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Radcalc for Windows 2.0: Transportation Packaging Software To Determine Hydrogen Generation and Transportation Classification

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RADCALC FOR WINDOWS 2.0: TRANSPORTATION PACKAGING SOFTWARE TO DETERMINE HYDROGEN GENERATION AND TRANSPORTATION CLASSIFICATION

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ABSTRACT

Radcalc for Windows[a] is a user-friendly menu-driven Windows-compatible software program with applications in the transportation of radioactive materials. It calculates the radiolytic generation of hydrogen gas in the matrix of low-level and high-level radioactive waste. It also calculates pressure buildup due to hydrogen and the decay heat generated 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 reportable quantity (RQ), radio-active, Type A or Type B, limited quantity (LQ), low specific activity (LSA), highway route controlled quantity (HRCQ), and fissile excepted using U.S. Department of Transportation (DOT) definitions and methodologies.

I. INTRODUCTION

Radcalc for Windows version 2.0 was developed for the U.S. Department of Energy (DOE) by Westinghouse Hanford Company (WHC) in 1994 and 1995. (Rust Federal Services of Hanford, Inc., and Fluor Daniel Northwest, Inc., among others, took over the WHC contract at the beginning of fiscal year 1997.) The menu-driven Microsoft[b] Windows-compatible computer code has two main applications in the packaging and transportation of radioactive materials. It calculates the radiolytic production of hydrogen gas and determines the percent of hydrogen by volume in packages used for the transportation of radioactive materials. The code's second main application is the classification of radioactive material shipments per DOT regulations. The code also 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. Radcalc has been verified and validated.

The code is available from Packaging Engineering, Rust Federal Services of Hanford, Inc., and is issued with a user's manual and a technical manual.[1] Radcalc for Windows is available on a 3.5-in. floppy disk and can be installed on an IBM[c]-compatible personal computer. It requires a 386 or higher processor with Microsoft Windows version 3.1 or higher.

II. SUMMARY OF FEATURES AND LIMITATIONS

Radcalc for Windows 2.0 calculates the radiolytic production of hydrogen gas. It is designed to be used for analyzing hydrogen production in radioactive ion exchange resins, liquids, and sludges that may be found in tank wastes or spent fuel basins, liquid samples, and other materials with a density between 0.6 and 3 g/cm^3. The code calculates the hydrogen gas generation rate, the quantity of hydrogen produced in a container during seal time, and the corresponding percent by volume of hydrogen gas in the package void volume. Pressure increases caused by hydrogen gas production are also included. Radcalc is not appropriate for calculations involving spent nuclear fuel, nor does it predict the production of hydrogen due to corrosion or chemical interactions.

Radcalc for Windows 2.0 classifies shipment quantities as RQ; radioactive; Type A or Type B; LQ;

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LSA I, II, and III; fissile excepted; and HRCQ using DOT regulations and definitions from the revised Title 49 of the *Code of Federal Regulations* (49 CFR), Subchapter C, "Hazardous Materials Regulations,"[2] effective April 1996.

Radcalc for Windows contains a decay algorithm that accesses a database of over 260 radionuclides. It calculates the activity of parent and daughter isotopes over the specified times. In addition, it calculates decay heat at the package seal time.

The code is applicable to 14 different container geometries, such as 30-, 55-, and 85-gal drums; the LR-56; the PAS-1 cask; ion exchange columns; and others. These containers are commonly used on the Hanford Site in Richland, Washington, and other DOE sites to transport radioactive waste. The code can be used for containers not modeled in the code by scaling the waste volume or by choosing a container similar in size to the waste matrix.

III. HYDROGEN GAS GENERATION IN RADCALC FOR WINDOWS

A. Background

High-level and low-level radioactive wastes produce ionizing radiation that may cause the radiolytic formation of hydrogen gas. A calculational technique for calculating the concentration of hydrogen generated by radiolysis in sealed radioactive waste containers was 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*.[3] The U.S. Nuclear Regulatory Commission (NRC) has also accepted the methodology, called the G-value method, for use in ensuring compliance with NRC Office of Inspection and Enforcement (IE) Information Notice No. 84-72.[4]

NRC IE Information Notice No. 84-72, "Clarification of Conditions for Waste Shipments Subject to Hydrogen Gas Generation,"[4] applies to any package containing water and/or organic substances, such as resins, binders, waste sludge, and wet filters, that could radiolytically generate combustible gases. The NRC requires that the hydrogen gas concentration be less than 5% by volume (or equivalent limits for other flammable gases) of the secondary container void for twice the expected shipment time.

