Radcalc: A Computer Program to Calculate the Radiolytic Production of Hydrogen Gas From Radioactive Wastes in Packages

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INTRODUCTION

Radcalc for Windows[1] is a menu-driven Microsoft[2] Windows-compatible computer code that calculates the radiolytic production of hydrogen gas in high- and low-level radioactive waste. In addition, the code also determines U.S. Department of Transportation (DOT) transportation classifications, calculates the activities of parent and daughter isotopes for a specified period of time, calculates decay heat, and calculates pressure buildup from the production of hydrogen gas in a given package geometry. Radcalc for Windows was developed by Packaging Engineering, Transportation and Packaging, Westinghouse Hanford Company, Richland, Washington, for the U.S. Department of Energy (DOE). It is available from Packaging Engineering and is issued with a user's manual and a technical manual (Green et al. 1995). The code has been verified and validated.

USES AND LIMITATIONS

The uses and limitations of Radcalc for Windows are listed in Table 1. Radcalc for Windows calculates the radiolytic production of hydrogen gas. It is designed to be used for low- and high-level radioactive waste such as ion exchange resins, liquids and sludges that may be found in waste tanks or spent fuel basins, liquid samples, and other materials with a density of less than 3 g/cm^3. The code calculates the quantity of hydrogen produced in a container and determines the corresponding percent of the void volume that is filled by the hydrogen. It also calculates the increase in pressure caused by the production of hydrogen. It does not, however, predict the production of hydrogen due to corrosion or chemical reactions. It is also not designed to be used for calculations involving spent nuclear fuel.

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[1]Windows is a trademark of Microsoft Corporation.

[2]Microsoft is a registered trademark of Microsoft Corporation.

Table 1. Radcalc for Windows Uses and Limitations.

Uses	Limitations
Radiolytic production of H_2 gas in high- and low-level radioactive liquids, sludges, ion exchange resins, and other materials with densities of less than 3 g/cm^3	Does not calculate H_2 production due to corrosion or chemical decomposition. Also, cannot be used for H_2 calculations in spent nuclear fuel
Transportation quantities: radioactive; Type A/Type B; limited quantity; low specific activity; highway route controlled quantity; fissile excepted, less than 15 g fissile radionuclides	Calculated on the assumption that the input is valid to three significant figures. Does not check other 49 CFR 173 fissile- excepted criteria
Radionuclide decay calculations	Limited by the radionuclide library (approximately 260 radionuclides)
Decay heat	Calculated at package seal time
Pressure buildup due to H_2 gas generation	Assumes ambient temperature

Radcalc for Windows classifies shipment quantities as radioactive, Type A or Type B, limited quantity, low specific activity, and highway route controlled quantity using DOT regulations and definitions from Title 49 *Code of Federal Regulations* (CFR) Subchapter C, "Hazardous Materials Regulations." It also calculates the quantity of fissile material and determines if the shipment is fissile-excepted based on the mass of fissile isotopes present. It does not check the other criteria for fissile exception that are authorized in 49 CFR 173.453.

Radcalc for Windows contains a robust decay algorithm, which accesses a database of over 260 radionuclides. It calculates the activity of parent and daughter isotopes over a user-specified period of time. In addition, it calculates the rate of decay heat generation at the package seal time.

The code is applicable to the 14 different container geometries listed in Table 2. These containers are routinely used to transport radioactive waste on the Hanford Site in Richland, Washington, and at many other DOE sites. The code can be used for containers not listed in Table 2 by scaling the waste volume or by choosing a container similar in size to the waste matrix.

Radcalc for Windows is available on a 3.5-in floppy diskette and can be installed on an IBM[3]-compatible personal computer. It requires a 386 or higher coprocessor with Microsoft Windows version 3.1 or higher.

HYDROGEN GAS GENERATION IN RADCALC FOR WINDOWS

Historical Background

High- and low-level radioactive wastes produce ionizing radiation, which may cause the radiolytic formation of hydrogen gas. A calculational technique for quantifying the concentration of hydrogen generated by radiolysis in sealed radioactive waste containers was

[3]IBM is a registered trademark of International Business Machines Corporation.

Table 2. Modeled Container Geometries.

