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Radcalc: An Analytical Tool for Shippers of Radioactive Material and Waste

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ABSTRACT

The U.S. Department of Energy (DOE) ships radioactive materials in support of its research and development, environmental restoration, and national defense activities. The Radcalc software program assists personnel working on behalf of DOE in packaging and transportation determinations (e.g., isotopic decay, decay heat, regulatory classification, and gas generation) for shipment of radioactive materials and waste.

Radcalc performs:

- The U.S. Department of Transportation determinations and classifications (i.e., activity concentration for exempt material Type A or B, effective A_1/A_2, limited quantity, low-specific activity, highway route controlled quantity, fissile quantity, fissile excepted, reportable quantity, list of isotopes required on shipping papers)
- DOE calculations (i.e., transuranic waste, Pu-239 equivalent curies, fissile-gram equivalents)
- The U.S. Nuclear Regulatory Commission packaging category (i.e., Category I, II, or III)
- Dose-equivalent curie calculations
- Radioactive decay calculations using a novel decay methodology and a decay data library of 1,867 isotopes typical of the range of materials encountered in DOE laboratory environments
- Hydrogen and helium gas calculations
- Pressure calculations.

Radcalc is a validated and cost-effective tool to provide consistency, accuracy, reproducibility, timeliness, quality, compliance, and appropriate documentation to shippers of radioactive materials and waste at DOE facilities nationwide. Hundreds of shippers and engineers throughout the DOE Complex routinely use this software to automate various determinations and to validate compliance with the regulations. The effective use of software by DOE sites contributes toward minimizing risk involved in radioactive waste shipments and assuring the safety of workers and the public.

INTRODUCTION

Radcalc is user-friendly software designed to automate packaging and transportation determinations for shipment of radioactive materials. Radcalc is used throughout the U.S. Department of Energy (DOE) Complex; by multiple Federal, state, and international agencies; and by public and commercial organizations. Radcalc capabilities include:

- Performs transportation classifications based on selected U.S. Department of Transportation (DOT) definitions and methodologies outlined in 49 CFR Chapter I, Subchapter C, "Hazardous Materials Regulations" [1]

- Performs calculations in accordance with selected methods prescribed by the DOE, U.S. Nuclear Regulatory Commission (NRC), U.S. Environmental Protection Agency, and International Conference of Radiological Protection

- Calculates the decay heat and activity of radionuclides and their daughter products at the end of a specified time interval

- Calculates the radiolytic production of hydrogen gas in a radioactive waste matrix

- Calculates the production of helium gas due to radioactive decay.

The capability to automatically import and export data provides users the ability to enter, evaluate, and report information within minutes.

RADCALC 4.0

Historical Development

GEND-041, *A Calculational Technique To Predict Combustible Gas Generation in Sealed Radioactive Waste Containers* [2], documents an EG&G Idaho, Inc., and the Electric Power Research Institute TMI-2 Technology Transfer Office methodology developed for quantifying the concentration of hydrogen generated by radiolysis in sealed radioactive waste containers.

NP-4938, *Methodology for Calculating Combustible Gas Concentration in Radwaste Containers* [3], documents that the GEND-041 methodology is acceptable for use in demonstrating acceptably low concentrations of hydrogen in low-level waste packages in compliance with NRC Office of Inspection and Enforcement Information Notice No. 84-72, *Clarification of Conditions for Waste Shipments Subject to Hydrogen Gas Generation* [4]. Using the GEND-041 methodology, the Electric Power Research Institute developed a simple spreadsheet to predict hydrogen gas concentrations. Three Mile Island EPICOR[1] II resin bed

[1] EPICOR is a trademark of the Epicor Software Corporation, Irvine, California.

measurements served as a benchmark for the spreadsheet showing that the model predicted hydrogen gas concentrations within 20 percent of measured concentrations.

Radcalc 1.0, issued in September 1995, adapted the GEND-041 methodology to calculate the production of hydrogen gas in the waste packages, incorporating a FORTRAN executable code to calculate decay. Radcalc 2.0 incorporated DOT requirements and an improved user interface. Radcalc 3.0 incorporated regulatory changes; consolidated, updated, and substantially expanded the isotopic database; isolated the calculation, package, and user interfaces to facilitate independent modification; and was developed to American Society of Mechanical Engineers (ASME) NQA-1-1994, *Quality Assurance Requirements for Nuclear Facility Applications*, [5] standards. Radcalc 4.0, issued in September 2004, incorporated DOT regulation changes, dose-equivalent curie (DE-Ci) and helium gas calculations, and an enhanced user interface.

DOT Determinations

Radcalc provides the following DOT determinations for radioactive material shipments:

- Radioactive
- Type A or Type B
- Effective A_1 or A_2 for mixture
- Limited quantity
- Low specific activity-I, -II, or -III
- Highway route controlled quantity
- Fissile quantity
- Fissile excepted
- Reportable quantity.

Radcalc also provides a list of isotopes that are required for shipping papers and labels (95 percent of the total A_1/A_2 values). Radcalc has a transportation library including all isotopes identified in 49 CFR 173, "Shippers—General Requirements for Shipments and Packagings" [6], 173.435, "Table of A_1 and A_2 values for radionuclides".

