B)—Gradual Release Requirement). Third is the groundwater travel time must be greater than specified limits (Section 60.113(2)—Ground Water Travel Time Requirement).

In all, therefore, there are six distinct performance measures. In general, a total system performance assessment code must allow for estimation of the three measures related to 40 CFR Part 191 and may (but not necessarily) also for the other three related to Section 60.113. In

Figure 1-1, the modules which may help in estimating all six performance measures are shown. Also shown in Figure 1-1 are the various steps that one needs to take to complete a performance assessment. These steps include conceptualization of processes as well as assembly of data suitable for inputting in the mathematical models.

1.2 REPORT CONTENT

The various processes coded in *SOTEC* are described in Chapter 2. The processes described include various failure modes for the waste package in Section 2.1 and nuclide release modes in Section 2.2. Release in both the aqueous and the gaseous phase are described. In Chapter 3, the structure of the *SOTEC* code is described. This includes descriptions of various subroutines included in Version 1 of *SOTEC*. Finally, input commands for *SOTEC* are explained in Chapter 4.



1-4

Figure 1-1. Regulatory Performance Measures

2 DESCRIPTION OF PROCESSES IN SOTEC

SOTEC considers processes expected to be important for release of radionuclides from the engineered barriers. The calculations begin with consideration of waste package corrosion. Corrosion failure can occur by general corrosion, pitting, and/or crevice corrosion. Container failure can also occur from time-dependent physical loading, especially as the container becomes weakened from corrosion. Subsequent to container breach, radionuclides are released into the surrounding rock in gaseous and liquid phases. Estimation of liquid and gaseous release requires consideration of chain decay, waste form alteration rate, and transport phenomena. Additionally, spatial variability is considered by *SOTEC*.

Spatial variability is considered in *SOTEC* by conceptualizing the repository volume as made up of a number of disjoint calculational cells. Conditions such as temperature, concentrations of ions in solution, waste loading, and materials properties all may vary among cells. However, the conditions within an individual cell are considered uniform.

Environmental conditions such as temperature, flow rates, and concentrations of individual ions may vary both spatially and temporally. Information concerning this variability may come either from standard input parameters or from output files created by other codes. In order to limit the amount of input data and limit code complexity, the assumption is made that (except for temperature) a separation of variables can be performed between spatial and temporal variations. For each parameter, the input appears as a time variant repository average (or reference) value and a set of load factors for the individual cells. The load factors vary among the cells and are specified in the input deck. The separation of variables is analogous to many analytical solutions for partial differential equations where spatial and temporal variability can be separated mathematically. In *SOTEC*, the separation of variables is made for convenience and no claim is made that the separation is rigorously correct.

The one exception to the above procedure is temperature. Separate time histories for temperature are read into each repository cell. This is done for reasons of compatibility between *SOTEC* and the temperature module which is external to *SOTEC*. For the same reason, the present version of *SOTEC* considers only either 7 or 17 separate cells in the repository. This limitation will be removed in future revisions of the code.

The calculation of the source term from the repository is comprised of three logical segments. The three segments are (i) temperature calculations, (ii) container failure, and (iii) release rate calculations. These subdivisions follow logically from the governing equations because there is no significant feedback among the three. Release rate is assumed not to impact corrosion rates and corrosion or release do not impact repository temperature. For this reason, temperature, corrosion, and release calculations can be performed independently. In the current version of *SOTEC*, corrosion and release calculations only are performed. Temperature calculations are

performed in a separate code. Because of the different time constraints for the corrosion and radionuclide release processes, separate time step loops are used for these two calculations.

2.1 WASTE PACKAGE FAILURES

Calculation of waste package failures due to initial defects, corrosion, mechanical processes and disruptions are considered. For corrosion processes, the corrosion potential is the master variable. For any given metal and set of environmental conditions, the corrosion potential is a major factor influencing the types of active corrosion processes and their rates. The importance of the corrosion potential as a master variable is reflected in the philosophy and implementation of the corrosion routines in *SOTEC*. The current version of *SOTEC* assumes that the corrosion potential is a function of temperature and radiation dose rate.

$$E_{corr} = E_1 + A(T-298) + \beta e^{\lambda t} \quad (2-1)$$

where

- A = empirical factor for temperature effect on corrosion potential
- β = empirical factor for radiolysis effect on corrosion potential (mV)
- λ = average decay rate for gamma emitters in waste
- E_1 = reference corrosion potential (mV)
- R = gas law constant
- T = temperature (Kelvin)
- t = time (years)
- E_{corr} = corrosion potential (mV)

The equation for the corrosion potential assumes that the influence of radiolysis varies linearly with gamma dose rate and that temperature affects the corrosion potential in a linear fashion. The impact of radiolysis appears as a maximum corrosion potential effect at time = 0, and declines with time with radioactive decay of gamma emitters. The dependence of the corrosion potential on temperature and radiolysis is presumed to be obtained from independent theoretical analysis (e.g., Macdonald and Urquidi-Macdonald, 1990) or experimental data and translated into functional forms allowed in *SOTEC*.

Rates of pitting and crevice corrosion depend directly on the corrosion potential. When the potential is elevated above a critical value which is dependent upon the metal, metal history, and solution composition, localized corrosion begins. The critical potential may be referred to as E_{pit} , E_{crev} , or E_{crit} depending upon the context and the type of corrosion involved. When the potential is lowered sufficiently, the localized corrosion cavities generally repassivate. This is sometimes referred to as the repassivation potential or protection potential E_{prot} .

SOTEC currently allows only one critical potential for each of pitting and crevice corrosion and the critical potential is assumed to be a function of temperature but not of solution composition. These expressions for the critical potential will be expanded in future versions of the code.

2.1.1 Uniform Corrosion

General corrosion is assumed to begin when repository temperatures drop below a specified (input) temperature. In the atmosphere, general corrosion is known to become significant when the relative humidity exceeds 80 percent. At the elevation of Yucca Mountain, this corresponds to a temperature of approximately 98°C assuming conservatively that the air is 100 percent water vapor. Once corrosion begins, the corrosion rate is described by a generic power law equation (for more discussion, see Williford, 1991) of the form:

$$r = ae^{\frac{b}{cT}} t^d C_1^e C_2^f \quad T < T^o \quad (2-2)$$

where r is the corrosion rate, a , b , c , d , e , and f are empirical constants, T is absolute temperature, and t is time. Instead of concentrations C_1 , C_2 , etc. of aggressive species, logarithm (to base 10) of the concentrations can be used. In some interpretations of this equation (e.g., Kubaschewski and Hopkins, 1962), b is the activation energy and c is the gas law constant.

General corrosion is assumed to represent a passivated metal and as such is independent of the corrosion potential.

2.1.2 Crevice Corrosion

Crevice corrosion is affected by a number of coupled processes. These include mass transfer, production of metal ions within the crevice and hydrolysis. In literature both the steady-state and transient models for crevice corrosion exist (see Watson and Postlethwaite, 1990 as an example). However, for inclusion in *SOTEC*, such models are too complex and computationally time consuming. A strategy to develop a simple parametric equation for crevice corrosion for use in *SOTEC* has been adopted. In the future versions of *SOTEC*, the parametric equation will be based on the results of complex physics-based models.

Crevice corrosion has two characteristics, initiation time and propagation rate. Initiation time is dependent upon the corrosion potential of the metal exceeding the critical potential for crevice corrosion initiation. After the critical potential is exceeded, an additional period of time is required for destruction of the passive film in the crevice leading to active corrosion. The current version of *SOTEC* conservatively ignores crevice corrosion initiation time. For small crevices, the initiation time will be very short relative to the time scales of

interest for waste isolation. For crevice geometries of a very large size, the initiation time may be of significance but will be difficult to predict with high confidence.

The repassivation potential is taken to be a function of temperature.

$$E_{rc} = E_r + \zeta T \quad (2-3)$$

where

E_{rc} = crevice repassivation potential (mV vs SHE)

E_r = reference potential (mV)

ζ = temperature correction factor.

Subsequent to initiation, crevice corrosion is assumed to penetrate the container at a constant empirical rate, r . The active corrosion rate can be estimated from an equation of the form:

$$r = \frac{i_a w}{\rho z F} \quad (2-4)$$

where, i_a is the active current density, w is the formula weight of the material, ρ is the material density, z is the ion valency, and F is the Faraday constant. *SOTEC* assumes a constant active corrosion rate.

2.1.3 Pitting Corrosion

Episodic evaporation and condensation of water on the surface of waste containers resulting in high concentration of aggressive ions makes them susceptible to pitting corrosion. Mechanistically, pitting corrosion is similar to crevice corrosion and it is possible to develop detailed models (Farmer et al., 1991) for predicting critical pitting potentials or electrochemical conditions within the pit. Alternatively, heuristic stochastic models may also be developed (Henshall, 1991). For development of the *SOTEC* code, a strategy similar to that for crevice corrosion will be followed. Detailed models for pitting corrosion are still under development and a constant propagation rate is assumed for pitting in Version 1.0 of *SOTEC*.

Pitting corrosion is assumed to begin when the critical potential for pitting is reached. The initiation time for pitting is assumed to be very short relative to the time frame of interest, thus pitting is assumed to begin immediately when the critical potential is exceeded. The critical potential is given by Eq. (2-3). The rate of penetration by pitting is assumed to be identical to the rate in crevice corrosion as described by Eq. (2-4).

2.1.4 Failures from Buckling

The only mechanical failure mode for the waste package included in *SOTEC*, Version 1.0, is buckling. Buckling can be caused by the sudden collapse of the borehole plug or the borehole walls, and by dynamic loads caused by seismic activity. For a given design, a time dependent waste package failure criteria due to buckling can be developed. The buckling of waste packages is essentially a three-dimensional phenomenon. Very often, detailed three-dimensional finite-element analysis is required to obtain a failure loading (static and dynamic) envelope for a given design. There are, in the literature, several analytical expressions for the buckling of cylindrical and plate structures. These are primarily for static loads that are symmetrically loaded with respect to a simplified geometry. In this version of *SOTEC*, the buckling loads are based on analytical expressions (Bazant and Cedolter, 1991) with significant simplifying assumptions.

The waste package is assumed to be a long slender cylinder subject to axial and vertical symmetric loads. The cylinder is unconstrained in the radial direction. As a conservative assumption, the stiffening effect of the wasteform inside the cylinder is neglected. The additional stiffness due to end plates is also not considered. The material properties are assumed to be functions of temperature. Only static or quasi-static loads are considered.

The buckling equations are developed for general conditions for various thickness to radius ratios. The approach adopted here considers two aspects of cylinder buckling. In the first evaluation, the load safety factor and critical buckling load are determined for three loading conditions: uniform radial load, uniform axial load, and uniform radial and axial load. Based on the current design configuration, only elastic buckling needs to be considered. Thus, in the second evaluation, only the critical thickness in the elastic buckling criteria is calculated by using the corresponding initial buckling load and safety factor.

2.1.4.1 Case 1 Evaluation

The following equations (for 304L stainless steel) represent the behavior in *SOTEC*, Version 1.0. For safety reasons, the load factor must be applied to the original critical buckling load in order to determine a conservative buckling criteria. In this version, three load factors are determined using the following empirical forms:

- Plastic Buckling

$$F = 1.5 \quad \text{for } (R/t) \leq 5 \quad (2-5)$$

- Elastic-Plastic Buckling

$$F = 1.5 + 1.5 \left[\frac{(R/t) - 5}{(R/t)_e - 5} \right] \quad \text{for } 5 \leq (R/t) \leq (R/t)_e \quad (2-6)$$

- Elastic Buckling

$$F = 3.0 \quad \text{for } (R/t) \geq (R/t)_e \quad (2-7)$$

where

$$(R/t)_e = \sqrt{\frac{E}{4(1-\nu^2)\sigma_{pl}}}$$

- F = load factor
- t = wall thickness, 0.01 (original) m
- R = radius, 0.33 (original) m
- (R/t) = radius to thickness ratio
- $(R/t)_e$ = minimum R/t for elastic buckling
- σ_{pl} = proportional limit stress = $0.8 \times$ yield stress, σ_y
- σ_y = yield stress, MPa = $170 \times [1-1 \times 10^{-3} \times (T-20)]$
- T = current temperature ($^{\circ}\text{C}$)
- E = elastic modulus, MPa = $182000 \times [1-6 \times 10^{-4} \times (T-20)]$
- ν = Poisson's ratio = $0.25 \times [1+4 \times 10^{-4} \times (T-20)]$

The critical buckling loads under uniform radial, uniform axial, and uniform radial and axial external loads are calculated. Buckling formulas for long thin-walled cylinders under uniform radial external loads are derived by:

- Elastic Buckling

$$P_{cr} = \frac{E}{4(1-\nu^2)} \left[\frac{t}{R_o} \right]^3 \quad (2-8)$$

- Elastic-Plastic Buckling

$$P'_{cr} = \frac{E_1}{4(1-\nu^2)} \left[\frac{t}{R_o} \right]^3 \quad (2-9)$$

● Plastic Buckling

$$P''_{cr} = \frac{t}{R_o} \sigma_y \quad (2-10)$$

where

P_{cr} = elastic buckling pressure, MPa

P'_{cr} = elastic-plastic buckling pressure, MPa

P''_{cr} = plastic buckling pressure, MPa

E_t = tangent modulus, MPa = $1600 \times [1 - 6 \times 10^{-4} \times T - 20] \times [1 - 3.9 \times 10^{-4} \times (\sigma - 275)]$ (σ represents the current pressure and T represents temperature °C).

Buckling formulas for long thin-walled cylinders under uniform axial external load (for elastic buckling only) are derived from:

$$P_{cr} = \frac{1}{\sqrt{3}} \frac{E}{\sqrt{1-\nu^2}} \frac{t}{R} \quad (2-11)$$

2.1.4.2 Case 2 Evaluation

As discussed earlier, the elastic buckling is considered more important because of the current design configuration. In the following, the critical thickness for elastic buckling criteria under different loading conditions is calculated. For a uniform radial external load, assuming a uniform radial external load, P_E , and a safety factor of three, buckling occurs when

$$\frac{P_E}{3} \geq P_{cr} \geq \frac{E}{4(1-\nu^2)} \left[\frac{t}{R_o} \right]^3 \quad (2-12)$$

The critical thickness, t_c , can be

$$t_c = \frac{0.32(a)}{1-(a)} \quad (2-13)$$

where

$$a = \frac{4P_E(1-\nu^2)^{1/3}}{3E}$$

For a uniform axial external load in the elastic range with a safety factor of three, buckling occurs when

$$\frac{P_E}{3} \geq P_{cr} \geq \frac{1}{\sqrt{3}} \frac{E}{\sqrt{1-\nu^2}} \frac{t}{R} \quad (2-14)$$

The critical thickness is

$$t_c = \frac{0.32(a)}{1-(a)} \quad (2-15)$$

where

$$(a) = \frac{P_E \sqrt{(1-\nu^2)}}{\sqrt{3}E}$$

For a uniform axial and radial external load, buckling failure occurs when

$$\frac{P_E}{3} \geq P_{cr} \geq \frac{0.92E}{\left[\frac{L}{R}\right] \left[\frac{R}{t}\right]^{2.5}} \quad (2-16)$$

The critical thickness, t_c , can be derived by using numerical analysis. Failure due to buckling is assumed to occur when the calculated critical thickness is greater than the current (corroded) thickness or when the external load exceeds the critical buckling load.

2.1.5 Initial Defective Waste Packages

A proportion of the containers in any cell may be specified to fail at $t=0$. These waste packages represent containers that were initially defective and/or were damaged during emplacement. These data are provided to SOTEC as an external input.

2.1.6 Failures Due to Disruptive Scenarios

SOTEC can handle scenarios in several different ways. Disruptive scenarios are considered in *SOTEC* through the SCENARIO option. The SCENARIO option allows the user to specify the time of occurrence and the number of containers failed for each cell by the specified disruptive scenario. The scenario can be different for each cell. Multiple scenarios can be handled in a single run if they impact different cells. However, only one scenario is allowed per cell. The other failure types, time dependent failure of the container by corrosion or buckling, and initial defectives, are only impacted by scenarios through the loss of containers left in the inventory.

Alternatively, the input parameters for any cell(s) can be perturbed to represent particular scenarios. For example, the water infiltration rate can be varied to simulate change in climate. Modules external to *SOTEC* are being developed to determine the number of failed containers because of various disruptive scenario classes such as fault-movement, damage due to seismic events and volcanism.

2.2 WASTE DISSOLUTION AND RADIONUCLIDE RELEASE

2.2.1 Introduction

Figure 2-1 shows a failed container as represented in the *SOTEC* code. Water is assumed to enter the container and exit again, carrying dissolved radionuclides only. (Colloids are treated as a subcase of dissolved radionuclides and will be discussed further in section 2.7.) The model also accounts for molecular diffusion. Radionuclides are released from the spent fuel waste form at a rate determined by the alteration rate of the uranium dioxide fuel as well as from crud, gap, grain boundary, and cladding. These compartments are listed in Figure 2-2. Radionuclides can leave the container at a rate no greater than that allowed by the solubility of the element in the water. The code allows for radioactive decay of each radionuclide and production from previous elements in the chain. The inventory of the unaltered uranium dioxide is calculated analytically by the Bateman equations (Foster, 1977). The inventory of released radionuclides inside the container is calculated by 4th order Runge-Kutta numerical integration (Press, 1989).

Molecular diffusion is accounted in the code by assuming a spherical source with no advection, and then adding the diffusive component to the advective component of transport out of the container. In the diffusion model, the inner surface of the sphere is held at the concentration or radionuclides in water inside the container. The concentration away from the waste form is held at zero. The diffusion model accounts for molecular diffusion, retardation and radioactive decay, but does not allow for production from a parent radionuclide.

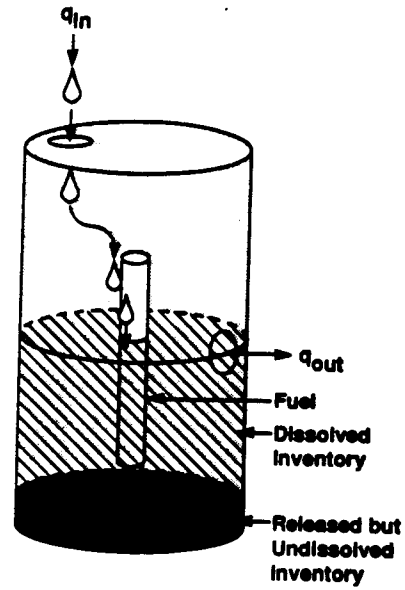


Figure 2-1. Dissolved Radionuclide Release Model

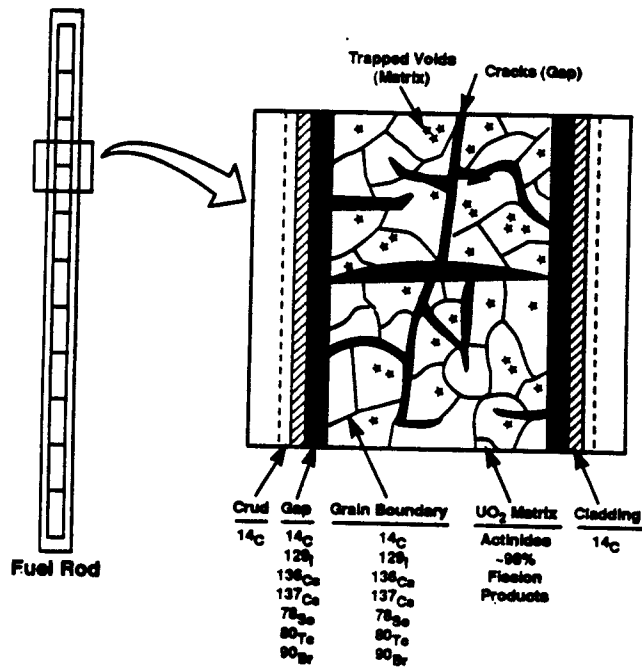


Figure 2-2. Locations of Radionuclides in Spent Fuel

Upon container failure, water is assumed to begin entering the container at a rate q_{in} . There is no outflow until a critical value V_{max} in the container is exceeded, at which point the outflow is:

$$q_{out} = q_{in} k_q \left(\frac{V}{V_{max}} \right)^2 \quad (2-17)$$

where k_q is an arbitrary "weir" coefficient chosen so that the numerical integration of volume occurs smoothly, V is the volume of water, and V_{max} is the maximum volume of water. For the present model, $k_q = 0.8$.