Container	Geometry	Inside modeled dimensions (diameter x height)	Outside modeled dimensions	Internal volume
55-gal drum	Cylinder	22.5 in. x 33.25 in. (57.2 cm x 84.46 cm)	22.6 in. x 33.41 in. (57.40 cm x 84.86 cm)	7.65 ft^3 (2.17E5 cm^3)
4x4 liner	Cylinder	120 cm x 120 cm (47.24 in. x 47.24 in.)	NA	47.9 ft^3 (1.36E6 cm^3)
5x5 liner	Cylinder	140 cm x 140 cm (55.12 in. x 55.12 in.)	NA	76.1 ft^3 (2.16E6 cm^3)
6x6 liner	Cylinder	180 cm x 180 cm (70.87 in. x 70.87 in.)	NA	162 ft^3 (4.58E6 cm^3)
30-gal drum	Cylinder	18 in. x 28 in. (45.72 cm x 71.12 cm)	18.1 in. x 28.1 in. (45.97 cm x 71.37 cm)	4.12 ft^3 (1.17E5 cm^3)
85-gal drum	Cylinder	26 in. x 37.9 in. (66.04 cm x 96.27 cm)	26.13 in. x 38.03 in. (66.37 cm x 96.60 cm)	11.6 ft^3 (3.30E5 cm^3)
Doorstop sample carrier	Cylinder	4.5 in. x 5.625 in. (11.43 cm x 14.29 cm)	6.38 in. x 11.38 in. (16.21 cm x 28.91 cm)	0.0518 ft^3 (1470 cm^3)
Ion exchange column	Cylinder	17.5 in. x 69 in. (44.45 cm x 175.3 cm)	18 in. x 69.5 in. (45.72 cm x 176.5 cm)	9.60 ft^3 (2.72E5 cm^3)
LR-56	Cylinder	140 cm x 320 cm (55.12 in. x 126.0 in.)	338 cm x 518 cm (133.1 in. x 203.9 in.)	174 ft^3 (4.93E6 cm^3)
Neutralized current acid waste	Cylinder	3.375 in. x 40.2 in. (8.573 cm x 102.1 cm)	3.75 in. x 48.88 in (9.53 cm x 124.2 cm)	0.208 ft^3 (5890 cm^3)
Onsite transfer cask	Cylinder	2.375 in. x 42.75 in. (6.033 cm x 108.59 cm)	5.505 in. x 45.87 in. (13.98 cm x 116.51 cm)	0.110 ft^3 (3104 cm^3)
PAS-1	Cylinder	18 in. x 21.88 in. (45.72 cm x 55.58 cm)	32.5 in. x 40 in. (82.55 cm x 101.6 cm)	3.22 ft^3 (91300 cm^3)
Sample Pig carrier	Cylinder	2.06 in. x 4.875 in. (5.23 cm x 12.38 cm)	6.71 in. x 9.275 in. (17.04 cm x 23.56 cm)	9.40E-3 ft^3 (22.2 cm^3)
Single pass fuel cask	Box	35.25 in. x 37.5 in. x 46.625 in. (89.54 cm x 95.25 cm x 118.4 cm)	57 in. x 59.25 in. x 69 in. (144.8 cm x 150.5 cm x 175.3 cm)	35.7 ft^3 (1.01E6 cm^3)

developed in a DOE study conducted by EG&G Idaho, Inc., and the Electric Power Research Institute (EPRI) TMI-2 Technology Transfer Office. The study resulted in report GEND-041, *A Calculational Technique To Predict Combustible Gas Generation in Sealed Radioactive Waste Containers* (Flaherty et al. 1986). The study also resulted in a presentation to the U.S. Nuclear Regulatory Commission (NRC), which gained acceptance of the methodology for use in ensuring compliance with NRC Office of Inspection and Enforcement (IE) Information Notice No. 84-72 (NRC 1984) concerning the generation of hydrogen within packages.

NRC IE Information Notice No. 84-72, *Clarification of Conditions for Waste Shipments Subject to Hydrogen Gas Generation*, applies to any package containing water and/or organic substances that could radiolytically generate combustible gases. The NRC requires that the hydrogen gas concentration be less than 5% by volume over a period of twice the expected shipment time.

Notice No. 84-72 is pertinent to shipments of resins, binders, waste sludge, and wet filters. The notice requires compliance by tests or measurements of a representative package. However, in April 1985 the NRC accepted the calculational method presented in report GEND-041 to confirm the absence of hydrogen in low-level waste packages (Deltete 1987).