Subsequently, EPRI developed a simple computer program in a spreadsheet format using the G-value method to predict hydrogen gas concentrations. The computer code was extensively benchmarked against Three Mile Island EPICOR II resin bed measurements. The benchmarking showed that the model predicted hydrogen gas concentrations within 20% of the measured values. Radcalc for Windows refines and broadens the approach used in the original spreadsheet to calculate the production of hydrogen gas in the waste matrix of packages.

B. 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. 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 species resulting from the interactions will depend upon the chemical structure of the absorbing medium and, for the most part, will be the same for different types of ionizing radiation. The quantity of the species formed will depend upon the amount of radiation energy absorbed within the material; the proximity of the different species formed that will affect the chances of species recombination; and, in some part, the chemical nature of the different 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*[5] was defined. The G value of a material is defined as the number of molecules formed or disasssociated per 100 eV energy absorbed. 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. G values are also radiation specific and depend upon the linear energy transfer (LET) of the radiation type. For instance, alpha particles have a much higher LET 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.

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[3] 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 code.[6,7] Along with the four original containers included in the Radcalc spreadsheet, ten additional containers have been added. The new containers are used in the DOE complex for the transportation of radioactive waste materials. The gamma absorption fraction ranges from a few percent for small packages to up to approximately 80%.

IV. TRANSPORTATION CLASSIFICATION DETERMINATIONS

Definitions, quantity calculations, and the A_1/A_2 database used in Radcalc for Windows are taken from the revised 49 CFR Subchapter C, "Hazardous Materials Regulations,"[2] effective April 1996. Radcalc for Windows determines shipment classification per DOT for radioactive material; Type A or Type B; LSA I, II, and III; LQ; HRCQ; and fissile excepted. It also calculates transuranic (TRU) waste per the *Hanford Site Solid Waste Acceptance Criteria*.[8]

Radioactive material: The content of a package is classified as radioactive material if the specific activity is greater than 0.002 μCi/g or 70 Bq/g.

Unity fraction/package type determination: The unity fraction is calculated following 49 CFR 173.433(d).[2] If the unity fraction is less than or equal to one, the payload is defined as Type A. If the unity fraction is greater than one, the package is Type B.

Daughter products in Radcalc for Windows: Radcalc for Windows uses the criteria outlined in 49 CFR 173.433(c)[2] for identifying daughter products in transportation classification calculations. In Type A/B, effective A_2, LQ, LSA, HRCQ, and RQ determination daughter products are included in the hazard of the parent, which is represented as the A_1/A_2 value. The definition, as contained in Table 49, is: a single radioactive decay chain is considered to be a single radionuclide when no daughter radionuclide has a half-life either longer than ten days or longer than that of the parent radionuclide. Radcalc for Windows has identified 47 radionuclides that are most frequently shipped as products of a parent radionuclide. These daughters are identified by Radcalc for Windows, and calculations are made accordingly unless the user identifies a daughter product as being shipped by itself as a medical isotope.

LSA determination: The total specific activity must be less than 10^{-6} A_2/g for a package to be eligible for LSA-I. The limit for LSA-II is 10^{-4} A_2/g for solids and gases and 10^{-5} A_2/g for liquids. Water with tritium concentration is LSA-II if it has an activity less than 0.8 TBq/L (20.0 Ci/L). The LSA-III limit, which applies only to solids, is 0.002 A_2/g. For mixed radionuclides, the sum of the ratios of specific activities of each radionuclide to its respective limit must be less than one. It is the responsibility of the user to verify that all other conditions for LSA are met as specified in 49 CFR 173.403[2] and 49 CFR 173.427, including the passage of a leaching test for LSA-III.