2.2.2 Waste Dissolution

2.2.2.1 Effective Container Failure Time

No waste can leave the container until it is perforated and water can enter and leave. The container fails from damage or corrosion at time T_{FAIL} . The temperature drops below the boiling point at time T_{COOL} . The model assumes that no water can enter or leave the container until $t > T_{COOL}$ and $t > T_{FAIL}$. The effective time at which fuel degradation can begin is given by:

$$T_{FTC} = \text{MAX}(T_{COOL}, T_{FAIL}) \quad (2-18)$$

2.2.2.2 Contact of Water with Fuel

Upon failure, water flowing into the container is assumed to come into intimate contact with the fuel. Radionuclides are released either instantaneously or congruently with the alteration of the UO_2 fuel. Upon contact with water, those radionuclides being released congruently are released at a rate tied to the alteration rate of the UO_2 and the inventory of remaining radionuclides in the fuel. The leaching time (T_{LEACH}) is defined as:

$$T_{LEACH} = 1/(FF \times ALT) \quad (2-19)$$

Where ALT is the fixed alteration rate of the wetted fuel, and FF is the fraction of fuel that is wetted at any time.

No alteration of the spent fuel occurs until T_{FTC} , and release from the spent fuel continues for T_{LEACH} years beyond T_{FTC} . The release stop time T_{LT} is defined as:

$$T_{LT} = T_{LEACH} + T_{FTC} \quad (2-20)$$

Subsequent to alteration of all the spent fuel, at time = TLT, additional release from the waste package may continue from the released but undissolved fraction. Oxidation of the nonwetted fuel is not considered in the liquid release module.

2.2.3 Solubility Limited Release

As the fuel is altered, it releases its radionuclide inventory at the alteration rate into the released but undissolved (RBU) inventory. This assumption is generally called the "congruent release" mode, and assumes that the radionuclides are contained in the matrix of the UO_2 . Radionuclides not contained in the matrix, but collected at the grain boundaries and in the cladding gap, can be assumed to be part of the RBU inventory at the moment of failure.

Release of radionuclides from the container is possible only in the dissolved state. The inventory of all isotopes of an element are collected, and the model determines if the hypothetical concentration of that element is greater than or less than the solubility limit. If the hypothetical concentration exceeds the solubility, then the actual concentration of the container water is held at the solubility limit. Otherwise, the concentration of the container water is the RBU inventory divided by the volume of water, V , contained in the container. There is an assumed minimum volume of water of 1 liter in the container to avoid dividing by zero. The concentration of the element in the container water is reapportioned to the isotopes when determining the release rates.

The mass of each radionuclide remaining in the container water and RBU inventory is calculated by a material balance describing the inputs of:

- radionuclide released from the fuel,
- production from radioactive decay of another radionuclide,
- losses from radioactive decay,
- advective flow,
- diffusion.

2.2.4 Advective Mass Transfer

Advective mass transfer from the container occurs at the rate at which water leaves; i.e., $W_{out,i} = C_i q_{out}$ where C_i is the aqueous concentration of radionuclide i . The radionuclide concentration also is the boundary condition for the diffusive mass transfer from the container.

2.2.5 Diffusive Mass Transfer

The model assumes that mass can be transferred by molecular diffusion, irrespective of convective mass transfer from flowing water into and out of the container.

Diffusive mass transfer is from an assumed spherical source into a domain unaffected by other containers and the flow rate of liquid water through the container and the surrounding rock. The boundary conditions are the concentration of water inside the container at the inner spherical boundary and zero concentration at the outer radius of the spherical boundary. The inner and outer radii, as well as the grid node locations, are input to the code at run time. The differential equation solved is:

$$K \frac{\partial C}{\partial t} = D \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial C}{\partial r} \right) - \lambda KC \quad (2-21)$$

where K is the retardation coefficient, C is the concentration, t is time, D is the coefficient of molecular diffusion, r is radius, and λ is the decay coefficient. The present formulation considers only decay of the radionuclide in question, and does not consider generation of the radionuclide from the chain decay.

The radius of the source sphere for diffusional mass transfer may be chosen so that the surface area is equal to that of the cylindrical container. The spherical geometry with no advection allows a relatively simple solution either numerically or analytically since it is strictly one dimensional. The present formulation for molecular diffusion is an approximation, since the container is not a sphere, and advective mass transfer is not considered in its solution. We believe that the formulation is conservative for the following reasons:

- The concentration of the inner boundary is held constant at the concentration inside the container, neglecting the substantial barrier of the container itself.
- Neglecting the production of the radionuclide from chain decay causes the concentration gradient to become steeper than it would be if chain production were allowed (for those radionuclides in chains).
- Setting the concentration at the outer boundary at zero causes the gradient to be steeper than if the effect of buildup from other containers were considered.

2.2.6 Numerical Solution

The numerical solution to Eq. (2-21) is obtained in a finite difference grid with varying grid spacing. Diffusion coefficient and effective porosity may vary in each finite difference cell. Each solution requires the solution of N linear algebraic equations, where N is the number of increments in the finite difference grid. The equations are tri-diagonal, and several of the terms do not vary with time. The solution of the equations with the Thomas algorithm is quick and efficient (Lapidus, 1962).

The rate of change of mass (m_i) of each radionuclide in the container is determined by the following equation:

$$\frac{\partial m_i}{\partial t} = m_{i-1}\lambda_{i-1} - m_i\lambda_i - C_i q_{out} - w_{di} \quad (2-22)$$

where

$$w_i = m_i \times FF \times ALT$$

The rate of release by molecular diffusion is calculated from the concentration gradient near the container:

$$W_{d_i}(t) = 4\pi r^2 \epsilon D \frac{(C_{o_i} - C_{1_i})}{\Delta R_1} \quad (2-23)$$

where C_{o_i} is the container concentration, C_{1_i} is the concentration at the first grid point, and ΔR_1 is the grid spacing between the innermost two points. The diffusive flux $W_{d_i}(t)$ is subtracted from the inventory at the end of each time step.

The parameters of water influx, q_{in} , alteration rate ALT, wetted fraction FF, diffusion coefficients, and maximum volume of water in the container V_{max} effectively allow the selection of several modes of potential release; e.g., a small V_{max} ensures the so-called "wet drip" case (Pigford, 1992) while a large V_{max} leads to the "bathtub" case. Very low flow would correspond to the "moist continuous" case where the only mechanism for liquid release would be diffusion.

2.2.7 Treatment of Colloids

Colloidal radionuclides are potentially important at the Yucca Mountain site. Radionuclides such as plutonium have large inventories and are long-lived, but have generally low solubilities. Under conditions of a high alteration rate of the fuel matrix, there is evidence that they form colloids when released from spent fuel (Wilson, 1990). *SOTEC*, Version 1.0 does not explicitly handle the release of colloids from spent fuel. A mechanistic treatment of the phenomena leading to growth of colloids from supersaturated liquids will be incorporated into the next update of *SOTEC*. By treating them as distinct species and assuming a large value for their solubilities, colloids can be approximately treated by the current version of *SOTEC*.

2.3 CALCULATIONS OF GASEOUS RELEASE OF ^{14}C

2.3.1 Introduction

^{14}C gas poses a potential problem for compliance with NRC regulations at the partially saturated Yucca Mountain site because in the gaseous form it could migrate quickly to the atmosphere. Its abundance in spent fuel—roughly 10 times the present EPA limit—and its long half life of 5720 years contribute to the potential problem.

^{14}C is generated in nuclear fuel by the neutron activation of ^{13}N , ^{13}C and ^{17}O contained in the fuel and other materials in the reactor core (Van Konynenburg, 1984). The main reservoirs for ^{14}C are the cladding, the cladding/fuel gap and grain boundaries, and the fuel itself. There will be little ^{14}C in vitrified defense waste stored at the site.

Park (1992) estimates the inventories of ^{14}C in BWR and PWR spent fuels shown in Table 2-1.

Table 2-1. Adjusted ^{14}C Content in Spent Fuel (Ci/MTHM) (After Park, 1992)

Type	Burnup Mwd/MTHM	UO ₂	Zirc.	Hardware	TOTAL
BWR	35,000	0.69	0.48	0.13	1.3
PWR	40,000	0.73	0.22	0.26	1.21
Average	----	0.72	0.31	0.21	1.24

Currently, Park (1992) estimates that there will be 70,000 MTHM of spent fuel in the repository, consisting of 22,500 MTHM of BWR fuel and 40,500 MTHM of PWR fuel. The inventory of ^{14}C is estimated to be 78,000 curies.

Van Konynenburg (1991) reports that the chemical forms of the ^{14}C inventories are uncertain, but that they are believed to exist partially as elemental carbon, carbides and oxycarbides in the fuel, and as dissolved carbon and carbides in the metal. The form in the cladding oxide is unknown, but is evidently at least partially in the reduced state.

Most of the ^{14}C in the fuel, cladding and hardware must first oxidize in order to be released in the gas pathway. Although elemental carbon is generally stable at low temperatures, thermodynamics in air favors the formation of gaseous compounds such as CO₂ and methane. Furthermore, carbon does not form protective oxide layers like corrosion-resistant metals. Van Konynenburg (1987) noted that ^{14}C was found to oxidize in an oxidizing environment with a radiation level of 10,000 rad/hr and a temperature of 275°C. Kopp and

Munzel (1990) showed ^{14}C dioxide releases from ^{14}C -doped zirconium sheets at temperatures as low as 200°C , with virtually no radiation, and in an atmosphere of mostly argon with air and water impurities. In very pure argon, however, there was virtually no release of ^{14}C . The dependence of release on O_2 content indicates that carbon was undergoing oxidation in the impure argon. While kinetic considerations might restrict the formation of gaseous compounds of the ^{14}C in the fuel, the model conservatively assumes that any ^{14}C available to become oxidized is converted to $^{14}\text{CO}_2$.

The ^{14}C gas release component of *SOTEC* considers that $^{14}\text{CO}_2$ is released from the fuel, cladding and hardware compartments upon failure of the container. Several of the release mechanisms depend on the presence of oxygen. While there may be small quantities of oxygen initially present in the container as impurity in the inert gas or as water, most of the oxidation would not occur prior to container failure.

The ^{14}C release model makes the following conservative and simplifying assumptions:

- There can be no release of gas prior to container failure;
- Spent fuel has no protection from the cladding;
- Once the container fails, it provides no resistance to the inflow of air or the outflow of CO_2 ;
- Upon container failure, a "prompt release fraction" of approximately 2.5 percent (Van Konynenburg, 1991) of the total inventory is released from the container instantaneously;
- Upon container failure, cladding and structural metals undergo oxidation, and release ^{14}C at the rate that the metal oxidizes;
- Upon container failure, oxygen diffuses into the spent fuel and oxidizes the ^{14}C to $^{14}\text{CO}_2$, which then diffuses out to the container.

The bases for the assumed release mechanisms are discussed in the following paragraphs.

2.3.2 Prompt Release Fraction

Upon failure of the container and cladding (no credit taken for the cladding integrity), a portion of the total ^{14}C inventory estimated at about 2.5 percent (Van Konynenburg, 1991) would be released quickly (i.e., within a few hundred years). This inventory represents the ^{14}C held loosely on the cladding and in the cladding/fuel gap and grain boundaries.

Experiments on ruptured spent fuel indicate that upon cladding failure, 0.5 percent of the inventory is released from the fuel/cladding gap (Wilson, 1990). The initial layer of oxide and crud on the cladding is the other readily available inventory of loosely held ^{14}C . This layer holds a relatively elevated level of ^{14}C , some produced by activation in situ but some probably picked up from the circulating water within the reactor. The chemical form of this ^{14}C is uncertain, but experiments in which cladding was heated indicate that the release rate of $^{14}\text{CO}_2$ appears to be controlled by a diffusion process from a layer of finite thickness.

Smith and Baldwin (1989) showed that as much of 2 percent of the spent fuel ^{14}C inventory was released in zircaloy cladding heated to 350°C for 8 hours. The release of ^{14}C from the oxide layer for short term experiments is shown in Figure 2-3. The release rate is consistent with diffusion out of a layer of finite thickness with an activation energy of between 19 and 25 Kcal/mole. However, the rate of release depended on the presence of air. In argon with a trace of air (50 ppm oxygen), release rates were lower by a factor of about 10, indicating that the carbon was in a reduced state and had to be oxidized before being released.

If it is considered that the release of ^{14}C from the cladding oxide layer is governed by the diffusion mechanism, it is possible to estimate the upper bound of release rate for the temperature range of interest, approximately 350 to 100°C . Molecular diffusion through the film of thickness $2L$ can be expressed by the partial differential equation:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (2-24)$$

where D is the diffusion coefficient, C is the concentration of $^{14}\text{CO}_2$, and x is the distance measured from the edge of the cladding toward the center. The boundary conditions that apply to Eq. (2-24) are:

$$\begin{aligned} C &= C_o \text{ at } x = 0 \\ \partial C / \partial x &= 0 \text{ at } x = L \end{aligned}$$

Equation (2-24) has the following solution (Carslaw and Jaeger, 1959):

$$\frac{C}{C_o} = 1 - \frac{4}{\pi} \sum_{n=0}^{\infty} \frac{-1^n}{2n+1} e^{-\frac{(2n+1)^2 \pi^2 \tau}{4}} \cos\left(\frac{(2n+1)\pi \xi}{2}\right) \quad (2-25)$$

where $\tau = Dt/L^2$, $\xi = x/L$ and L is $1/2$ the cladding thickness.

Results of Eq. (2-25) in terms of the dimensionless distance x/L , C/C_o and Dt/L^2 are shown in Figure 2-4. For $Dx/L^2 = 1.5$, virtually all $^{14}\text{CO}_2$ would have diffused out of the

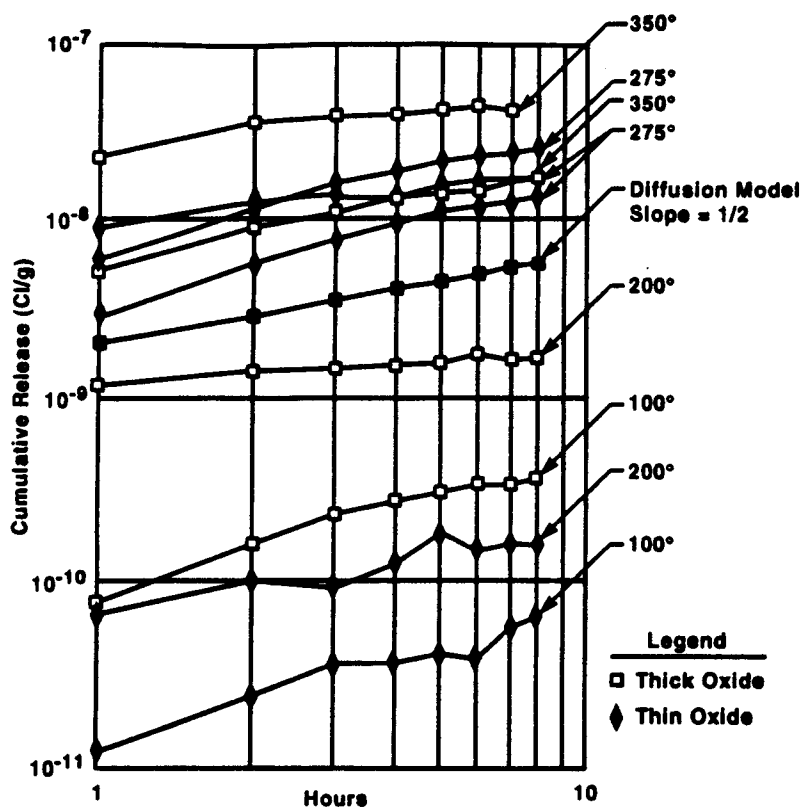


Figure 2-3. ¹⁴C Release from Oxide Layer (after Smith and Baldwin, 1989)

system. The experimental data of Smith and Baldwin indicate virtually complete release of ¹⁴C from the oxide layer in 8 hours at 350°C, and estimate an activation energy of between 19 and 25 kilocalories per mole. Since the fraction removed from the sample is related to the parameter Dt/L^2 , we can estimate the time for an equal degree of removal for lower temperatures. Time to reach an equivalent concentration profile is inversely proportional to the diffusion coefficient; i.e.,

$$\frac{D_1 t_1}{L_1^2} = \frac{D_2 t_2}{L_2^2} \quad (2-26)$$

or

$$t_2 = t_1 \frac{D_1}{D_2} \quad (2-27)$$

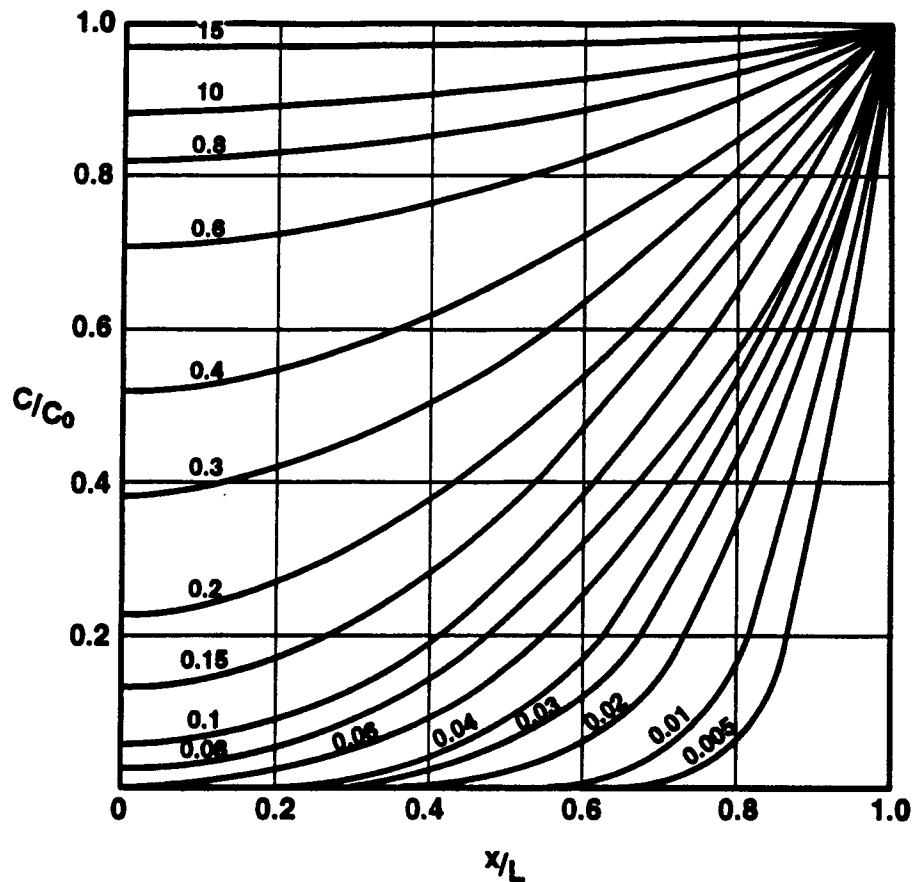


Figure 2-4. Evaluation of Eq. (2-25) (Carslaw and Jaeger, 1959). The Numbers on the Curves are the Values $\frac{Dt}{L^2}$

The ratio D_1/D_2 can be related by the Arrhenius equation to be:

$$\frac{D_1}{D_2} = e^{-\frac{E}{R}\left(\frac{1}{T_2} - \frac{1}{T_1}\right)} \quad (2-28)$$

The time for an equivalent release at lower temperatures is illustrated in Table 2-2, assuming $t_1 = 8$ hours at 350°C for $E = 19$ and 25 Kilocalories/mole. R is the gas law constant and the subscripts refer to the two different temperatures. Equation (2-25) was used to estimate t_2 for a range of activation energies and temperatures.