Subsequently, EPRI developed a simple computer program in a spreadsheet format utilizing the GEND-041 calculational methodology to predict hydrogen gas concentrations. The computer code was extensively benchmarked against TMI-2 (Three Mile Island) EPICOR II resin bed measurements. The benchmarking showed that the model developed predicted hydrogen gas concentrations within 20% of the measured concentrations. The computer code has since been dubbed Radcalc and is still accepted today by the NRC for certificates of compliance for shipping casks containing materials that may result in the radiolytic formation of hydrogen gas. Radcalc for Windows utilizes the computational methodology outlined in GEND-041 to calculate the production of hydrogen gas in the waste matrix of packages.

Theoretical Basis

Radiolysis is the chemical change that occurs in materials as a result of ionizing radiation. The majority of radiolysis that occurs in radioactive waste materials is due to interactions initiated by alpha particles, beta particles, and photons (gamma rays and x-rays). As ionizing radiation travels through a medium, the atoms and molecules within the trajectory of the particle or photon will absorb energy and be ionized or left in an excited state. The quantity of radiolytic products formed will depend upon the material composition, as well as the radiation energy absorbed and the incidence of species recombination.

The types of species produced radiolytically depend upon the chemical structure of the material through which the radiation is traveling. The major reactions in the radiolysis of water are:

$$H_2O \rightarrow e^-_{aq}, OH, H, H_2, H_2O_2, HO_2$$

The addition of substances, such as nitrates, to the water will change the chemical components produced and the quantities of these components.

In order to relate the quantity of a species produced radiolytically to the amount of energy absorbed, a term arbitrarily called the *G value* was defined (Burton 1952). The G value of a material is defined as the number of molecules formed or disassociated per 100 eV energy absorbed. The G values are species and material specific; that is, $G(H_2)$ refers to the G value for the production of hydrogen gas and will differ for different absorbing media. The G values are also radiation specific and depend upon the linear energy transfer of the radiation type. For instance, alpha particles have a much higher linear energy transfer than beta particles, and the G value will be correspondingly greater.

Using the G value concept, the production of hydrogen can be calculated by multiplying the total decay heat or energy produced over a specified period of time of radiation type j for radionuclide i by the fraction of decay heat or energy absorbed in the medium for that radiation type by the medium-specific G value for the jth radiation type. Summing this product for all radiation types and radionuclides in the material and multiplying by the mass of the medium and a conversion factor will result in the quantity of hydrogen produced.

Hydrogen Gas Generation Calculations

The hydrogen gas volume is calculated as

$$H(t) = K \sum_{ij} G_j \, D_i(t) \, \mathscr{E}_{ij} \quad ,$$

where

- i = Index for radionuclide type
- j = Index for radiation type. Radiation types are
  - $j = \alpha$ (hereafter referred to as "heavy particle") which are decays of heavy charged particles, such as alpha and spontaneous fission, as well as delayed neutrons;
  - $j = \beta$ (hereafter referred to as "beta-type") which are all electron-related radiation, such as electrons, positrons, conversion-electrons, and Auger electrons; and
  - $j = \gamma$ (hereafter referred to as "gamma") which includes all electromagnetic radiations such as gamma rays, x-rays, annihilation radiation, and internal bremsstrahlung.
- G = Hydrogen G value for the given radiation type (molecules/100 eV)
- $D_i(t)$ = Total disintegration for the radionuclide over time t
- $\mathscr{E}_{ij}$ = Total energy absorbed
- K = Conversion factor from molecules to volume for an ideal gas at 20 °C and standard pressure (101.325 kPa).

For known discrete gamma radiation, the total energy absorbed is calculated as follows:

$$\mathscr{E}_{i\gamma} = \sum_{k} E_{i\gamma k} A_{i\gamma k} F(\rho, E_{i\gamma k}) + \overline{E'_{i\gamma}} \quad ,$$

- k = Index for each discrete gamma
- $E_{i\gamma k}$ = kth discrete gamma energy
- $A_{i\gamma k}$ = Fraction of decays exhibiting the kth gamma
- $F(\rho, E_{i\gamma k})$ = Fraction of energy absorbed in the waste matrix, as discussed in Chapter 3
- $\overline{E'_{i\gamma}}$ = electromagnetic radiation not accounted for amongst the known discrete known discrete gammas, usually very low energy x-rays.

$\overline{E'_{i\gamma}}$ is calculated from

$$\overline{E'_{i\gamma}} = \overline{E_{i\gamma}} - \sum_{k} E_{i\gamma k} A_{i\gamma k} \quad .$$

where $\overline{E_{i\gamma}}$ is the total average gamma radiation.