LQ determination: The activity limits listed in Table 7 of 49 CFR 173[2] are used for LQs. A mixture of radionuclides is considered an LQ when the sum of the ratios of activities of each radionuclide to its corresponding limit, as indicated in Table 7, does not exceed one and the total quantity of ^{235}U is less than 15 g.

HRCQ determination: A package contains an HRCQ if it contains more than 3,000 times the A_1 or A_2 value or 27,000 Ci or 1,000 TBq. For mixed radionuclides, the quantity is considered an HRCQ if the sum of the ratios of the activity of each radionuclide to its most restrictive corresponding limit exceeds one.

Fissile excepted determination: Radcalc for Windows 2.0 specifies radioactive material as fissile excepted under 49 CFR 173.453(b)[2] if the total mass of the following isotopes is greater than 15 g: ^{238}Pu, ^{239}Pu, ^{241}Pu, ^{233}U, and ^{235}U. If the physical form is liquid and the total mass of ^{238}Pu, ^{239}Pu, ^{241}Pu, ^{233}U, and ^{235}U is not greater than 5 g/10 L, it is also fissile excepted. The code also specifies radioactive material as fissile excepted under 49 CFR 173.453(e)[2] if the mass of ^{239}Pu and ^{241}Pu does not exceed 20% of the total mass of plutonium. This test is only applied when there is no more than 1 kg total of plutonium and there is no ^{233}U or ^{235}U present.

TRU waste determination: Radcalc for Windows 2.0 specifies waste as TRU if the concentration of the following are greater than 100 nCi/g of the waste matrix: ^{233}U, radium sources (^{223}Ra and ^{226}Ra), and all alpha-emitting radionuclides with a Z number greater than 92 and half-lives greater than 20 years. This

classification is based on the criteria given in WHC-EP-0063-04, *Hanford Site Solid Waste Acceptance Criteria*.[6]

RQ determination: The unity fraction for RQ determination is calculated in Radcalc for Windows as the sum of the ratios of activities in curies of each radionuclide to its respective RQ limit in curies per 49 CFR 172.101.[2] If the RQ unity fraction is greater than or equal to one, the package is considered to contain an RQ.

V. UNIT CONVERSIONS

The user may input quantities in English units (e.g., inches, feet, pounds) or in units in the Systeme International (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.

VI. RADCALC FOR WINDOWS DATABASES

A. Radionuclide Databases

Radcalc for Windows utilizes the Fusion Energy Nuclear Data Library (FENDL)/D-1.0[4] database for hydrogen gas generation and heat decay calculations. The FENDL/D-1.0 database is the Evaluated Nuclear Data File/B-VI decay database library supplemented by experimental data from the Evaluated Nuclear Data Structure Data File.

Radcalc for Windows uses the Oak Ridge Isotope Generation and Depletion Code (ORIGEN)2[9] database for decay calculations. ORIGEN2 is managed and distributed by Oak Ridge National Laboratory and is widely accepted and used in national laboratories and the nuclear industry. Radcalc for Windows uses the ORIGEN2 database for decay calculations.

B. G Value Data

The G value is defined as the number of molecules formed or disassociated in a medium per 100 eV of absorbed energy (molecules/100 eV). G values were extensively researched, and a list of published G values is presented in document SD-TP-RPT-014.[10] 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 on the basis of G beta = G gamma and G alpha = 4 * G gamma. This is in keeping with G value information given in *An Introduction to Radiation Chemistry*.[11]

C. Transportation Data

Radcalc for Windows 2.0 uses the definitions and A_1 and A_2 values from the revised Title 49 CFR 173.435[2] effective April 1996. It also uses the *Hanford Site Solid Waste Acceptance Criteria*[6] for the definition of TRU waste.

VII. VERIFICATION AND VALIDATION OF RADCALC FOR WINDOWS

Radcalc for Windows was extensively validated as documented in *Radcalc for Windows Volume II: Technical Manual*.[1] The code is accompanied by verification cases to be run on a system after installation. The verification problems test the models incorporated in Radcalc for Windows and ensure the proper installation of the code on a computer system.

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