Table 2-2. Diffusion of $^{14}\text{CO}_2$ from Surface Oxide

T°C	t ₂ , yrs E=19 Kcal/mol	t ₂ , yrs E=25 Kcal/mol
350	0.00091	0.00091
300	0.0035	0.0053
250	0.017	0.043
200	0.12	0.55
150	1.3	13
100	27	690
75	169	7800

Results presented in Table 2-2 indicate that for temperatures as low as 75°C, release of the ^{14}C by diffusion out of the oxide film would be virtually complete within 10,000 years, and possibly much less time. Furthermore, there may be sufficient oxygen present in an unfailed container as an impurity or dissociated water to allow partial escape of $^{14}\text{CO}_2$ from the cladding into the container prior to failure. On the other hand, there are few direct data on $^{14}\text{CO}_2$ releases from zirconium at low temperature.

The observation that release from the cladding depended on the presence of oxygen indicates that the ^{14}C may be in a reduced state in the oxide film. The oxidation to $^{14}\text{CO}_2$ might depend on the combined effects of temperature, oxidizing environment and ionizing radiation. Considering the uncertainties, we must assume a conservative model for release of ^{14}C from the oxide layer. In the present model, the entire quantity of ^{14}C in each container contained in the "prompt release fraction," 2.5 percent of the spent fuel inventory, is assumed to be released to the geosphere at the time of container failure.

2.3.3 Cladding Integrity

Protection of the fuel by the cladding has been ignored in the Phase 2 source term models for liquid and gaseous release. We believe this to be a conservative assumption, but it is difficult to prove to the contrary that the cladding would survive for long periods of time. Factors in favor of cladding integrity are:

- Cladding is usually a highly corrosion-resistant zirconium alloy, but some of the older fuel was clad in stainless steel.

- Most fuel rods emplaced in the containers will be intact, but some will have a small number of undetected pinholes or cracks in the cladding, generally considered to be lower than 1 percent.

Factors in favor of loss of cladding integrity are:

- Under circumstances of sufficiently high temperature in an oxidizing environment to cause oxidation of the fuel to lower-density U_3O_8 , fuel rods with cladding defects are known to split further. At temperatures above 250°C , fuel oxidation was high enough to cause propagation of defects, but below 250°C the defects were not observed to propagate (Einziger and Kohli, 1984). Those defected rods that do split allow oxygen to further oxidize fuel, causing the defect to spread down the length of the fuel rod. The container must first fail to allow oxidation of the fuel and further splitting of the fuel rods.
- While cladding is highly corrosion resistant, there are factors such as hydride reorientation that could lead to failure.
- Cladding could fail by mechanical breakage caused by handling errors, container buckling or earthquakes.
- The fuel rods will be pressurized up to 50 atmospheres, so the cladding will be under stress.

The *SOTEC* Version 1.0 model assumes that the cladding does not protect the UO_2 from oxidation. The protection the cladding may provide will be included as an enhancement to the next version of *SOTEC*.

2.3.4 Release of ^{14}C from Oxidation of Cladding

The cladding oxidation layer is about 10 microns thick initially. The cladding metal itself is on the order of 0.5 to 1.0 mm thick, and contains the bulk of the ^{14}C in the cladding. The zirconium alloy generally used for cladding is highly corrosion resistant, but in an oxidizing environment with elevated temperatures, could oxidize. Upon oxidation, the ^{14}C contained in the metal as elemental carbon, carbides and oxycarbides could be released as $^{14}\text{CO}_2$.

Studies of zircaloy degradation in air have been performed in connection with dry-cask storage (Einziger and Kohli, 1984). The gain in weight of zircaloy samples attributed to oxidation has been determined to proceed in two modes. The first mode, known as the "pretransition" phase, shows weight gain proportional to the cube root of time until the oxide thickness reaches a critical value. The "post-transition" phase shows a linear weight gain for

oxide layer thickness greater than the critical value. It is most likely that the cladding reaches post-transition within the reactor core.

Post-transition cladding oxidation appears to follow an Arrhenius relationship (Gazarelli, 1980), i.e.:

$$\frac{\Delta w}{\Delta t} = A e^{-\frac{E}{RT}} \quad (2-29)$$

where A is the coefficient, mg/dm^2 , E = activation energy, calories/mole, R = gas constant, $1.987 \text{ cal}/\text{mole}/^\circ\text{K}$ and T = absolute temperature, Gazarelli presents several empirical formulas for the post-transitional weight gain due to oxidation. Table 2-3 gives the coefficients A and E for four empirical formulas for post-transition oxidation. Figure 2-5 compares the empirical formulas for the rate of oxide growth (K_L)(mg/dm^2).

Table 2-3. Zircaloy Oxidation Coefficients (Post Transition) (Gazarelli, 1990)

Name	A, mg/dm^2	E, cal/mol	Comments
Hilner	1.12E8	24895	
Van der Linde	2.3E9	28613	
Dyce	6.53E9	29915	
Gazarolli	2.21E9	28200	
Dalgaard	1.87E7	22200	no pretransition

Figure 2-5 shows calculated oxide thickness to 10,000 years for the five formulas evaluated for one of the hotter containers with an assumed failure time of 100 and 500 years respectively. Calculated oxide thickness is highest for the Dalgaard formula. The release model for cladding oxidation conservatively adopts the Dalgaard formula and makes the following additional assumptions:

- $^{14}\text{CO}_2$ is released from the zircaloy at the rate that the metal oxidizes. Zirconium is a strong oxygen getter, so oxygen concentrations available to oxidize carbon would be limited until the zirconium oxidizes.
- Other irradiated structural metal buried with the waste (other than the container itself) is conservatively included with the zircaloy cladding as a source of ^{14}C .

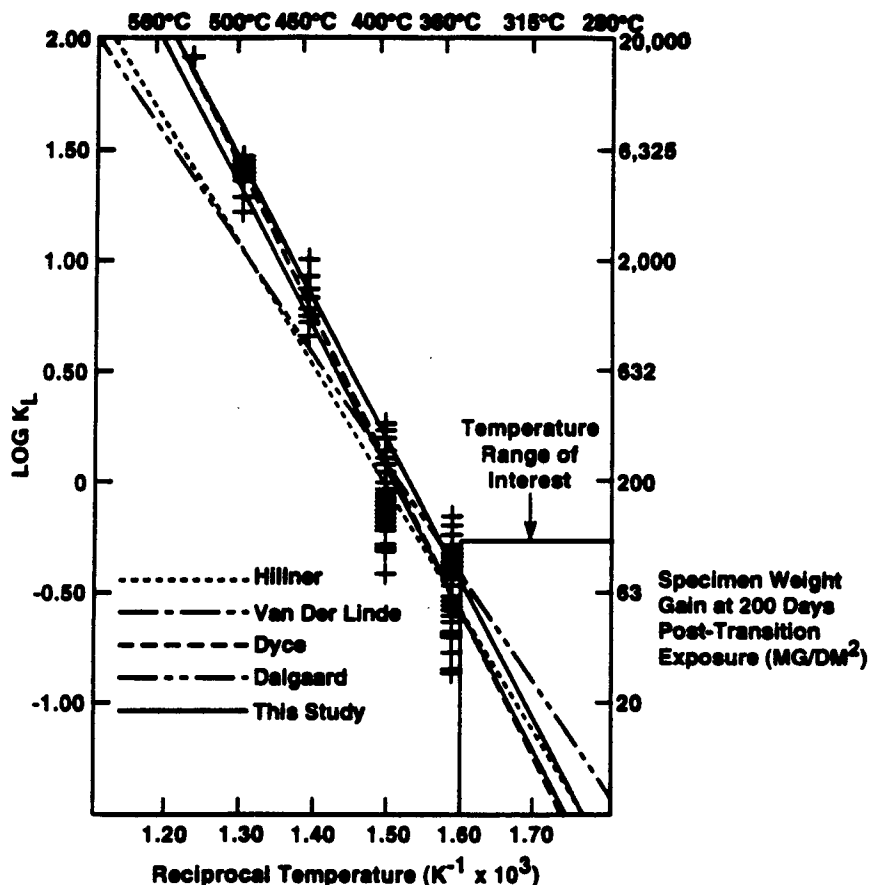


Figure 2-5. Comparison of Post-Transition Corrosion Models with Data (Gazarelli, 1980)

2.3.4.1 Release from UO_2

The largest inventory of ^{14}C is contained in the spent fuel tied up as solid solutions, elemental carbon, carbides and oxycarbides.

- UO_2 oxidizes at a rate controlled by the diffusion of oxygen through the grain boundaries and oxide film.
- The oxygen concentration at the oxide/fuel boundary will be zero, because all oxygen is being consumed by fuel oxidation.
- ^{14}C can oxidize only after the fuel oxidizes.

- $^{14}\text{CO}_2$ will be released from the spent fuel at the rate that the spent fuel oxidizes.
- $^{14}\text{CO}_2$ must diffuse outward through the oxide film and grain boundaries.
- Concentration profiles for both oxygen and $^{14}\text{CO}_2$ are at steady state, although the position of the boundary changes with time.

The last assumption will be relaxed to include unsteady state diffusion at a later stage of development.

Investigations of the oxidation of grains of spent fuel indicate that the rate of oxidation is controlled by at least diffusion through two barriers; the grain boundaries and the film of oxidized fuel surrounding each grain (Einziger, 1988). Irradiated spent fuel contains numerous cracks which allow gas to easily permeate the mass. The smallest scale of interest is the individual grains, which are on the order of 10 to 20 microns in diameter.

2.3.4.2 Fuel Oxidation Model

The present model assumes that the fuel mass in each container can be represented by two concentric spheres as shown in Figure 2-6. The outer sphere represents the diffusion barrier for the grain boundaries, and has the equivalent spherical diameter of a fuel fragment, about 0.2 cm. The inner sphere represents the diffusion barrier through the oxide film on the surface of the fuel grain, taken as a sphere with a diameter of about 20 microns. The boundary conditions for the model are zero oxygen concentration at the fuel/oxide interface and atmospheric oxygen concentration at the outer diameter.

Diffusion of oxygen through the fuel grain will be governed by the following partial differential equation:

$$\frac{\partial C}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 D \frac{\partial C}{\partial r} \right) \quad (2-30)$$

where C = oxygen concentration, t = time, r = radius from center of sphere, D = diffusion coefficient.

The diffusion coefficient D is a function of temperature, and differs for the oxide layer and grain boundary layers. The boundary between the spent fuel grain and the oxide layer changes as the oxide layer grows, making this a moving boundary problem. The rate that the boundary recedes is governed by the diffusion rate of oxygen at the interface:

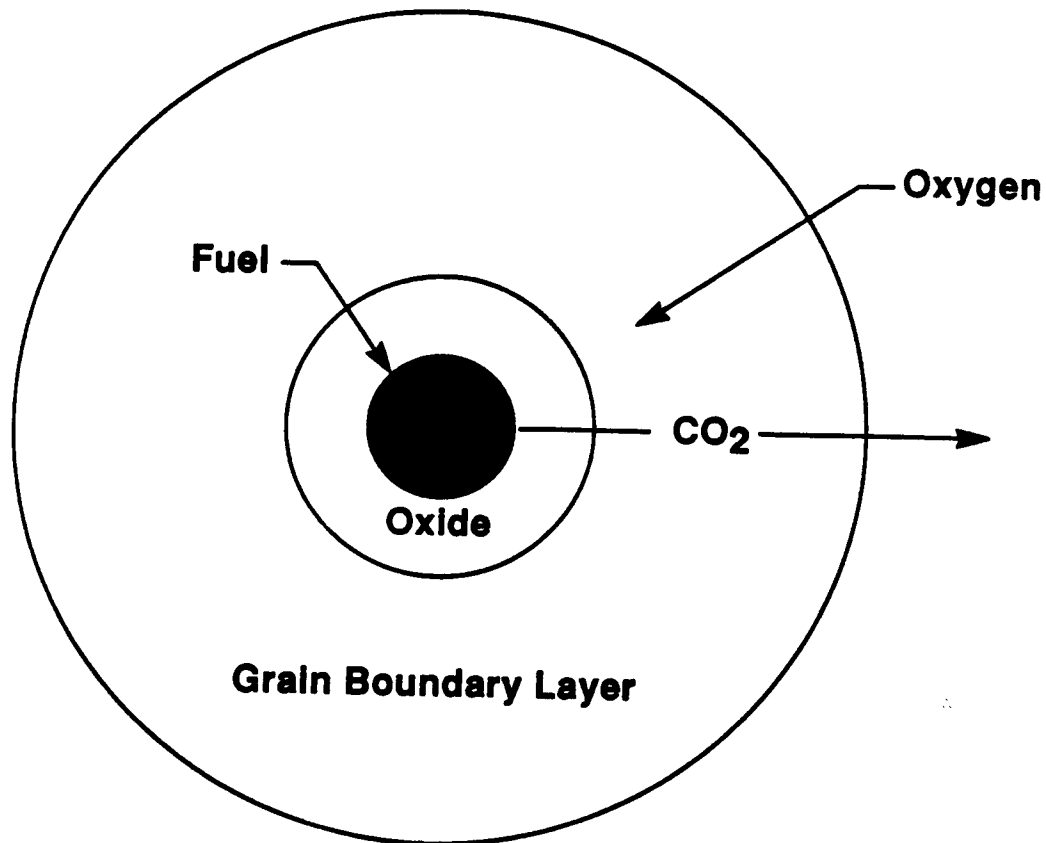


Figure 2-6. ¹⁴C Gaseous Release Model

$$\frac{\partial R}{\partial t} = N_{ox} \frac{D}{\rho_{ox}} \left. \frac{\partial C}{\partial r} \right|_{r=r'} \quad (2-31)$$

where r' = radius of fuel/oxide interface, D = diffusion coefficient in the oxide, cm^2/yr , and ρ_{ox} is the density of the oxide, moles/cm^3 . The term N_{ox} is the conversion factor for UO_2 in terms of moles UO_2 oxidized per mole O reaching the boundary. X-ray diffraction analyses of samples of oxidized fuel indicate that for the temperature ranges likely to be encountered in the repository, most of the oxide formed will be U_4O_9 , although stoichiometrically, the oxide appears to be U_3O_7 , because of excess oxygen loosely held by the lattice (Einzig, 1992). For the purposes of the present analysis, N_{ox} will be taken to be 3, i.e., 3 moles of UO_2 will be oxidized by one mole of O ($1/2$ mole O_2).

2.3.4.3 $^{14}\text{CO}_2$ diffusion model

The model for the release of $^{14}\text{CO}_2$ is similar to the UO_2 oxidation model. Diffusion of $^{14}\text{CO}_2$ through the fuel grain boundary layer and oxide layer will be governed by the following partial differential equation:

$$\frac{\partial C_c}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 D \frac{\partial C_c}{\partial r} \right) - \lambda C_c \quad (2-32)$$

where $C_c = ^{14}\text{CO}_2$ concentration, $t =$ time, $r =$ radius from center of fragment and λ is the decay coefficient for ^{14}C . The outer boundary conditions are:

$$C_c = 0 \text{ at } r = R_1$$

At the inner boundary, $^{14}\text{CO}_2$ enters the oxide layer from the just-oxidized fuel. The gradient of $^{14}\text{CO}_2$ concentration is adjusted to account for diffusion:

$$\left. \frac{\partial C_c}{\partial r} \right|_r = \frac{3M}{4\pi r_o^3 D_o} \frac{dr}{dt} e^{-\lambda t} \quad (2-33)$$

where M is the initial inventory of ^{14}C , $D_o =$ diffusion coefficient in the oxide layer. The ^{14}C diffusion model depends on the UO_2 oxidation model to provide the position of the moving boundary and the source flux of $^{14}\text{CO}_2$ at the inner boundary as the oxide layer grows. Release of $^{14}\text{CO}_2$ at the outer boundary of the fragment is calculated from the concentration gradient at that boundary:

$$q_{14c} = 4\pi r_1^2 \left. \frac{\partial C_c}{\partial r} \right|_{r_1} \quad (2-34)$$

where $q =$ the rate of release from the fragment, curies/year and $D_1 =$ the diffusion coefficient in the boundary diffusion layer.

A simple semi-analytical solution to Eq. (2-30) was developed which depends on the assumption that the O and $^{14}\text{CO}_2$ gradients were at steady state.

2.3.4.4 Steady State Fuel Oxidation Model

At steady state, Eq. (2-32) for O concentration reduces to:

$$\frac{\partial}{\partial r} \left(r^2 \frac{\partial C}{\partial r} \right) = 0 \quad (2-35)$$

which has the general solution:

$$C = \frac{A}{r} + B \quad (2-36)$$

where A and B are constants.

The boundary condition for oxygen is atmospheric concentration at the surface of the sphere and zero concentration at the boundary between the U_3O_7 and the fuel:

$$\begin{aligned} C &= C_o, \text{ at } r = R_1 \\ C &= 0, \text{ at } r = r' \end{aligned}$$

Between r' and R_o , diffusion coefficient D_o applies, and between R_o and R_1 , D_1 applies. For a composite hollow sphere between r and R_1 , therefore, the mass rate of oxygen transport to the surface of the fuel at steady state is:

$$\text{flux} = 4\pi D_o r^2 \left. \frac{dC}{dr} \right|_{r=r'} = \frac{4\pi(C_o - C_1)}{\left[\frac{1}{D_o} \left(\frac{1}{r'} - \frac{1}{r_o} \right) + \frac{1}{D_1} \left(\frac{1}{r_o} - \frac{1}{r_1} \right) \right]} \quad (2-37)$$

The rate of growth of the film is related to the flux of oxygen. In terms of the oxidation rate of the fuel surface, Eq. (2-37) becomes:

$$\frac{dr_o}{dt} = \frac{(C_o - C_1)N_{ox}}{\left[\frac{1}{D_o} \left(\frac{1}{r'} - \frac{1}{r_o} \right) + \frac{1}{D_1} \left(\frac{1}{r_o} - \frac{1}{r_1} \right) \right] r^2 \rho_u} \quad (2-38)$$

where C_o = the concentration at the surface of the fuel, taken here to be zero, and C_1 = concentration of oxygen at the surface of the fuel, taken here to be the volumetric concentration in the atmosphere, 0.2 moles/22,400 cm³.

The rate of growth depends on r_0 and the diffusion coefficients D_0 and D_1 which are functions of fuel temperature. The temperature of the fuel is estimated from the temperature of the container (generated from program TEMPER externally) corrected with an empirical formula determined from a calculational exercise on fuel temperature shown in Figure 2-7 (O'Neal, 1984). The growth of the oxide layer is calculated by numerically integrating Eq. (2-38) with a Runge-Kutta algorithm for the time-varying properties.