For heavy particle and beta-type radiation the total energy absorbed is simply the average decay energy for the given disintegration type per disintegration. E_{ij}, $E_{i\gamma k}$, and $A_{i\gamma k}$ all come from the Fusion Energy Nuclear Data Library (FENDL)/D-1.0 database discussed in the database section.

UNIT CONVERSIONS

The user may input quantities in English units (e.g., inches, feet, pounds) or in units in the International System of Units (e.g., centimeters, meters, grams). Radcalc for Windows will display the input quantities in terms of the units used when entered, as well as in terms of centimeters and grams, which are the units used internally for the calculations.

RADCALC FOR WINDOWS DATABASES

Radionuclide Databases

Radionuclide information for hydrogen gas generation and heat decay rate calculations is taken from the FENDL/D-1.0[4] database. The FENDL/D-1.0 database is the ENDF/B-VI decay database library supplemented by experimental data from the Evaluated Nuclear Data Structure Data File. Decay algorithm calculations are dependent upon the Oak Ridge Isotope Generation and Depletion Code (ORIGEN)2 (Croff 1980) database. ORIGEN2 is managed and distributed by Oak Ridge National Laboratory and is widely accepted and used in national laboratories and the nuclear industry.

Gamma Absorption Fraction Input Parameters

Radcalc for Windows uses the total energy emitted by heavy particle and beta-type decay in calculating the volume of hydrogen produced. However, only a percent of gamma energy will be absorbed in the package and waste. The original spreadsheet (Flaherty 1986) developed to calculate hydrogen gas generation uses fitted curves for four container types to calculate the absorbed gamma dose in the waste material.

For use in Radcalc for Windows, these same four curve fits have been recalculated using the Monte Carlo N-Particle transport code (Breismeister 1993, Carter 1994). Along with the 4 original containers included in the Radcalc spreadsheet, 10 additional containers have been added. The new containers are used in the DOE complex for the transportation of radioactive waste materials.

G Value Data

G values were extensively researched and a list of published G values is presented in document WHC SD-TP-RPT-014 (Green 1994). Radcalc for Windows uses a condensed version of this list. When maximum G values are cited for a material, the G value is used for all radiation types. When G values are missing for a radiation type, they are calculated

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[4]FENDL/D Version 1, January 1992, is a decay data library for fusion (and other) applications. Summary documentation by A. B. Pashchenko. Index No. IAEA-NDS-167 in *Index to the IAEA-NDS-Documentation Series*.

on the basis of G beta = G gamma and G alpha = 4 * G gamma. This approach is in keeping with G value information given in *An Introduction to Radiation Chemistry* (Spinks and Woods 1990).

Transportation Data

Radcalc for Windows uses A_1 and A_2 values to determine classifications. The A_1 and A_2 values were taken from the 49 CFR 173.435 table of A_1 and A_2 values for radionuclides. When A_1 and A_2 values were not available, they were calculated using the methodology outlined in 49 CFR 173.433. All gases are assumed to be uncompressed and the corresponding A_2 value for uncompressed gas was used. The data was extensively checked to verify input values.

CODE VALIDATION AND VERIFICATION

Test cases were designed to exercise the various routines and models programmed into Radcalc for Windows and validate its performance. Each test case includes (1) a discussion of the case, (2) independent check results, and (3) Radcalc for Windows results. Results from each of the test cases are included in the Radcalc for Windows documentation. Radcalc for Windows is accompanied by a set of six test cases designed to ensure the proper installation of the program on a computer system.

SUMMARY

Radcalc for Windows is a user-friendly Windows-compatible software program designed to support the transportation of radioactive materials. It calculates the radiolytic generation of hydrogen gas in the matrix of low- and high-level radioactive waste. It also calculates pressure buildup due to hydrogen generation and the rate of decay heat generation in a package at seal time. It computes the quantity of a radionuclide and its associated products for a given period of time. In addition, the code categorizes shipment quantities as radioactive, Type A or Type B, limited quantity, low specific activity, highway route controlled quantity, and fissile-excepted quantity using DOT definitions and methodologies.

Radcalc for Windows has been extensively tested and validated. It uses NRC-accepted methodology for the calculation of the production of hydrogen gas. The radionuclide database is taken from the well-established FENDL/D 1.0 and ORIGEN2 databases. The transportation database and calculations are based directly on 49 CFR Subchapter C data and methodologies. When used appropriately, Radcalc for Windows can be expected to give accurate and consistent results.

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