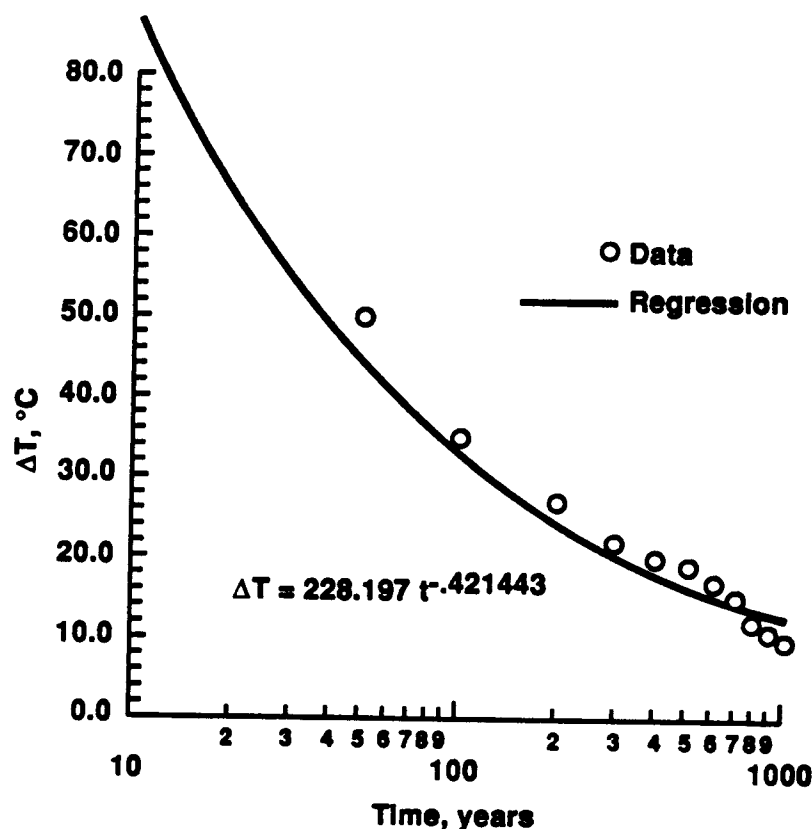


Figure 2-7. Fit of Temperature Difference between Container Skin and Maximum Fuel. Vertical Emplacement, 48.4 kW/acre 3.3 kW/container at 10 yr.

2.3.4.5 Steady-State $^{14}\text{CO}_2$ Transport Model

$^{14}\text{CO}_2$ generated at the oxide/fuel interface must diffuse out through the oxide and grain boundary layers to leave the fuel fragment. In the steady state assumption, the gradient of $^{14}\text{CO}_2$ in the radial direction is steady, but because the ^{14}C decays radioactively, the gradient will be somewhat different than for a nondecaying substance. While it is possible to have a separate model for diffusion of $^{14}\text{CO}_2$ at steady state, it would be much more

complicated (involving series of Bessel functions) than the model necessary for the diffusion of the nondecaying oxygen.

In order to test the need to include the effects of ^{14}C decay, the simpler case was solved for diffusion of $^{14}\text{CO}_2$ diffusing through a one-dimensional uniform oxide layer of thickness L . The boundary condition of this problem are concentration of $^{14}\text{CO}_2 = C_1$ at $x = 0$, and 0 at $x = L$, with diffusion coefficient D . The differential equation at steady state that describes the concentration of $^{14}\text{CO}_2$ in the layer would be:

$$\frac{d^2C}{dx^2} - \lambda C = 0 \quad (2-39)$$

where λ is the decay coefficient. For a nondecaying substance at steady state, the flux F would be simply:

$$F = DC_1/L \quad (2-40)$$

For a decaying substance, the solution of Eq. (2-39) in terms of the gradient at $x = L$ gives the flux F' :

$$F' = \frac{2DC_1\sqrt{a}}{e^{-\sqrt{a}L} - e^{\sqrt{a}L}} \quad (2-41)$$

where $a = \lambda/D$.

The ratio of the approximate model (no decay) to the complete steady state model with decay is therefore:

$$\frac{F}{F'} = \frac{e^{-\sqrt{a}L} - e^{\sqrt{a}L}}{2L\sqrt{a}} \quad (2-41)$$

For parameter values typical of the present work, Eq. (2-41) was evaluated for conditions likely to lead to the largest difference between the model with and without decay, diffusion through the largest possible thickness of 0.001 cm of oxide at the relatively low temperature of 100°C. At this temperature, $D_o = 5.7\text{E-}8$ cm²/year for the parameters used in the current example. The decay coefficient (λ) = 1.21E-4/year. Therefore, Eq. (2-41) yields a ratio $F'/F = 1.002$. From this approximate analysis we can conclude that the effects of radioactive decay of ^{14}C on the flux can be neglected, and therefore the rate of release of $^{14}\text{CO}_2$ is equal to the rate at which the gas is liberated at the surface of the fuel.

2.3.4.6 Overall Model Conservatism

We believe the ^{14}C release rate model to be conservative for the following reasons:

- The protection of the fuel from oxidation afforded by the cladding is ignored in phase 2. Zircaloy is a highly corrosion resistant material, and it is likely that it would protect the fuel following container failure for a substantial period of time. Protection of the fuel even for a few hundred years would have a substantial impact on the calculated release rate because the greatest potential for release is the period during which the fuel has the highest temperature.
- Most of the ^{14}C in the fuel, cladding and hardware is likely to be in a reduced state, and must first become oxidized to be released in the gas pathway, although thermodynamics in air favors the formation of gaseous compounds such as CO_2 and methane. While kinetic considerations might restrict the formation of gaseous compounds of the ^{14}C in the fuel, the model conservatively assumes that any ^{14}C available to be oxidized is converted to $^{14}\text{CO}_2$.
- The model assumes there is no resistance of $^{14}\text{CO}_2$ once it is released from the fuel fragment; i.e., there is no resistance for diffusion through the long length of the failed fuel rod, or through pinhole failures of the container. This conservatism is relatively less important for the far-field release calculations because of the long periods involved; that is, diffusion and barometric pumping could allow virtually all $^{14}\text{CO}_2$ to escape to the geosphere, even for relatively small failure holes. Resistance to release from the container might be more important in evaluating compliance with the NRC subsystem requirements of 10 CFR 60.113 which are more sensitive to short-term rates of release.

2.3.4.7 Parameter estimation and model verification/validation

The ^{14}C source term model is based on an abstraction of many complex processes. Therefore, the model can only be compared to data on UO_2 oxidation. It relates UO_2 oxidation to diffusion through two layers of material and outward diffusion of $^{14}\text{CO}_2$ through the same two layers. The simplifying assumptions taken in the model are:

- The fuel is represented by concentric spheres, of a single set of dimensions; i.e., the irregular shape of the fragments and grains is not taken into account.
- The current version of *SOTEC* allows only steady state diffusion.
- No effects of the container or cladding on the diffusional processes are taken into account.
- The increase in surface area caused by oxidation of the grains is not taken into account.
- While there are some direct data on release of $^{14}\text{CO}_2$ from cladding and the grain/gap inventory, releases from the largest inventory in the fuel itself are lacking. There are data on UO_2 oxidation, but there are apparently no data available for release of $^{14}\text{CO}_2$ from fuel grain oxidation.

Nevertheless, the model was considered to be realistic enough that the parameters necessary for its implementation could be obtained from experimental data.

2.3.4.8 Data on Spent Fuel Oxidation

Several investigators have collected data on the oxidation in air of spent reactor fuel in connection with intermediate storage of spent fuel (Einziger, 1984, 1988, 1991a, 1991b, 1992; Woodley, 1989; Thomas, 1991). The main concern of these investigators was the degradation of the waste form for fuel in contact with oxygen at temperatures of several hundred °C (e.g., dry storage in air). Consequently, the experimental programs concentrated on the physical changes to the fuel resulting from oxidation in failed fuel rods rather than the releases of radionuclides.

Quantitative data on fuel oxidation in air was basically of four types; Thermal Gravimetric Analysis (TGA), dry bath analysis, ceramography and x-ray crystallography. In TGA, samples of spent fuel were exposed to air at fixed temperatures, and the gain in sample weight due to oxidation was recorded continuously with an analytical balance. In the dry bath tests, samples were kept in small crucibles held at fixed temperatures in an aluminum block, and weighed periodically outside the apparatus. The advantage of the dry bath tests were that they could be performed for very long times, on the order of years, while the TGA experiments were limited to shorter periods, up to a few months.

Ceramography is the inspection of thin slices of spent fuel to visually observe the growth of oxide films around individual fuel grains. These observations gave

valuable, quantitative information about the sizes of fuel grains necessary for the implementation of the mathematical model and the rate of growth of the oxide thickness. In addition, ceramography gave qualitative information about the mechanisms of oxidation; e.g., the fact that the film of oxide appears to be growing at a consistent rate throughout the sample indicated that diffusion of oxygen through grain boundaries and cracks probably was much faster than the diffusion across the oxide layer itself.

X-ray crystallography gave quantitative information on the chemical species of the oxide formed at different temperatures. Among the more interesting indications of x-ray crystallography was the observation that for temperatures below about 200°C, the oxide formed was primarily U_4O_9 , even though it appeared to be U_3O_7 stoichiometrically (Einziger, 1992).

2.3.4.9 *Uses of the data for parameter identification*

Aside from identifying the form of the model, only the quantitative data on grain size, growth of oxide film thickness and sample weight were used to determine the model parameters for fuel oxidation. Data on sample weight gain and film thickness at fixed temperatures were put into the form of "conversion fraction" of UO_2 to U_3O_7 versus time. The model was then exercised to predict the conversion of UO_2 grains to U_3O_7 versus time. The only model parameters that could vary were:

- Grain size (i.e., radius of inner sphere);
- Fragment size (i.e., radius of outer sphere);
- Diffusion coefficient through oxide layer at the reference temperature of 200°C;
- Diffusion coefficient through the grain boundary layer at the reference temperature of 200°C;
- Activation energy for diffusion coefficient (considered to be equal for both layers).

The parameter identification does not at the present time take into account any differences between fragment sizes, grain sizes or types of fuel.

Data on fuel conversion were collected in eight sets from TGA, Dry Bath and ceramography, representing experimental periods from 700 to 12,000 hours, with the lower temperatures represented by the longer periods. Although data were available on whole fuel fragments as well as crushed fuel, only the whole-fragment data were used for the parameter

identification for Phase 2 because these samples were more like the actual spent fuel that would be in the repository.

The parameter identification was performed by manually varying the parameters and observing the agreement between predicted and observed conversion for the eight data sets. This manual process was facilitated by a computer program, UOXB, which exercised the model specifically for this purpose and represented the results graphically on the monitor screen. Parameters giving the best comparisons are given in Table 2-4.

Table 2-4. Model Parameters from Manual Identification

MODEL PARAMETER	VALUE
Grain radius, cm	0.001
Fragment radius, cm	0.1
Ref Film Diffusion Coef., cm ² /yr	5.256E-4
Ref. Grain Diffusion Coef, cm ² /yr	5.942E-3
Reference Temperature, C	200
Activation Energy, Kcal/mol	32

Graphical results of the model/data comparison are given in Figure 2-8(a) through (h). While there is a degree of scatter in the data, the model prediction is quite reasonable for the parameters chosen. Bear in mind that only one set of parameters was used to represent what are certainly nonhomogeneous samples. More identification will be performed in order to estimate the ranges of parameters and the correlation of parameters that encompass the range of the conversion data. These ranges and correlations will be necessary for the Latin Hypercube analyses.

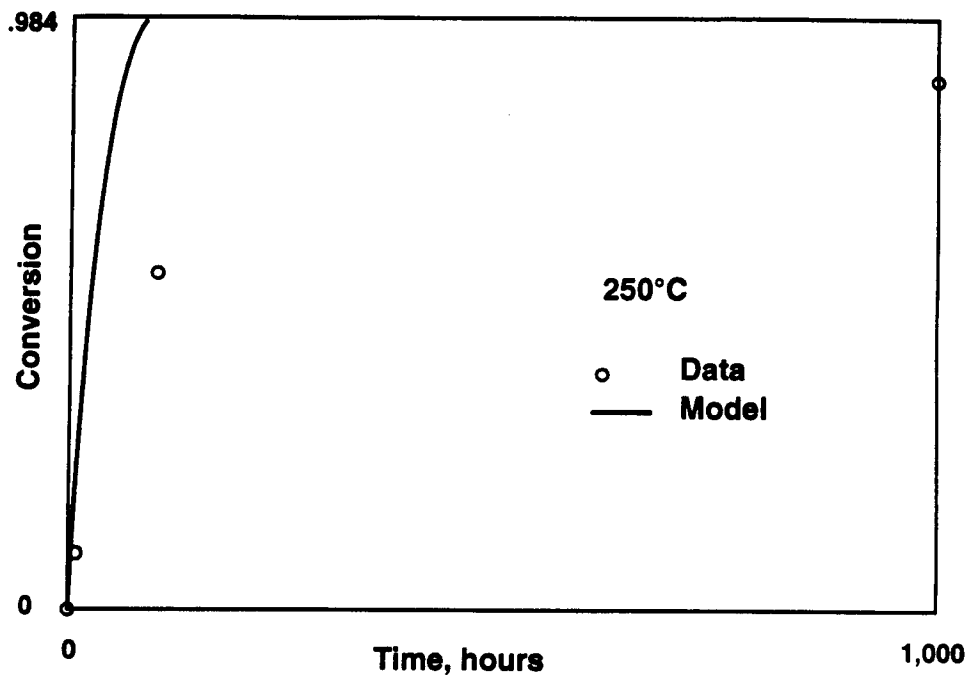


Figure 2-8(a). Model-Prototype Comparison, 250°C

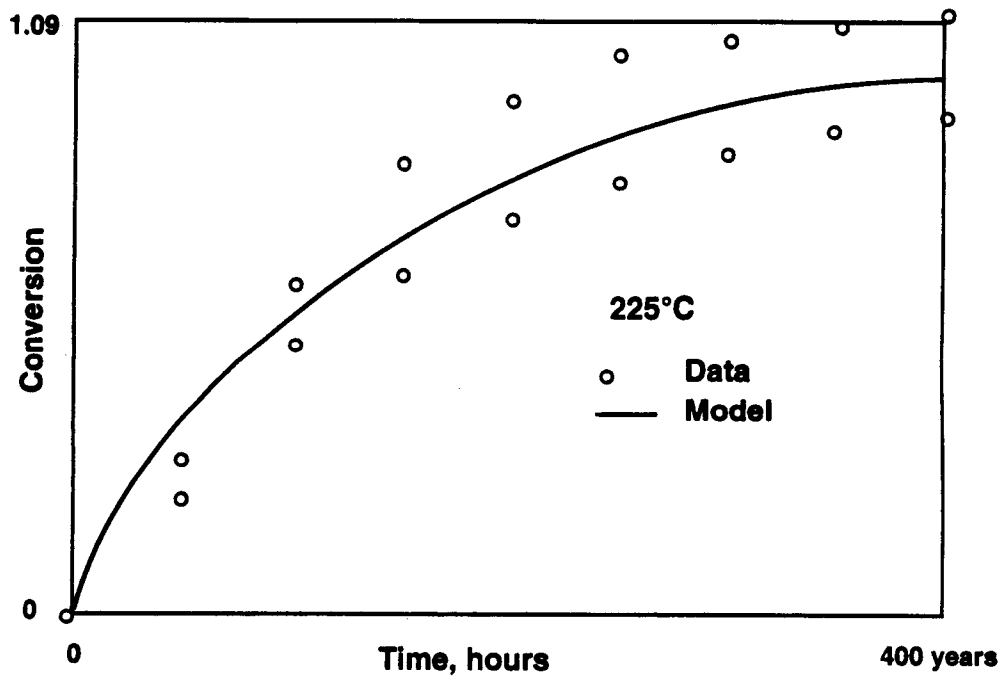


Figure 2-8(b). Model-Prototype Comparison, 225°C

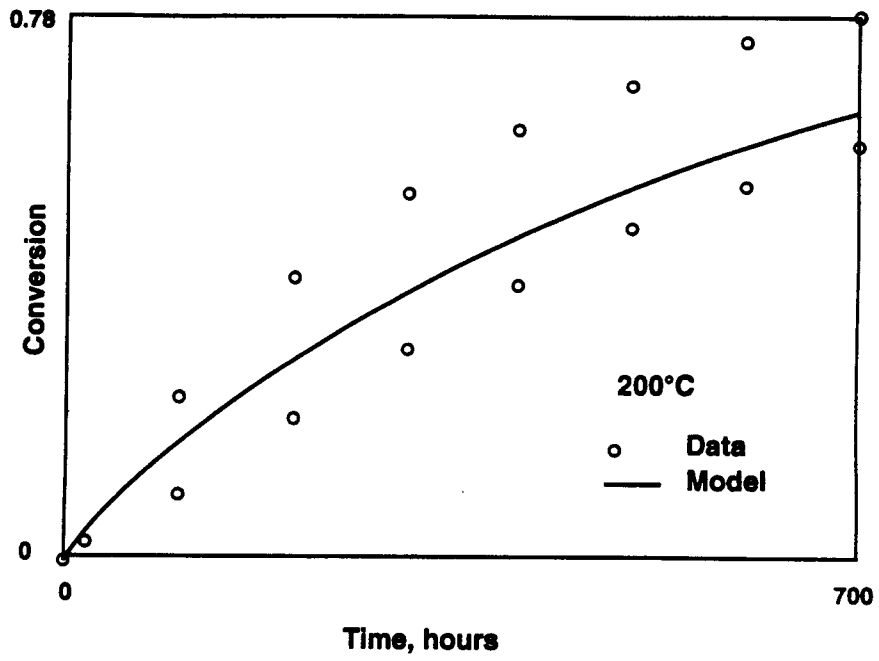


Figure 2-8(c). Model-Prototype Comparison, 200°C

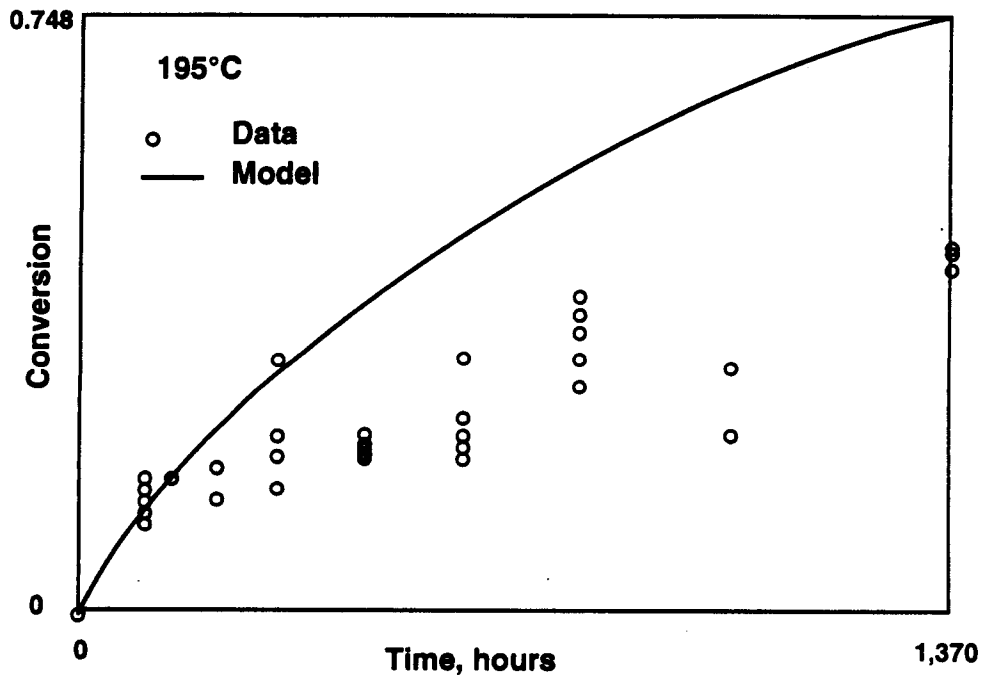


Figure 2-8(d). Model-Prototype Comparison, 195°C

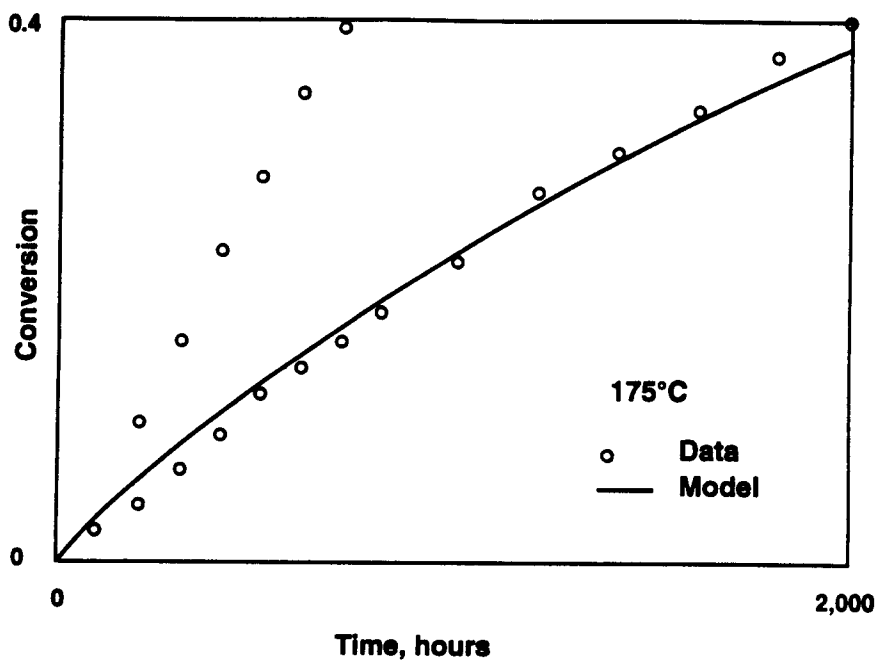


Figure 2-8(e). Model-Prototype Comparison, 175°C

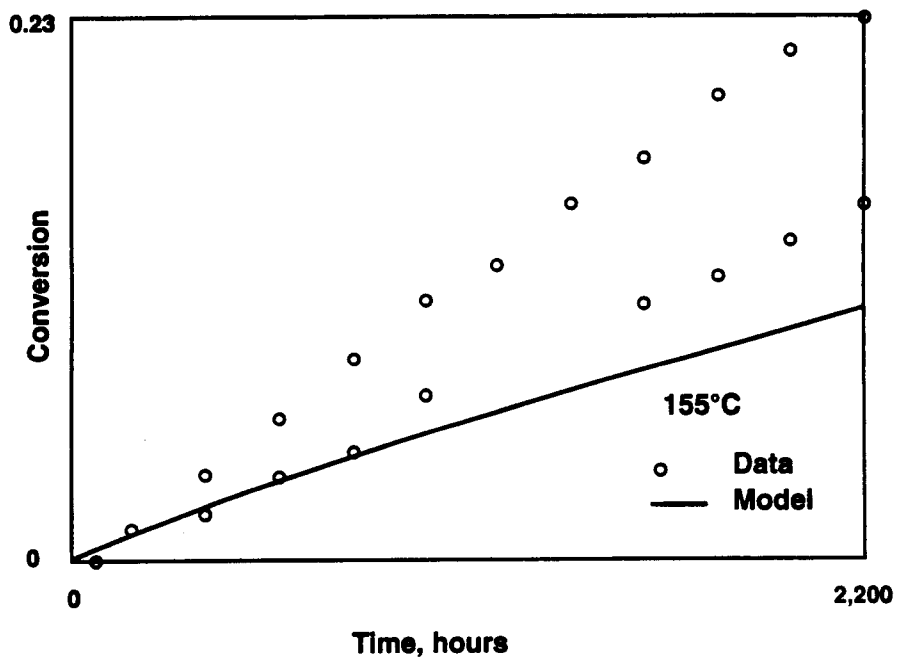


Figure 2-8(f). Model-Prototype Comparison, 155°C

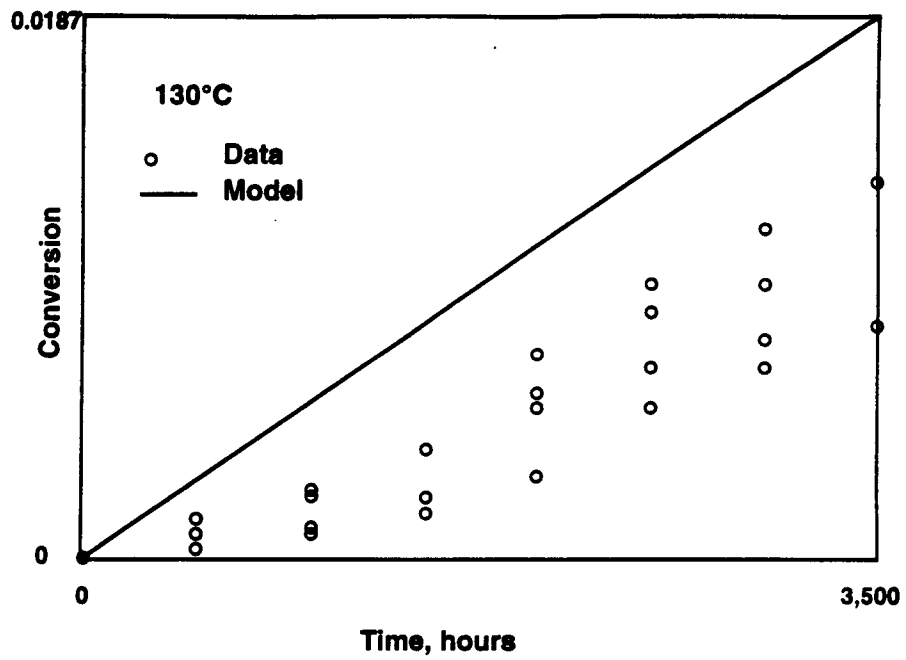


Figure 2-8(g). Model-Prototype Comparison, 130°C

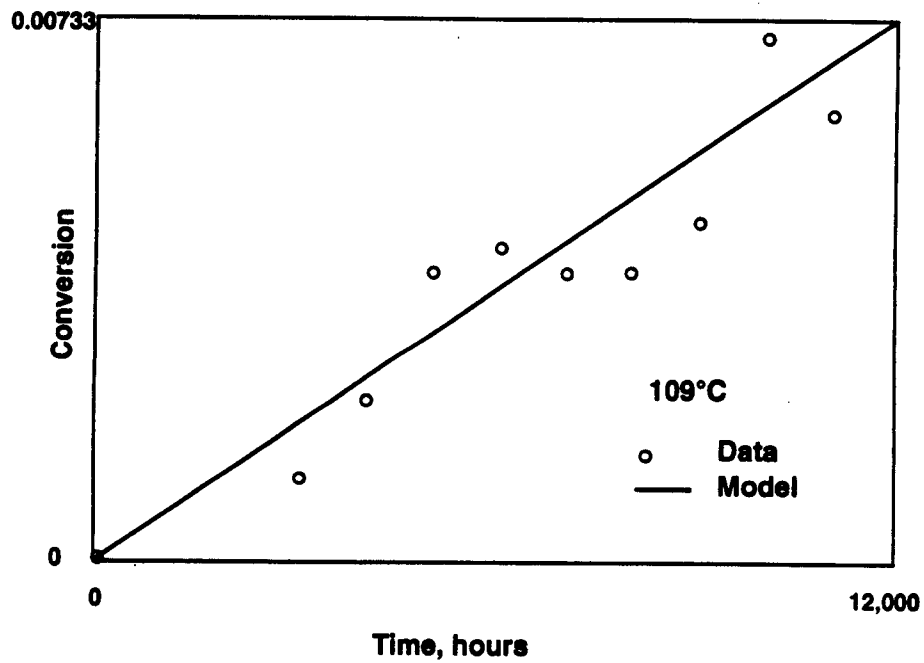


Figure 2-8(h). Model-Prototype Comparison, 109°C

3 OVERVIEW OF *SOTEC* COMPUTER CODE

3.1 INTRODUCTION

SOTEC is written in a format compatible with preFOR, a preprocessor for FORTRAN files (Janetzke and Sagar, 1991a). preFOR is designed to process code written in FORTRAN in which preFOR commands have been embedded. The output from preFOR is a standard FORTRAN file that can be compiled like any other FORTRAN file. preFOR commands allow the programmer significant flexibility in coding. preFOR commands include creation of code blocks that can be inserted at appropriate places. These insertions can be made conditional on use of a particular central processing unit, thereby increasing code portability. In addition, the preFOR utility provides other features such as numbering of lines in the code, deletion of comments, and trimming of trailing characters. preFOR is written in ANSI standard FORTRAN 77. *SOTEC* has been tested on VAX computers, Sun workstations and CRAY supercomputers.

The first task in *SOTEC* is to read the input data in the subroutine INPUT. The primary input data is in the file "sotec.inp." A number of auxiliary files may be specified in the main input file. Most data input in *SOTEC* is through the RDFREE utility (Janetzke and Sagar, 1991b). The RDFREE utility was designed for creating a user friendly data input mechanism for FORTRAN programs. It consists of a suite of subroutines written in ANSI standard FORTRAN 77. These subroutines are designed for reading alphanumeric data without the usual format constraints that are inherent to FORTRAN. That is, the RDFREE utility can be used to read data without specifying any format type such as "I, E, F, A, or G" and generally without constraints of sequence. The basis of RDFREE is to associate alphanumeric data with appropriate keywords. The keywords and the data associated with each keyword are selected by the developer of the FORTRAN application program. The user's guide for RDFREE is recommended reading for all users of *SOTEC*.

After reading the input deck, *SOTEC* loops through time and over cells to determine the lifetime of all the containers in the cells. After the corrosion loop, the number of containers in each cell that have failed by initial defects, scenarios, and time dependent failure are summarized. The liquid and gaseous release subroutines (*LIQUID* and *CANIS*) are called for each failure category with nonzero waste package failures. This requires 7 to 21 calls to the release modules. After calling the release modules, the releases from each cell are combined and written to output files.

3.2 WASTE PACKAGE FAILURES

SOTEC calculates waste package failures by looping through time and over the number of cells. At the beginning of each time step, the subroutine *TIMVAL* is called to estimate the time dependent values of a number of variables by linear interpolation of input data.

In the corrosion calculations, the corrosion potential and rate(s) of pitting and crevice corrosion are estimated as part of the main program. The general corrosion rate is calculated in the subroutine *UNICOR* as a function of time based upon the temperature and solution composition surrounding a waste package. After corrosion occurs, the current container thickness is passed to the subroutine *BUCKLE*. Subroutine *BUCKLE* determines if an individual container will fail by buckling. Buckling is dependent upon time varying loads and container thickness. After the buckling and corrosion steps, the subroutine *FAIL* is called to determine if the containers in a particular cell have failed. Failure times are recorded for each cell.

Subsequent to the time loops, the waste package failures for each cell are organized into 21 potential categories depending on the cell number and type of failure (i.e., time dependent, initial defective, and scenario specified).

Subroutine *LIQUID* carries out the numerical integration of the inventory of water and radionuclides in the container, integrating the rates for the numerical integration calculated in subroutine *DERIV*. Subroutine *SOURCE* checks for the state of failure of the container. Once the container fails, it begins calculating the inventory of water in the container and radionuclide inventories in the unaltered uranium dioxide and in the released radionuclides. Water is assumed to enter the container near the top, and to begin to flow out once it reaches the lowest hole that supports liquid flow. The volume of water within the container is calculated numerically by the Runge-Kutta method. This subroutine also initiates the calculation of the diffusive transport (see subroutine *DFLUX*).

3.3 DESCRIPTION OF RELEASE RATE MODULE LIQUID AND ITS SUBROUTINES

3.3.1 *DERIV*

Subroutine *DERIV* calculates the derivatives for the Runge-Kutta integration of the ordinary differential equations describing the system state in terms of water volume and radionuclide inventory for radionuclides released in the liquid phase. Radionuclides are released from the uranium dioxide fuel at a rate determined by the alteration rate. The radionuclides then enter the RBU inventory to determine the concentration in the container water. All isotopes of the same element are summed, and a test performed to determine that, if the inventory were dissolved in the available water, the solubility limit would be exceeded. If solubility is exceeded, the concentration of the water is the solubility of that element. If not, the concentration of that element is its RBU inventory divided by the volume. The concentration is reappportioned according to the ratio of isotopes of the element. Release rate from the container by advection and diffusion is tied directly to the concentration of each radionuclide in the water.

Advective transport from the container is simply the concentration of the radionuclide in the container water times the flow rate out of the container. There is no advective release prior to overflow of the container.

Diffusive release is calculated as if there were no advection, and the resulting diffusive flux added to the advective flux. Diffusive flux is calculated from a sphere with the inner concentration held at the concentration of the container water, and the outer boundary condition held at zero concentration. Subroutine *DFLUX* calculates the diffusive flux using a one dimensional finite difference method with variable grid spacing in spherical coordinates, and a backward-in-time numerical solution.

3.3.2 *DECAY*

Subroutine *DECAY* calculates the quantity of each radionuclide remaining in the unconverted fuel using an analytical solution with the Bateman equations:

$$N_{ij} = \prod_{k=1}^{j-1} \lambda_k N_{i0} \sum_{l=i}^j \frac{e^{-\lambda_l t}}{\prod_{\substack{m=i \\ m \neq l}}^j (\lambda_m - \lambda_l)} \quad (3-1)$$

where N_{ij} = contribution from the i th chain member to the j th chain member,

$$\begin{aligned} \lambda_j &= \text{decay coefficient of } j\text{th member of chain} \\ N_{i0} &= \text{initial quantity of } i\text{th member of chain} \\ t &= \text{time} \end{aligned}$$

The total quantity of the j th chain member at any time is therefore:

$$N_j = \sum_{i=1}^j N_{ij} \quad (3-2)$$

3.3.3 *SETDIF*

Subroutine *SETDIF* sets some of the coefficient matrix for the numerical diffusion solution. Only those coefficients that do not change with the radionuclide are set.

3.3.4 *DFLUX*

Subroutine *DFLUX* calculates the backward-in-time solution for the diffusion.

3.3.5 *DIAG3*

Subroutine *DIAG3* is the Thomas solution for a tridiagonal set of linear algebraic equations.

3.4 DESCRIPTION OF INPUT FOR SUBROUTINE *LIQUID*

Data for subroutine *LIQUID* consist mainly of a table of source term radionuclides, a description of the decay of chains of radionuclides including initial quantity and half-lives, retardation coefficients in the nearby rock, elemental solubilities, and fractions of the radionuclide in the fuel and quick-release compartments. In addition to these data, the following parameters must be input:

3.5 RELEASE RATE MODULE FOR ^{14}C

3.5.1 *CI4REL*

Calculates the release of ^{14}C in the gas phase from waste containers in an unsaturated site.

The model considers that the ^{14}C in the spent fuel is contained in three compartments:

- Cladding oxide and gap (quick release) fraction;
- Cladding and structural metal;
- Uranium dioxide.

The first compartment is the "quick release" fraction, and is released upon failure of the container. The second and third compartments release their inventory over time, and depend upon the temperature following container failure.

3.6 DESCRIPTION OF SUBROUTINES IN ^{14}C RELEASE MODULE

3.6.1 *CANIS*

The main subroutine for ^{14}C release

3.6.2 *SOLVEZ*

Release from zirconium metal upon oxidation

LININT2: Linear interpolation routine

3.6.3 DERIVS

Rate of release of ^{14}C from fuel for numerical integration

3.6.4 RKQC

Variable time step driver for Runge-Kutta integration

3.6.5 RK4

4th order Runge-Kutta integration routine

Subroutine *CI4REL* calculates the releases from a cell whose properties represent containers in the repository. Inputs to the subroutine are by an argument list and a named common block /C14GAS/.

4 INPUT COMMANDS FOR *SOTEC*

The input to *SOTEC* is from a data file that is created using appropriate keywords. These keywords and the numeric and/or alpha data associated with them are explained in detail in this chapter.

The RDFREE library of subroutines is utilized to read and interpret the data. RDFREE subroutines are designed to read all characters (alpha and numeric) in a record of any length and analyze this record to assign input values to various parameters associated with the keyword of that record. Details about RDFREE are available in a separate document (Janetzke and Sagar, 1991b) and will not be repeated here. However, it should be mentioned in brief that this form of input allows liberal use of comment lines, blank lines, as well as writing of input in English-like constructs. The entry of numeric values in a record is free of any rigid format requirements.

This section defines the *SOTEC* keywords in alphabetical order. The modifiers are grouped under their respective keywords also in alphabetical order. A detailed description of the syntax of the input mechanism is given in Appendix II. Note that all logical records must end with a special symbol which is |.

The descriptions of keywords is given in six parts called SECTION, PURPOSE, SYNTAX, DATA ITEMS, COMMENTS, and EXAMPLE. The descriptions of modifiers are made of five parts, the same as keywords except the SECTION part is omitted.

The SECTION part is used for keywords only. It indicates the section of the input file in which the keyword is used.

The PURPOSE part gives a functional description of the keyword or modifier and indicates the action requested by its use.

The SYNTAX part shows the general form of the input record along with the position of any data fields. The keyword or modifier is given in uppercase and the generic data identification is given in lowercase if present.

The DATA ITEMS part is a table which describes each data field of the keyword. The data field may be one of two types string or numeric. The numeric data values can be input in any form (integer, fixed point, or exponent form) and any column following the keyword or modifier. String values must be enclosed in single quotes ('). The minimum and maximum columns indicate the permissible range of numeric data, but for string data they indicate the minimum and maximum number of characters allowed in the string. The default/required column gives the default value if the keyword/modifier is optional, but if they are required

parameters the word 'required' is displayed. The description column describes the data field with respect to the keyword/modifier shown in the upper right of the page.

The COMMENTS part gives further details of keyword and data items and hints on their usage.

The EXAMPLE part gives an example of a logical record which uses the keyword/modifier.

KEYWORD

CHEMICAL

SECTION

This keyword belongs to the environmental parameter section of the input file.

PURPOSE

The CHEMICAL keyword is used to specify data that pertain to the chemical environment in the near-field. Time-dependent chemical concentration data is read from an external file created by the user. There are two modifiers associated with this Keyword. These are FORMULA and CELL_L. Species names and their formula weights are specified by the modifier FORMULA. The CELL_L modifier specifies the space-dependent chemical loads.

SYNTAX

CHEMICAL string; modifier - data; modifier - data

DATA ITEMS

Field	Description	Minimum Size	Maximum Size	Default Name
String	SOTEC chemical data file name	1 character	64 characters	Chefil

STRUCTURE OF EXTERNAL FILE

The external file is for reading time-dependent chemical concentration data. This is a formatted file which may be written by another program designed to calculate chemical concentrations or it may be created by the user in some other way. The file is assumed to contain a table, the first column of which is time. The number of data sets in the table is provided in the second line. The number of data sets in the file should be less than or equal to the dimensioning parameter MAXTIM. Further description is provided below.

Line 1: file title—data in this line is for file identification only and is not used anywhere.

Line 2: file section name up to 9 characters long and number of entries in the table. There can be up to MAXCHE sections in this file, each section representing the concentration of a specific chemical. MAXCHE is a dimensioning parameter that specifies the maximum number of chemicals to be considered. The names of chemical

species are read by using the modifier FORMULA explained later. Number of entries (nsets) for each chemical species in the table must be less than the dimensioning parameter MAXTIM. The format of this line is: (A9,I10). That is, the first nine columns contain the section name (which in this case is the name of the chemical species) and the number of table entries are read in I10 format. If the number of entries in the table are less than 2 than it is assumed that the species concentration are invariant with time.

Lines 3, 4 & 5: header lines, e.g., titles of columns in the table. These two lines are for ease of reading the table only and may be blanks.

Next nsets lines: data to be read. Format is 2e10.1 where the first number in a line is the time and the second, the concentration. The units of concentration are kg of species per kg of water.

Example of chefil:

time-dependent data on concentration of chemical species

CL	3
time	ref concn
(years)	(ppm)

----	-----
0.	31.5e1
1.e2	27.1e1
1.e3	18.e1

NO3	2
time	concentration
(years)	(ppm)

----	-----
0.	1.2e3
1.e4	1.2e3

COMMENTS

Chemical species names are also used in other input sections such as to specify the aggressive ions for corrosion. The names used in the table above should be consistently used in all sections of the input file, that is if nitrate ion is represented by NO3, then NO3 should be used as the name of nitrate ion where ever it occurs in the input file. Any name up to nine characters can be selected. For example, NITRATE, NO3+ are equally acceptable as NO3 used in the example.

The first two chemical species are required to be pH and Eh. The default parameters in the code assume other species will be in units of ppm.

EXAMPLE

CHEMICAL time-dependent data in 'concen.dat' file

FORMULA: no of species = 2

species name formula weight

CL 36

NO3 65

CELL-L: chemical load factors

from cell to cell che load

1 5 1.0 |

KEYWORD

CHEMICAL

MODIFIER

CELL_Load

PURPOSE

To specify cell-dependent chemical load factor. The time-dependent data on concentration of chemical species is read from an external file. The cell-dependent load factor specified by this modifier when combined with the time-dependent data provide a space and time-dependent concentration of various species.

SYNTAX

CELL_Load: from cell n1 to cell n2, load factors = v1, v2, ..., vn

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ the number of cells defined by keyword GEOMETRY	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Cell load factor for first chemical species	> 0	Any	1.0
v2	Cell load factor for second chemical species	> 0	Any	1.0
...

Field	Description	Minimum Value	Maximum Value	Default Value
vn	Cell load factor for nth chemical species	> 0	Any	1.0

COMMENTS

As many load factors as there are species can be specified by this modifier. The time-dependent species concentration read from the external file (see CHEMICAL keyword) is multiplied by these load factors to obtain concentration in each cell.

The load factors must be provided in the same sequence as these are read by the modifier FORMULA.

EXAMPLE

CHEMICAL time-dependent data from 'chem.dat'

```

CELL_load
\\ \   from cell  to cell  f1   f2   f3
      1         5      1.1  1.3  1.0

```

KEYWORD

CHEMICAL

MODIFIER

FORMULA

PURPOSE

This modifier specifies the names of all chemical species (other than radionuclides) and their formula weights that may be used in calculating the source term. In *SOTEC* Version 1.0, these are used primarily for calculation of the corrosion rates of container material.

SYNTAX

FORMULA n; string1,v1; string2,v2; ...; stringn,vn

DATA ITEMS

Field	Description	Minimum value or Size	Maximum Value or Size	Default Value
n	Number of chemical species	At least 1	≤ MAXCHE	MAXCHE
String1	Name of first chemical species	At least 1 character	Nine characters	None
v1	Formula weight of first chemical species	> 0	Any	None
String2	Name of second chemical species	At least 1 character	Nine characters	None
v2	Formula weight of second chemical species	> 0	Any	None

Field	Description	Minimum value or Size	Maximum Value or Size	Default Value
...	None
Stringn	Name of nth chemical species	At least 1 character	Nine characters	None
vn	Formula weight of nth chemical species	> 0	Any	None

COMMENTS

Whenever species names are required by other keywords of *SOTEC*, these should be consistent with the names defined by the FORMULA modifier. Currently, the MAXCHE parameter is set to 30. The first two chemical parameters in the file are specified to be Eh and pH.

EXAMPLE

CHEMICAL species time-dependent data on 'chem.dat'

FORMULA: read 5 species

```

///   name      formula weight
///   ---      -
Eh           1
pH           1
SO4          96
CL           35.4
HCO3         61

```

KEYWORD

CONTAIner

SECTION

This keyword belongs in the container data section of the input file.

PURPOSE

All data pertaining to the waste container is specified with modifiers associated with this keyword. Two options for determining container failures are provided—either by the use of mechanistic corrosion models or by the use of statistical distributions. Note also that containers may fail due to mechanical loads specified by the keyword MECHANICAL. The modifiers used with this keyword are: MATERIAL type, wall THICKNESS, number of initially DEFECTIVE, number of failures due to some SCENARIO, names of aggressive IONS, parameters for UNIFORM corrosion, parameters for PITTINING corrosion, parameters for hydrogen EMBRITTLement, parameters for STRESS corrosion, parameters for CREVICE corrosion, parameters for MICROBIAL corrosion, parameters for GALVANIC corrosion, parameters for determining failures from a STANDARD statistical distribution, and values of empirical distribution for determining failures. In SOTEC, Version 1.0, only the uniform, pitting, and crevice corrosion modes are implemented.

SYNTAX

CONTAIner: modifier: data; modifier: data; ...

DATA ITEMS

None.

COMMENTS

Some of the modifiers associated with different corrosion modes are not active at this time. Only the active modifiers are described in detail.

EXAMPLE

CONTAIner properties and parameters:

MATERIAL: 1 (stainless steel)

formula wt = 80; density = 2000; metal ion diff = 0

Youngs modulus = 5.e6, shear modulus = 4.e6

Poisson ratio = 1.25

THICKNESS of wall

\\	from cell	to cell	wall thickness
	1	10	0.02

DEFECTIVE initially

\\ from cell to cell # defective
1 10 0

SCENARio failures

\\ from cell to cell mean failures
1 10 5

IONS for corrosion: NO3 and CL

UNIFORM corrosion parameters:

crit temp = 373; a = 0.1; b = 1.2e-5; c = 8.134

d = 0.5, 0(concen val); 0.5, 1(log concn val); 0.6 |

KEYWORD

CONTAIner

MODIFIER

CREVICe corrosion

PURPOSE

Parameters for calculation of crevice corrosion are specified by this modifier. In SOTEC, version 1.0, the crevice corrosion model is a simple algebraic equation obtained by regressing the numerical results of a more mechanistic model based on solving the partial differential equation of electro-migration. The regressed equation is of the following form:

$$t = a c^b ; \text{ where } c = g/(l i_p) \text{ and}$$

$$r = (i_a w)/(F d z)$$

where, g = crevice gap, l = crevice length, i_p = passive current density, i_a = active current density, F = Faraday constant, d = material density, z = ion valency, w = formula weight of material, a,b = empirical constants derived from regression, t = initiation time for crevice corrosion, and r = rate of crevice corrosion.

Parameters needed for the application of the simple model are described below.

SYNTAX

CREVICe corrosion parameters: v1, v2, ... , v15

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
v1	Temperature above which crevice corrosion does not occur	> 0	< 100°C	96°C
v2	Crevice gap	> 0	Any	None
v3	Crevice length	> 0	Any	None

Field	Description	Minimum Value	Maximum Value	Default Value
v4	Passive current density	> 0	Any	None
v5	Active current density	> 0	Any	None
v6	Charge valency	> 0	Any	None
v7	Faraday constant	> 0	Any	None
v8	Temperature T1 below which the v9 and v10 coefficients apply	> 0	Any	25°C
v9	Empirical coefficient	Any	Any	0.116
v10	Empirical coefficient	Any	Any	1.29
v11	Temperature T2; coefficients v12 and v13 apply for temperatures between T1 and T2	> T1	Any	50°C
v12	Empirical coefficient	Any	Any	0.148
v13	Empirical coefficient	Any	Any	1.34
v14	Empirical coefficient for temperature greater than T2	Any	Any	0.116

Field	Description	Minimum Value	Maximum Value	Default Value
v15	Empirical coefficient for temperature greater than T2	Any	Any	1.471

COMMENTS

See text for description of the crevice corrosion model.

EXAMPLE

CONTAINER properties and parameters

CREVICE parameters: temp = 96, gap = 0.0001, length = 0.001,
 passive current = 0.25, active current = 0.35, valency = 2,
 Faraday Constant = 96000, TEMP = 25, a = 0.115, b = 1.30,
 TEMP = 50, a = 0.116, b = 1.50, c = 0.117, d = 1.80

KEYWORD

CONTAIner

MODIFIER

DEFECTive

PURPOSE

The number of initially defective containers is specified by this modifier.

SYNTAX

DEFECTive: from cell n1 to cell n2, value = v1

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ the number of cells specified by the GEOMETry keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Number of defective containers	≥ 0	< the number of containers in the cell	0

COMMENTS

Omit this modifier if there are no initially defective containers.

EXAMPLE

CONTAIner properties and parameters:
DEFECTive containers

\\	from cell	to cell	# defectives
	1	5	1
	6	10	0

KEYWORD

CONTAINER

MODIFIER

ECORR

PURPOSE

Parameters for corrosion potential and localized corrosion.

SYNTAX

ECORR: V1, V2, ... , V8

DATA ITEMS

Field	Description	Minimum Size	Maximum Size	Default value
V1	E1	Any	Any	None
V2	A	Any	Any	None
V3	ζ	Any	Any	None
V4	β	Any	Any	None
V5	λ	Any	Any	None
V6	Epit	Any	Any	None
V7	Ecrevice	Any	Any	None
V8	Active corrosion rate	> 0	Any	None

COMMENT

See text for description of corrosion calculations.

EXAMPLE

CONTAINER properties and parameters

ECORR: 300, 0.74, -1000, 100, 0.0231049, 320, 340, 1e-4

KEYWORD

CONTAIner

MODIFIER

IONS

PURPOSE

Names of aggressive ions for the corrosion process are specified by this modifier. The names specified here must be included in the list of names provided with the keyword CHEMICAL, modifier FORMULA.

SYNTAX

IONS: s1, s2, ... , Sn

DATA ITEMS

Field	Description	Minimum Size	Maximum Size	Default value
s1	Name of first aggressive ion	1 character	6 characters	None
s2	Name of second aggressive ion	1 character	6 characters	None
...
sn	Name of nth aggressive ion	1 character	6 characters	None

COMMENT

The number of ions specified should be less than equal to those specified with the CHEMICAL keyword.

EXAMPLE

CONTAIner properties and parameters
IONS for corrosion: NO3 and CL

KEYWORD

CONTAINER

MODIFIER

MATERIAL

PURPOSE

To specify properties of the container material. In the current models of SOTEC, some of the properties read are not used.

SYNTAX

MATERIAL: v1, v2, ... , v7

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
v1	Type of container material; 1 = steel, 2 = copper	1 (steel)	2 (copper)	1
v2	Formula weight of material	> 0	Any	1.0
v3	Density of material	> 0	Any	1.0
v4	Metal ion diffusion coefficient	> 0	Any	0
v5	Young's Modulus	> 0	Any	None
v6	Shear Modulus	> 0	Any	None
v7	Poisson's ratio	> 0	Any	None

COMMENTS

The formula weight, metal ion diffusion coefficient, and material density are currently not used in any formulation in *SOTEC* and these can be specified as zero. These are for future use.

EXAMPLE

CONTAINER MATERIAL properties:

formula wt = 0, density = 0, diff coef = 0

Youngs modulus = 1.e6, shear mod = 1.e6, Poisson ratio = 1.2

KEYWORD

CONTAIner

MODIFIER

PITTING corrosion

PURPOSE

Parameters for calculation of pitting corrosion are specified by this modifier. In *SOTEC*, version 1.0, a probabilistic pitting model is used. The probability of pit initiation is assumed to depend upon the time of exposure of the material to pitting environment. Similarly, the pit growth rate is assumed to depend upon the age of the pit. Various parameters needed for this probabilistic model are described below.

SYNTAX

PITTING parameters: v1, v2, ... , v8

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
v1	Temperature above which pitting corrosion does not occur	Any	Any	96°C
2	Probability of pit initiation	0	1.0	None
v3	Probability of pit death	0	1.0	None
v4	Pit age at which pit becomes stable and after which it does not die	> 0	Any	None

Field	Description	Minimum Value	Maximum Value	Default Value
v5	Rate of pit diameter growth	> 0	Any	None
v6	Rate of pit depth growth	> 0	Any	None
v7	Factor for changing initiation probability with age	> 0	Any	None
v8	Factor for changing pit growth with age	> 0	Any	None

COMMENTS

See text for description of the pitting model.

EXAMPLE

CONTAINER properties and parameters

PITTING parameters: temp = 96, initiation prob = 0.1,
 death prob = 0.09, stable age = 0.001, dia growth rate = 1.e-5,
 depth growth rate = 1.e-6, factor for prob = 0.8,
 factor for growth = 0.9

KEYWORD

CONTAINER

MODIFIER

SCENARIO failures

PURPOSE

To specify the number of containers that fail due to some disruptive scenario such as volcanism. The time at which such a scenario occurs is to be read by the system code or it can be read in the *SOTEC* itself. Scenario failures are assumed to have a Gaussian distribution and the 5th and 95th percentiles are specified as needed by the LHS module. If the 95th percentile is specified as zero, then it is assumed that the number of failures is constant.

SYNTAX

SCENARIO: from cell n1 to cell n2, values = v1, v2

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ the number of cells specified by the GEOMETRY keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Failure time	≥ 0	None	0
v2	Number of containers failed	> V1 or 0	< number of containers	None

COMMENTS

Omit this modifier if there are no failures due to disruptive scenarios.

EXAMPLE

CONTAINER properties and parameters:

SCENARIO failures

\\	from cell	to cell	time	number
	1	5	100	3
	6	10	500	1

KEYWORD

CONTAIner

MODIFIER

THICKNess

PURPOSE

Container wall thickness is specified by this modifier.

SYNTAX

THICKNess: from cell n1 to cell n2, value = v1

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ the number of cells specified by the GEOMETry keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Container wall thickness	> 0	Any	1.0

COMMENTS

None.

EXAMPLE

CONTAIner properties and parameters

THICKNess of wall

```

\\
    from cell  to cell  wall thickness
      1       10       0.025
    
```

KEYWORD

CONTAIner

MODIFIERUNIFORM
corrosion**PURPOSE**

To specify parameters of the uniform corrosion model. In Version 1 of *SOTEC*, an empirical expression for uniform corrosion rate of the following form is assumed:

$$r = a \exp\{b/(cT)\} t^d C_1^e C_2^f \dots ; T < T^*$$

where, r is the corrosion rate, a , b , c , d , e , and f are empirical constants, T is temperature, and t is time. Instead of concentrations C_1 , C_2 , etc., logarithms (to base 10) e.g., $\log_{10} C_1$, etc. can also be used. In some interpretations of this equation, $b =$ Gibb's free energy and $c =$ gas constant. The equation is used only if the temperature is less than some specified temperature, T^* .

SYNTAX

UNIFORM corrosion parameters: v_1, v_2, \dots, v_n

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
v1	Temperature T^*	Any	Any	96°C
v2	a	> 0	Any	1.0
v3	b	Any	Any	1.255e-5 J/mol-K
v4	c	Any	Any	8.134 J/mol-K
v5	d	Any	Any	0.5

Field	Description	Minimum Value	Maximum Value	Default Value
v6	Flag signifying concentration or log concentration of first aggressive ion	0 (use concentration value)	1 (use log concentration value)	0
v7	e	Any	Any	0.5
v8	Flag repetition for next aggressive ion	Same as v6	Same as v6	0
v9	Exponent repetition for next aggressive ion	Same as v7	Same as v7	0.5

COMMENTS

By adjusting parameters, many types of models may be fitted by the uniform corrosion equation used above.

EXAMPLE

CONTAINER properties and parameters:

UNIFORM corrosion parameters:

temp = 96; a=1.0, b=1.e-5, c=8.1, d=0.5; 0,.5; 1,1

KEYWORD

END

SECTION

This keyword belongs in the calculation section of the input file

PURPOSE

This key word signifies the end of the input data stream. When END is encountered in the input file, reading of input is stopped and calculations are begun. Therefore, it must be the last data record of the input file.

SYNTAX

END |

DATA ITEMS

None.

COMMENTS

If the END keyword is not the last record in the input file, the remainder of the file is ignored.

EXAMPLE

END |

KEYWORD

GEOMETry

SECTION

This keyword belongs to the calculation section of the input file.

PURPOSE

This is the primary keyword for specifying the geometric parameters such as repository dimension, number of distinct computational cells, waste package dimensions, etc. as will become evident from the various modifiers explained later.

SYNTAX

GEOMETry modifiers data |

DATA ITEMS

All data items are associated with modifiers as explained in the following pages.

COMMENTS

Four modifiers are associated with the GEOMETry keyword. These are: (1) REPOSITory; (2) CELL; (3) PACKAGe; (4) BOREHole. Data associated with each is explained on the following four pages. The GEOMETry keyword can be used anywhere in the input file.

EXAMPLE

```

GEOMETRY
REPOSITory = 2000 m x 1500 m x 10 m; 10 cells
\\ read cell characteristics
CELL      volume  no of packages
1 to 5    2.e4    1500
6 to 10   3.e5    2000
\\ volmax = vol after which water flows out of pac
\\ rsphere = radius of equivalent sphere
PACKAGe
\  cell  length  dia  volmax  rspher
  1 to 4  4.2    1.5  3.5    3.15
  5 to 10 3.8    1.25 2.5    2.85
\\ read characteristics of emplacement hole
BOREHole
  cell  length  dia  orientation
  1 to 10 5    2.0 1 (vertical)
|

```

KEYWORD

GEOMETry

MODIFIER

BOREHole

PURPOSE

To specify length, diameter, and orientation of the emplacement borehole. This data is cell dependent.

SYNTAX

BOREHole from cell n1 to cell n2, properties = v1,v2,v3

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ the number of cells specified with the GEOMETry modifier	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Borehole length	> package length	Any	1.0
v2	Borehole diameter	> than package diameter	Any	1.0
v3	Borehole orientation	1 (Vertical emplacement)	2 (Horizontal emplacement)	1

COMMENTS

The space between the borehole wall and the outer surface of the waste package is assumed to contain packing material which may be air.

EXAMPLE

	GEOMETry BOREHole				
\\	from cell	to cell	length	dia	orientation
	1	3	2.5	1.2	1

KEYWORD

GEOMETry

MODIFIER

CELL

PURPOSE

Characteristics of the computational cell or compartment are read by this modifier. The cell characteristics include its volume and the number of waste packages.

SYNTAX

CELL from cell n1 to cell n2 properties are v1, v2

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ number of cells specified with REPOSItory modifier	1
n2	Ending cell number	≤ n1	Same as above	MAXCEL
v1	Cell volume	> 0	Any	1.0
v2	Number of waste packages	> 0	Any	1

COMMENTS

Data can be provided cell by cell or in groups of cells. However, all four of the numeric fields must be specified. That is even when data is to be provided cell by cell, the syntax will be: from cell 1 to cell 1, vol = v1 and no of packages = 10.

EXAMPLE

	GEOMETry	CELL		
///	from	to	volume	packages
	1	3	5.e3	1200
	4	4	15000	800

KEYWORD

GEOMETry

MODIFIER

PACKAGe

PURPOSE

This modifier specifies the size of the waste package.

SYNTAX

PACKAGe from cell n1 to cell n2, properties = v1,v2,v3,v4

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ number of cells specified with REPOSITory modifier	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Package length	> 0	Any	1.0
v2	Package diameter	> 0	Any	1.0
v3	Liquid volume that can be contained in a container without overflow	> 0	Any	1.0
v4	Radius of equivalent sphere	> 0	Any	1.0

COMMENTS

v4 is the radius of a sphere whose volume is equal to the volume of the container.

EXAMPLE

GEOMETry PACKAGe
\\ from cell to cell len dia overflow sphere
1 3 3.0 1.0 1.2 2.25

KEYWORD

GEOMETry

MODIFIER

REPOSItory

PURPOSE

The REPOSItory modifier with the keyword GEOMETry is used to read size of the repository and the number of cells or compartments into which the repository is divided for computation purposes.

SYNTAX

REPOSItory: v1, v2, v3, v4

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
v1	Repository length	> 0	Any	1.0
v2	Repository width	> 0	Any	1.0
v3	Repository height	> 0	Any	1.0
v4	Number of cells or compartments	≥ 1	\leq the dimensioning parameter MAXCEL	1

COMMENTS

MAXCEL is a dimensioning parameter. Its value is currently 20.

EXAMPLE

GEOMETry

REPOSItory l = 50 m; w = 20 m; h = 5 m; n = 2 |

KEYWORD

<p>MECHANical parameters</p>

SECTION

This keyword belongs in the environmental section of the input file.

PURPOSE

Parameters related to mechanical stresses are specified through the use of this keyword. Two modifiers are associated with it. These are YIEREF and LOAREF. Reference yield stresses are specified with the help of YIEREF, and LOAREF is used to specify the mechanical loads on the system.

SYNTAX

MECHANical parameters: modifier:data; modifier:data

DATA ITEMS

None.

COMMENTS

See description of modifiers YIEREF and LOAREF for greater detail.

EXAMPLE

MECHANical parameters

```

YIEREF: data on file 'MECFIL'
\\      load factors are as follows
\\      from cell  to cell  comp  tensile  shear
          1      5      1.e6  1.e6     1.e4

LOAREF: data on file 'MECFIL'
\\      load factors are as follows
\\      from cell  to cell  comp  tensile  shear
          1      5      8.e6  9.e6     5.e3

```

KEYWORD

MECHANical
parameters

MODIFIER

LOAREF

PURPOSE

The reference loads are specified with this modifier. Up to three different loads can be specified. These could in general be compressive, tensile, and shear loads or may have other appropriate interpretations depending upon the mechanical model selected, e.g., one of these could represent circumferential torque. The time-dependent data is on an external file while the cell dependent load factors are on the main input file.

SYNTAX

LOAREF: string; from cell n1 to cell n2, values = v1,v2,v3

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
String	Name of external file containing time-dependent load data	1 character	9 characters	'MECFIL' is the default name of this file
n1	Beginning cell number for cell-dependent data	> 0	≤ to the number of cells specified with the GEOMETry keyword	1
n2	Ending cell number for cell-dependent data	≥ n1	Same as above	MAXCEL

Field	Description	Minimum Value	Maximum Value	Default Value
v1	Cell load factor for first load	> 0	Any	None
v2	Cell load factor for second load	> 0	Any	None
v3	Cell load factor for third load	> 0	Any	None

STRUCTURE OF EXTERNAL FILE

The external file for reading time-dependent loads is the same one which also contains the time-dependent yield stresses. See keyword MECHANical, modifier YIEREF for complete description and example.

EXAMPLE

MECHANical parameters are specified below

```

LOAREF: read the 'MECFIL' file for time-dependent loads
///      cell-dependent load factors are as follows
///      from cell    to cell    comp  tensile  shear
           1          10         1.0   0.9     0.8

```

KEYWORD

MECHANical
parameters

MODIFIER

YIEREF

PURPOSE

The reference yield stresses are specified with this modifier. Up to three different yield stresses can be specified. These could in general be in compression, tension, and shear or may have other interpretations depending upon the mechanical model selected, e.g., one of these could represent circumferential yield stress. The time-dependent data is on an external file while the cell dependent load factors are on the main input file.

SYNTAX

YIEREF: string; from cell n1 to cell n2, values = v1,v2,v3

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
String	Name of external file containing time-dependent yield stress data	1 character	9 characters	'MECFIL' is the default name of this file
n1	Beginning cell number for cell-dependent data	> 0	≤ the number of cells specified with the GEOMETry keyword	1
n2	Ending cell number for cell-dependent data	≥ n1	Same as above	MAXCEL

Field	Description	Minimum Value	Maximum Value	Default Value
v1	First yield stress load factor	> 0	Any	None
v2	Second yield stress load factor	> 0	Any	None
v3	Third yield stress load factor	> 0	Any	None

STRUCTURE OF EXTERNAL FILE

The external file is for reading time-dependent mechanical loads data. This is a formatted file which may be written by another program designed to calculate yield stresses or it may be created by the user in some other way. The file is assumed to contain a table, the first column of which is time. The number of data sets in the table is provided in the second line. The number of data sets in the file should be less than or equal to the dimensioning parameter MAXTIM. Further description is provided below.

Line 1: file title—data in this line is for file identification only and is not used anywhere.

Line 2: file section name up to 9 characters long and number of entries in the table. There can be four sections to this file. The section headings for this file are 'YIELD' and 'LOAD'. The second section deals with the LOAREF modifier (see below). Only those sections contained in the file will be read. Number of entries (nsets) for each section in the table must be less than the dimensioning parameter MAXTIM. The format of this line is: (A9,I10). That is, the first nine columns contain the section name (which in this case is the name of the chemical species) and the number of table entries are read in I10 format. If the value of nsets is specified to be less than 2 then it is assumed that the liquid volumetric content is invariant in time.

Lines 3 & 4: header lines, e.g., titles of columns in the table. These two lines are for ease of reading the table only and may be blanks.

Next nsets lines: data to be read. Format is 2e10.1 where the first number in a line is the time and the second to fourth columns contain the liquid or the steam content or the flow rate (depending on the section) in the three zones. The units of liquid and steam

content are cubic meters of liquid or steam per cubic meter of rock volume and that of flow are cubic meter per square meter per year.

Each section begins with the line 2 above.

Example of 'MECFIL'
time-dependent data on mechanical parameters
YIEREF 3

time	comp	tensile	shear
----	-----	-----	-----
0.	5.e6	6.e6	3.e5
1.e3	4.e6	5.e6	2.e5
1.e4	3.e6	4.e6	1.e5

LOAREF 2

time	comp	tensile	shear
----	-----	-----	-----
0.	1.e9	1.e5	1.e6
1.e4	1.e12	1.e5	1.e6

EXAMPLE

MECHANICAL parameters are specified below

YIEREF: read the 'MECFIL' file for time-dependent yield stresses

\\ cell-dependent load factors are as follows

\\	from cell	to cell	comp	tensile	shear
\\	1	10	1.0	0.9	0.8

KEYWORD

NUMERICs data

SECTION

This keyword belongs in the section of the input file where time steps and spatial grid elements are specified.

PURPOSE

This keyword and its associated modifiers are used to specify spatial grids and time steps and any other information needed to do the numerical calculations.

SYNTAX

NUMERICs data: modifier, data; modifier, data; ...

DATA ITEMS

None.

COMMENTS

None.

EXAMPLE

NUMERICs data:

SIMTIM: start = 0 yrs, end = 10000 yrs, step = 100, step multiplier = 1.0, max step = 1000,

KEYWORD

PACKINg hydrology

SECTION

This keyword belong in the environmental section of the input file.

PURPOSE

The space between the outer boundary of the waste package and the wall of the waste emplacement borehole is assumed to be filled with packing material. This packing material may be air in which case its porosity may be specified to be one. POROSity and MASS_Fraction are the two modifiers associated with this keyword. Packing material porosity is read by the POROSity modifier and the liquid and steam volumetric contents are read by using MASS_Fraction modifier.

SYNTAX

PACKINg hydrology: modifier:data; modifier:data |

DATA ITEMS

None.

COMMENTS

All data items are associated with the modifiers. The same external data file as used for the ROCK hydrology keyword is also used for this keyword.

EXAMPLE

```

PACKINg hydrology
  POROSity :
\\    from cell  to cell  porosity
      1         5         1.0
  MASS_Fraction:
      time-dependent data from 'ROCHYD.DAT' file
\\    load factors are as follows
\\    from cell  to cell  load factor
      1         5         1.0 |

```


KEYWORD

PACKING

MODIFIER

MASS_Fraction

PURPOSE

To specify the liquid and steam volumetric content (it could be mass fraction if so desired) of water and steam in the packing material around the waste container.

SYNTAX

MASS_Fraction: string; from cell n1 to cell n2 load = v1

DATA ITEMS

Field	Description	Minimum Value or size	Maximum Value or Size	Default Value
String	File name containing time-dependent data on water and steam content in the packing	1 character	9 characters	File name must be the same as used for the rock hydrology data. Default name is 'HYDFIL'
n1	Beginning cell number	> 0	≤ the number of cells specified by the GEOMETry keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Cell dependent load factor for water	Any	Any	Any

Field	Description	Minimum Value or size	Maximum Value or Size	Default Value
v2	Cell dependent load factor for steam	Any	Any	Any

COMMENTS

Refer to keyword ROCK, modifier LIQUID for a description of the external file. The time-dependent data for the packing hydrology is included in the same file in which the rock hydrology data is provided. The section heading for identifying the packing data in that file is 'PACKIN'.

EXAMPLE

PACKING hydrology:

```

MASS_Fraction data on 'rochyd.dat' file
\\      cell dependent load factors are as follows
\\      from cell to cell liquid steam
          1      5      0.9  1.0

```

KEYWORD

PACKINg
hydrology

MODIFIER

POROSItY

PURPOSE

To read the porosity of the packing material.

SYNTAX

POROSItY from cell n1 to cell n2 por = v1

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ the number of cell specified by the GEOMETry keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Porosity of packing in cells from n1 to n2	≥ 0	1.0	0.6

COMMENTS

None.

EXAMPLE

PACKINg properties:

POROSItY:

```

\\
    from cell 1 to cell 5 porosity 1.0
    
```

KEYWORD

ROCK hydrology

SECTION

This keyword belongs in the environmental parameter section of the input file.

PURPOSE

The ROCK keyword is used to read the hydrologic characteristics of the rock in the near-field of the repository. The near-field rock is assumed to consist of three zones: (1) the damaged rock immediately surrounding the emplacement borehole, (2) the disturbed rock zone next to the damaged rock and the (3) intact host rock. These three zones may have distinct hydrologic characteristics.

SYNTAX

ROCK modifier:data; modifier:data; ...; modifier:data |

DATA ITEMS

None.

COMMENTS

The modifiers associated with this keyword are: (1) POROSity, (2) LIQUID, (3) STEAM, (4) FLOW, and (5) DEPTH. Each one is explained in the following.

EXAMPLE

```

ROCK hydrologic parameters
  POROSity
  \ \   from cell   to cell   zone1   zone2   zone3
        1           5         0.5     0.4     0.3
  LIQUID (water) concentrations
    time dependent data from 'ROCHYD.DAT' file
    load factors are:
  \ \   from cell   to cell   zone1   zone2   zone3
        1           5         1.3     1.01    1.0
  STEAM concentrations also from 'ROCHYD.DAT' file
    steam load factors are:
  \ \   from cell   to cell   zone1   zone2   zone3
        1           5         1.0     1.0     1.1
  FLOW rates from 'ROCHYD.DAT' file also
    flow load factors are:
  \ \   from cell   to cell   zone1   zone2   zone3
        1           5         1.1     1.0     1.2

```

DEPTH of water in borehole (min and max)
\\ from cell to cell min depth max depth
1 5 1.2 m 2.4 m |

KEYWORD

ROCK hydrology

MODIFIER

DEPTH

PURPOSE

The minimum depth is that which must be exceeded for flow to occur from the borehole; the maximum depth is the one at which inflow to the borehole is equal to the outflow. By assigning different values to these two, a variety of cases for near-field release may be analyzed.

SYNTAX

DEPTH from cell n1 to cell n2 properties = v1, v2

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default value
n1	Beginning cell number	> 0	≤ the number of cells specified by the GEOMETry keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Minimum depth before water seeps out of the borehole	> 0	≤ borehole length	1.0
v2	Maximum depth of water in the borehole	> v1	Same as above	1.0

COMMENTS

Data from this keyword is not used in SOTEC Version 1.0.

EXAMPLE

	ROCK hydrology			
	DEPTH			
\\	from cell	to cell	min depth	max depth
	1	5	1.2	2.5

KEYWORD

ROCK hydrology

MODIFIER

FLOW rate

PURPOSE

To read the rate of liquid flow in the three zones of the near-field rock. Time dependent data, if any, is contained in the external file that also contains the LIQUID and STEAM contents.

SYNTAX

FLOW rate: string; from cell n1 to cell n2, values = v1, v2, v3

DATA ITEMS

See table associated with keyword ROCK and modifier LIQUID.

COMMENTS

The external file name should be the same as for LIQUID and STEAM modifiers. The default file name is 'HYDFIL'.

EXAMPLE

```
ROCK hydrology
  liquid FLOW rates: file = 'ROCHYD.DAT'
\\
  load factors are as follows
\\
    from cell  to cell  zone 1  zone 2  zone 3
      1         4       1.1    0.9    0.5
```


KEYWORD

ROCK hydrology

MODIFIER

LIQUID content

PURPOSE

To specify the volumetric liquid content in the three zones of the near-field rock.

SYNTAX

LIQUID string; from cell n1 to cell n2, values = v1, v2, v3

DATA ITEMS

Field	Description	Minimum Value or Size	Maximum Value or Size	Default Value
String	File name for rock hydrology data	1 character	64 characters	'hydfil'
n1	Beginning cell number	> 0	≤ the number of cells specified by the GEOMETry keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Cell load factor for zone 1	Any	Any	1.0
v2	Cell load factor for zone 2	Any	Any	1.0
v3	Cell load factor for zone 3	Any	Any	1.0

STRUCTURE OF EXTERNAL FILE

The external file is for reading time-dependent hydrologic data. This is a formatted file which may be written by another program designed to calculate hydrologic environment or it may be created by the user in some other way. The file is assumed to contain a table, the first column of which is time. The number of data sets in the table is provided in the second line. The number of data sets in the file should be less than or equal to the dimensioning parameter MAXTIM. Further description is provided below.

Line 1: file title—data in this line is for file identification only and is not used anywhere.

Line 2: file section name up to 9 characters long and number of entries in the table. There can be four sections to this file. The section headings for these are 'LIQUID', 'STEAM ', 'FLOW ' and 'MASS_F'. The last section deals with the PACKING hydrology keyword but has been combined with this external file for convenience. Only those sections contained in the file will be read. Number of entries (nsets) for each chemical species in the table must be less than the dimensioning parameter MAXTIM. The format of this line is: (A9,I10). That is, the first nine columns contain the section name (which in this case is the name of the chemical species) and the number of table entries are read in I10 format. If the value of nsets is specified to be less than 2 than it is assumed that the liquid volumetric content is invariant in time.

Lines 3 & 4: header lines, e.g., titles of columns in the table. These two lines are for ease of reading the table only and may be blanks.

Next nsets lines: data to be read. Format is 2e10.1 where the first number in a line is the time and the second to fourth columns contain the liquid or the steam content or the flow rate (depending on the section) in the three zones. The units of liquid and steam content are cubic meters of liquid or steam per cubic meter of rock volume and that of flow are cubic meter per square meter per year.

Each section begins with the line 2 above.

Example of hydfil

```
time-dependent data on rock hydrology
LIQUID 3
time   zone 1   zone 2   zone 3
-----
0.     0.20     0.15     0.15
1.e3   0.20     0.15     0.15
1.e4   0.15     0.15     0.15
STEAM 2
```

time	zone 1	zone 2	zone 3
----	-----	-----	-----
0.	0.10	0.05	0.0
1.e4	0.0	0.0	0.0
FLOW	2		
time	zone 1	zone 2	zone 3
----	-----	-----	-----
0.	1.e-6	1.e-6	1.e-6
1.e5	2.e-6	2.e-6	2.e-6
MASS_F	2		
time	liquid	steam	
----	-----	-----	
0.	1.0	0.0	
1.e4	1.0	0.0	

COMMENTS

The file specified by the string field must exist and be available. The data read under the MASS_F section heading in the external data file is a modifier of the PACHING hydrology keyword. Time dependent liquid and steam volumetric contents in the packing material are read in this section.

EXAMPLE

ROCK hydrology: time-dependent data from 'ROCHYD.DAT'
 cell loads are as follows:

\\	from cell	to cell	zone 1	zone 2	zone 3
	1	5	1.1	1.2	1.1

KEYWORD

ROCK hydrology

MODIFIER

POROSItY

PURPOSE

The purpose of this modifier is to specify the porosities of the rock in the near-field of the waste emplacement holes. The near-field rock is described in terms of three zones—the damaged, disturbed and the intact rock. Different values of porosities can be read for these three different zones.

SYNTAX

POROSIties: from cell n1 to cell n2, porosities = v1, v2, v3

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ the number of cells defined by the GEOMETry keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Porosity of zone 1 (damaged rock in immediate vicinity of borehole)	> 0	1.0	0.2
v2	Porosity of zone 2 (disturbed rock located next to damaged rock)	> 0	1.0	0.02

Field	Description	Minimum Value	Maximum Value	Default Value
v3	Porosity of zone 3 (intact host rock)	> 0	1.0	0.002

COMMENTS

Any of the zones can be omitted by putting its porosity value to the porosity value of the other zones.

EXAMPLE

ROCK hydrology

POROSities of rock zones

\\	from cell	to cell	zone 1	zone 2	zone 3
	1	5	0.4	0.3	0.25

KEYWORD

ROCK hydrology

MODIFIER

STEAM content

PURPOSE

Similar to the LIQUID modifier, the steam volumetric content is specified by this modifier.

SYNTAX

STEAM string; from cell n1 to cell n2, values = v1, v2, v3

DATA ITEMS

See Table under keyword ROCK and modifier LIQUID

COMMENTS

The external file for STEAM is the same as for the LIQUID modifier. See the LIQUID modifier for its description and example.

EXAMPLE

STEAM content: time-dependent data from 'ROCHYD.DAT' file
\\ cell load factors are as follows
\\ from cell to cell zone 1 zone 2 zone 3
1 5 1.0 1.1 1.2

KEYWORD

THERMAL

SECTION

This keyword belongs to the environmental parameter section of the input file.

PURPOSE

This keyword is one of the four keywords for defining the near-field environment. It specifies the thermal environmental parameters. Time-dependent average temperature is read from an external file. Two modifiers—HEAT and CRITICAL are also associated with this keyword.

SYNTAX

THERMAL string; modifier : data; modifier: data

DATA ITEMS

Field	Description	Minimum Size	Maximum Size	Default Name
String	SOTEC thermal data file name	1 character	64 characters	Temfil

STRUCTURE OF EXTERNAL FILE

The external file is for reading time-dependent temperature data. This is a formatted file which may be written by another program designed to calculate temperatures or it may be created by the user. The file is assumed to contain a table, the first column of which is time. The number of data sets in the table is provided in the second line. Further description is provided below.

Line 1: file title—data in this line is for file identification only and is not used anywhere.

Line 2: file section name up to 9 characters long and number of entries in the table. Currently, there is only one section in the thermal data file and its name is 'THERMA'. The number of entries (nsets) in the table must be less than the dimensioning parameter MAXTIM. The format of this line is: (a9,i10). That is, the first nine columns contain the section name and the number of table entries are read in I10 format. If the value of nsets is less than 2 then it is assumed that temperatures do not vary with time.

Lines 3 & 4: header lines, e.g., titles of columns in the table. These two lines are for ease of reading the table only and may be blanks.

Next nsets lines: data to be read. Format is ne10.1 where n is the number of columns in the table, n can be any number from 1 to 4.

Example of tempfil:

temperature data

THERMA 3

time ref temp ave temp

0.	31.5e1	21.2e1
1.e2	27.1e1	18.e1
1.e3	18.e1	13.e1

COMMENTS

The reference temperature refers to the temperature of the container wall while the average temperature is that of the rock. The name of the external file should conform to specific file naming conventions applicable to the particular system on which the user is working.

EXAMPLE

THERMA data on 'THERMAL.DAT' external file.

HEAT load is cell dependent

\\	from cell	to cell	heat load
	1	3	2.1
	4	10	1.1

CRITICAL temperature for liquid water to exist is 98°C |

KEYWORD

THERMAI

MODIFIER

CRITICAL

PURPOSE

To read the temperature at which water exists in a liquid form. It is used to check whether the process of aqueous corrosion can begin.

SYNTAX

CRITICAL temperature = v1 °K

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
v1	Temperature at which water exists in liquid form	Any	Any	96°C

COMMENTS

The temperature should be in °K.

EXAMPLE

THERMAI CRITICAL temperature = 37.3

KEYWORD

TRANSPort parameters

SECTION

This keyword belongs in that section of the input file that specifies all of the near-field transport parameters

PURPOSE

Transport parameters are specified with this keyword and associated modifiers. The modifiers associated with this keyword are RDIFFUision, PDIFFUision, FORWARD rate of matrix dissolution, and flow VELOCity. These modifiers are described in the following pages.

SYNTAX

TRANSPort parameters: modifier, data; modifier, data; ...

DATA ITEMS

All data items are associated with modifiers.

COMMENTS

None.

EXAMPLE

TRANSPort parameters:

RDIFFUision coefficient in rock:

```
\\ from cell to cell zone 1 zone 2 zone 3
   1      5    1.e-6  1.e-6  1.e-5
```

PDIFFUision coefficient in packing material

```
\\ from cell to cell value
   1      5    1.e-4
```

FORWARD rate constant for waste dissolution

```
\\ from cell to cell rate cons colloidal frac
   1      5    1.e-3    0.0
```

VELOCity of flow in file 'VELREF'

```
\\ from cell to cell x-vel fac y-vel fac
   1      5    1.0    0.9
```

KEYWORD

TRANSPort parameters

MODIFIER

FORWARDd dissolution rate

PURPOSE

The forward dissolution rate of the waste and the rate of colloid formation is specified by this modifier.

SYNTAX

FORWARDd dissolution rate: from cell n1 to cell n2, values = v1,v2

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number for this specification	> 0	≤ the number of cells specified with the GEOMETry keyword	1
n2	Ending cell number for this specification	≥ n1	Same as above	MAXCEL
v1	Forward rate of waste matrix dissolution rate	≥ 0	Any	0.0
v2	Initial mass of UO ₂ in each container (Hs)	≥ 0	Any	0.0

COMMENTS

The colloid formation rate is not utilized in Version 1.0 of SOTEC.

EXAMPLE

TRANSPort properties:

FORWARD dissolution rate:

\\	from cell	to cell	dissolution rate	initial mass
	1	5	1.e-6	2E3

KEYWORD

TRANSPort
parameters

MODIFIER

PDIFFUSion
coefficient

PURPOSE

To specify the diffusion coefficient in the packing material.

SYNTAX

PDIFFUSion coefficient: from cell n1 to cell n2, value = v1

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ the number of cells specified by the keyword GEOMETry	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Diffusion coefficient in the packing material	≥ 0	Any	None

COMMENTS

None.

EXAMPLE

TRANSPort parameters

PDIFFUtion coefficient in packing

\\	from cell	to cell	diffusion coef
	1	5	1.e-3

KEYWORD

TRANSPort parameters

MODIFIER

RDIFFUision

PURPOSE

The diffusion coefficients in the three rock zones (damaged, disturbed, and intact rock) are specified by this modifier.

SYNTAX

RDIFFUision coefficient: from cell n1 to cell n2, values = v1,v2,v3

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default value
n1	Beginning cell number	> 0	≤ the number of cells specified with the GEOMETry keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Diffusion coefficient in the damaged rock zone next to borehole wall	≥ 0	Any	None
v2	Diffusion coefficient in disturbed rock zone next to damaged rock	Same as above	Any	None

Field	Description	Minimum Value	Maximum Value	Default value
v3	Diffusion coefficient in intact host rock next to disturbed rock	Same as above	Any	None

COMMENTS

None.

EXAMPLE

TRANSPort parameters

RDIFFUision coefficients:

\\	from cell	to cell	zone 1	zone 2	zone 3
	1	4	1.e-4	1.e-5	1.e-6

KEYWORD

TRANSPort parameters

MODIFIER

VELOCity of liquid

PURPOSE

To specify the liquid phase flow velocity. Either one- or two-dimensional velocity may be specified.

SYNTAX

VELOCity: string; from cell n1 to cell n2, values = v1,v2

DATA ITEMS

Field	Description	Minimum Value or Size	Maximum Value or Size	Default Value
String	Name of the external file containing time-dependent velocity data	1 character	64 characters	'VELFIL'
n1	Beginning cell number of specification of cell dependent 'load' factor	> 0	≤ the number of cells specified with the GEOMETry keyword	1
n2	Ending cell number for the above specification	≥ n1	Same as above	MAXCEL
v1	'Load' factor for velocity in x- or r-direction	Any	Any	0.0

Field	Description	Minimum Value or Size	Maximum Value or Size	Default Value
v2	'Load' factor for velocity in y-direction	Any	Any	0.0

DESCRIPTION OF EXTERNAL FILE

The external file is a formatted file and its structure is described below.

Line 1: Title of the file. This is for easy identification of the file and is not used .

Line 2: The number of data sets (nsets) contained in the file are read in this line. The format is (I10).

Line 3 and 4: These contain headers for the data.

Next nsets lines: the numeric data in format (nE10.1) where n is the number of numeric values in a line. The maximum value of n is 4.

Example of the external file 'VELFIL':

```

VELOCITY DATA
4
time   vel x   vel y
-----
0.0    1.e-4   0.0
1.e2   2.e-4   0.0
1.e3   1.e-4   0.0
1.e4   5.e-3   0.0

```

COMMENTS

The external file contains time-dependent data which is average over the entire repository. The cell 'load' factors are used to get velocities in each cell.

EXAMPLE

```

TRANSPort parameters
  VELOCities: time data on 'VELFIL' file
\\ load factors are given below
\\ from cell to cell   Vx   Vy
   1       5       1.2   0

```

KEYWORD

USER

SECTION

This keyword belongs in the header section of the input file.

PURPOSE

To specify the name of the user. This name will be printed in the output file.

SYNTAX

USER string |

DATA ITEMS

Field	Description	Minimum Size	Maximum Size	Default name
String	Name of the User	1 character	64 characters	None

EXAMPLE

USER: JWALTON |

KEYWORD

WASTE

SECTION

This keyword belongs to that section of the input file that contains all the data about properties and parameters of the waste form.

PURPOSE

Properties and other parameters pertaining to the waste form are specified by this keyword and its associated modifiers. The structure of this input is still under development. In Version 1.0 of *SOTEC*, two modifiers—ELEMENT and AREA are associated with it.

SYNTAX

WASTE characteristics

DATA ITEMS

None. Data is associated with the modifier (see next page).

COMMENTS

Data associated with this keyword will be reorganized in the next version of *SOTEC*.

EXAMPLE

WASTE characteristics
ELEMENT data on file 'ELEFIL'

KEYWORD

WASTE

MODIFIER

AREA

PURPOSE

To specify the wetted surface area of the waste form as a proportion of the total surface area of waste form.

SYNTAX

AREA of waste form: from n1 to n2, value = v1

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
n1	Beginning cell number	> 0	≤ the number of cells specified with the GEOMETRY keyword	1
n2	Ending cell number	≥ n1	Same as above	MAXCEL
v1	Surface area of waste per waste package in the cell	> 0	1.0	1.0

COMMENTS

None.

EXAMPLE

WASTE characteristics

AREA of waste form in a container

\\	from cell	to cell	area
	1	5	2.5
	6	6	2.8

KEYWORD

WASTE

MODIFIER

CARBON

PURPOSETo specify the parameters for the ^{14}C model.**SYNTAX**

CARBON V1, V2, ... V15

DATA ITEMS

Field	Description	Minimum Value	Maximum Value	Default Value
V1	Oxide layer thickness	0	Cladding thickness	None
V2	Grain radius	>0	Any	None
V3	Outer BC	0	Any	None
V4	UO ₂ density	>0	Any	None
V5	D _{ref} for inner oxide layer	>0	Any	None
V6	D _{ref} grain boundary layer	>0	Any	None
V7	E _{act} for diffusion	Any	Any	None
V8	T _{ref}	0	Any	None
V9	Mole ratio	1	Any	None
V10	Radius of UO ₂ fragment	>0	Any	None
V11	Cladding thickness	0	Any	None

Field	Description	Minimum Value	Maximum Value	Default Value
V12	¹⁴ C in fuel	0	Any	None
V13	¹⁴ C in cladding	0	Any	None
V14	¹⁴ C in crud	0	Any	None
V15	¹⁴ C in gap/grain	0	Any	None

COMMENTS

None.

EXAMPLE

WASTE characteristics

AREA of waste form in a container

\\	from cell	to cell	area
	1	5	2.5
	6	6	2.8

*KEYWORD**WASTE**MODIFIER**ELEMENt***PURPOSE**

This modifier is used to specify the names of elements, their isotopes, their occurrence in chains, and their inventory. The entire set of data is read from a formatted file.

SYNTAX

ELEMENt data: string

DATA ITEMS

Field	Description	Minimum Size	Maximum Size	Default name
String	File name containing data on radioelements	1 character	64 characters	'ELEFIL'

DESCRIPTION OF EXTERNAL FILE

The external file is a formatted file and its structure is described below.

Line 1: number of elements (nelem) to be considered (integer)

Line 2: line 2 contains the name of an element, number of chains in which this element appears, elemental solubility, and its sorption (partition) coefficient. The format of each line is (A3,2X,I5,2F10.0). That is, the radioelement name (ELEM) occupies the first 3 columns, columns 4 and 5 are blank, the number of chains (NCON) in which it appears is in columns 6 to 10, the elemental solubility (SOL) in columns 11 to 20, and the sorption coefficient (RDE) in columns 21 to 30.

Next NCON lines: two integer numbers are on each line. The first integer represents the chain number in which the radioelement specified in line 2 appears, the second number is the location of the radioelement in the chain. For example for an element which is the second member (i.e., first daughter of the parent nuclide) of the 4th chain,

this line will appear as: 4 2. As many chains as there are elements may be specified and the number of members in each chain are limited to five.

The above records are repeated for each radioelement.

Next line: the total number of chains and the size of each chain are specified in this line. For example if there are 4 chains and they contain 1,4,5 and 2 members then this line appears as: 4 1 4 5 2.

Next line: For each chain in order, the atomic mass of the isotope, its half life, its inventory in the matrix, and its inventory in the cladding gap is specified. The format is (E10.0,4x,A6,4E10.0). The first ten columns contain the atomic weight, columns 11 to 14 are blank, columns 15 to 20 have the name of the isotope, columns 21 to 30 have the half life, columns 31 to 40, the matrix inventory, and columns 41 to 50, the gap inventory.

Example of 'ELEFIL' file

```

4
  CM      2 .24e-6      100.0
1 1
2 1
  PU      4 .01e-3      100.0
1 2
3 2
4 1
  U       5 0.95e-3      2.0
1 3
1 4
2 3
3 3
4 2
  AM      2 .24e-6      200.0
2 2
3 1
4 4 3 3 2
  246.0   CM246   5.50e03   0.0655   0.00
  242.0   PU242   3.79e05   3.61     0.00
  238.0   U238    4.51e09   0.666    0.00
  234.0   U234    2.47e05   0.0       0.00
  245.0   CM245   9.30e03   0.3066   0.00
  241.0   AM241   4.58e02   3.55     0.00
  241.0   U233    1.62e05   6.01e-5  0.00

```

243.0	AM243	7.95e03	35.9	0.00
239.0	PU239	2.44e04	2.44e04	0.00
235.0	U235	7.10e08	0.0361	0.00
240.0	PU240	6.58e03	11.07	0.00
236.0	U236	2.39e07	0.0538	0.00

COMMENTS

In Version 1.0 of *SOTEC*, the entire waste data is contained in this file. In the next version, this file will be broken into sections for easy readability as has been done with other external input files. The inventory data refers to one single container. Waste form is restricted to spent fuel. The unit of inventory is curies.

EXAMPLE

WASTE characteristics
ELEMENT data on file 'ELEFIL'

5 REFERENCES

- Bazant, Z.P., and L. Cedolin. 1991. *Stability of Structures*. New York, New York: Oxford University Press.
- Carslaw, H.S., and J.C. Jaeger. 1959. *Conduction of Heat in Solids*. Oxford, England: Clarendon Press.
- Code of Federal Regulations. 1992. 40 CFR Part 191. U.S. Government Printing Office, Washington, D.C.
- Code of Federal Regulations. 1992. 10 CFR Part 60. U.S. Government Printing Office, Washington, D.C.
- Einzigler, R.E., and R. Kohli. 1984. Low-temperature rupture behavior of Zircaloy-Clad pressurized water reactor spent fuel rods under dry storage conditions. *Nuclear Technology* 67:107-123.
- Einzigler, R.E., and H.C. Buchanan. 1988. *Long-term, low-temperature oxidation of PWR spent fuel*. WHC-EP-0070. Richland, Washington: Westinghouse Hanford Co.
- Einzigler, R.E. 1991. *Effects of an Oxidizing Atmosphere in a Spent Fuel Packaging Facility*. Proceedings of FOCUS '91: American Nuclear Society: 88-99.
- Einzigler, R.E. 1991. Evaluation of the Potential for Spent Fuel Oxidation Under Tuff Repository Conditions. HEDL-7452. Richland, Washington: Engineering Development Laboratory.
- Einzigler, R.E., S.C. Marschman, and H.C. Buchanan. 1991. Spent-fuel dry-bath oxidation testing. *Nuclear Technology* 94:383-393.
- Einzigler, R.E., H.C. Buchanan, L.E. Thomas, and R.B. Stout. 1992. Oxidation of spent fuel in air at 75 and 195 C. *High-Level Radioactive Waste Management* 1449-1457.
- Farmer, J.C., G.E. Gdowski, R.D. McCright, and H.S. Abluwablia. 1991. Corrosion Model for performance assessment of high-level radioactive-waste containers. *Nucl. Eng. Design*. In press.
- Foster, A.R., and R.L. Wright. 1977. *Basic Nuclear Engineering*. Boston, Massachusetts: Allyn and Bacon. Third Edition.

-
- Gazarelli, F., D. Jorde, R. Manzel. 1980. *Review of PWR Fuel Rod Waterside Corrosion Behavior*. Electric Power Research Institute. Palo Alto California.
- Grambow, B. 1989. *Spent fuel dissolution and oxidation—An evaluation of literature data*. SKB Technical Report: 89-13.
- Henshall, G.A. 1991. *Stochastic models for predicting pitting corrosion damage of HLRW containers*. Proceedings of FOCUS '91, American Nuclear Society: 8.
- Janetzke, R.W., and B. Sagar. 1991a. *preFOR: A Pre-processor for FORTRAN Files*. CNWRA 91-003. San Antonio, Texas: CNWRA.
- Janetzke, R.W., and B. Sagar. 1991b. *RDFREE: A FORTRAN Utility for Format Free Input*. CNWRA-91-005. San Antonio, Texas: CNWRA.
- Kopp, D., and H. Munzel. 1990. Release of volatile carbon-14 containing products from Zircaloy. *J. Nuclear Materials* 173:1-6.
- Kubaschewski, O., and B.E. Hopkins. 1982. *Oxidation of Metals and Alloys*. London, England: Butterworth Publishers: 36.
- Lapidus, L. 1962. *Digital Computation for Chemical Engineers*. New York, New York: McGraw-Hill.
- Macdonald, D.D., and M. Urquidi-Macdonald. 1990. Thin-layer mixed-potential model for the corrosion of high-level nuclear waste canisters. *Corrosion* 46(5):380-390.
- O'Neal, W.C., D.W. Gregg, J.N. Hockman, E.W. Russell, and W. Stein. 1984. *Preclosure analysis of conceptual waste package designs for a nuclear waste repository in tuff*. UCRL 53595. Livermore, California: Lawrence Livermore National Laboratory (LLNL).
- Park, U.S. January 1992. Regulatory overview and recommendations on a repository's release of carbon-14. Science Applications International Corp. San Diego, California.
- Pigford, Th.H., P.L. Chambré, W. Lee. 1992. A Review of Near-Field Mass Transfer in Geologic Disposal Systems, *Radioactive Waste Management and the Nuclear Fuel Cycle* 16(3)(4).
- Press, W.H., B.P. Flannery, S.A. Teukolsky, and W.T. Vetterling. 1989. *Numerical Recipes*. New York, New York: Cambridge University Press.

-
- Sagar, B., and R.W. Janetzke. 1991. *Total System Performance Assessment Computer Code: Description of Executive Module*. CNWRA 91-009. San Antonio, Texas: CNWRA.
- Smith, H.D., and D.L. Baldwin. 1989. *An investigation of thermal release of C-14 from PWR spent fuel cladding*. Proceedings of FOCUS '89. Las Vegas, Nevada: 46-49.
- Thomas, L.E., O.D. Slagle, and R.E. Einziger. 1991. Nonuniform oxidation of LWR spent fuel in air. *J Nuclear Material* 184:117-126.
- Van Konynenburg, R.A., C.F. Smith, H.W. Culham, and C.H. Otto. November 1984. Behavior of carbon-14 in waste packages for spent fuel in a repository in tuff. UCRL-90855. Livermore, California: Lawrence Livermore National Laboratory.
- Van Konynenburg, R.A. 1987. *Carbon-14 in Waste Packages for Spent Fuel in a Tuff Repository. Scientific Basis for Nuclear Waste Management*. Materials Research Society 84:185-196.
- Van Konynenburg, R.A. 1991. *Gaseous Release of Carbon-14: Why the High-Level Waste Regulations Should be Changed*. Proceedings of the 1992 International High Level Waste Management Conference. Las Vegas Nevada: 313-319.
- Watson, M., and J. Postlethwaite. 1990. Numerical simulation of crevice corrosion of stainless steel and nickel alloys in chloride solutions. *Corrosion* 46(7):522-530.
- Williford, R.E. 1991. *Uncertainties in container failure time predictions*. Mat. Res. Soc. Symp. Proc. 212:335-342.
- Wilson, C.N. 1990. Results from NNWSI series 3 spent fuel dissolution tests. PNL-7071. Richland, Washington: Battelle Pacific Northwest Laboratories.
- Woodley, R.E., R.E. Einziger, and H.C. Buchanan. 1989. Measurement of the oxidation of spent fuel between 140 and 225°C. *Nuclear Technology* 85:74-88.