NRC Determination

Radcalc categorizes radioactive contents as Category I, II, or III in accordance with NRC Regulatory Guide 7.11, *Fracture Toughness Criteria of Base Material for Ferritic Steel Shipping Cask Containment Vessels With a Maximum Wall Thickness of 4 inches (0.1 m)* [7].

DOE Determinations

Radcalc categorizes waste using selected DOE requirements including the following:

- Transuranic waste classification in accordance with the DOE Manual M435.1-1, Chg. 1, *Radioactive Waste Management Manual* [8]

- Pu-239 equivalent activity in accordance with Appendix B of DOE/WIPP-02-3122, *Contact-Handled Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant* [9]

- Pu-239 fissile-gram equivalent calculations in accordance with Section 3.1.2 of the *TRUPACT-II Authorized Methods for Payload Control (TRAMPAC)* (WTS 2003) [10].

DE-Ci

Radcalc calculates DE-Ci values in accordance with FGR11/EPA-520/1-88-020, *Limiting Values of Radionuclide Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion, and Ingestion* [11]. In addition, Radcalc calculates DE-Ci values in accordance with ICRP 72, *Age-Dependent Doses to Members of the Public from Intake of Radionuclides: Part 5 Compilation of Ingestion and Inhalation Dose Coefficients* [12].

Radioactive Decay

Radcalc uses a decay data library of 1,867 isotopes to calculate the activity and decay products over a specified period. Radcalc also calculates the decay heat of the nuclear material in a package. The Radcalc nuclear database is based on a combination of the *Fusion Evaluated Nuclear Data Library (FENDL)* (IAEA 1988) [13] and the *Joint Evaluated File (JEF)* (OECD 1993) [14]. The Radcalc atomic mass database is based on "The NUBASE Evaluation of Nuclear and Decay Properties" (Audi 1997) [15]. Users may input source term in grams, becquerels, or curies and convert between units; multiply the source term by a factor to increase or decrease activity; and import data from or export data to other files or applications.

Hydrogen and Helium Gas Calculations

Radcalc can calculate the radiolytic production of hydrogen gas in packages containing radioactive material and hydrogenous material or, alternatively, calculate the $G_{effective}$ value for a specified hydrogen-gas generation rate. Radcalc calculates percent hydrogen in a specified period or will iterate the time needed to reach a specified hydrogen gas concentration. Radcalc contains an extensive database of G values.

Radcalc calculates the production of helium gas due to alpha decay. This calculation utilizes the total number of alpha decays from the decay algorithm and assumes that each alpha decay produces one atom of helium gas.

Pressure Calculations

Radcalc calculates package pressurization associated with hydrogen, oxygen, and helium generated in the package using the ideal gas law and assuming the package is sealed at atmospheric pressure and at 0 °C. Atmospheric pressure and temperature changes are ignored. Radcalc calculates pressure for cases where the hydrogenous material is or is not primarily water. Radcalc assumes that the radiolysis of water produces both hydrogen and oxygen whereas the radiolysis of other materials produces only hydrogen gas.

Limitations

Users are responsible to account for the limitations described below. Users must:

- Understand, interpret, and implement regulations and associated guidance documents
- Verify that Radcalc source documents are appropriate and current for the user's calculations
- Understand the purpose and limits of the reference material selected
- Evaluate both numeric and non-numeric requirements established in the regulations
- Understand the terminology, methodologies, and limitations of hydrogen gas calculations
- Review limitations described in the user documentation
- Regularly review known problems and limitations published on the website.

Quality Assurance

Energy*Solutions* Federal Services, Inc. (Energy*Solutions*), maintains Radcalc on behalf of the DOE Office of Transportation. Energy*Solutions* works under contract to DOE using a quality assurance (QA) program documented in FS-WO-QAPP-001, *Federal Services Hanford Quality Assurance Program Plan* [16], which incorporates the following requirements:

- Eighteen Basic Requirements prescribed in 10 CFR 71, "Packaging and Transportation of Radioactive Materials," Subpart H, "Quality Assurance" [17]

- 10 CFR 72, "Licensing Requirements for the Independent Storage of Spent Nuclear Fuel, High-Level Radioactive Waste, and Reactor-Related Greater than Class C Waste," Subpart G, "Quality Assurance" [18]

- Ten criteria prescribed in 10 CFR 830, "Nuclear Safety Management," and 830.122, "Quality assurance criteria" [19]

- DOE Order 414.1B, *Quality Assurance* [20]

- Eighteen Basic and Supplement Requirements of ASME NQA-1-1994 [5] (and ASME NQA-1-2000 [21]).

As of January 2, 2008, Radcalc will be managed through a new contract between DOE and Project Enhancement Corporation; it is anticipated that Energy*Solutions* will continue in its technical current role, at least in the near term, under the new contract.

Energy*Solutions* implements FS-WO-QAPP-001 [16] using written, approved procedures documented in FSWO-QAP-001, *Quality Assurance Procedures* [22]. Radcalc is Classification Level 1 software (software used in safety class or safety-significant applications) as defined in those procedures.

The *Software Quality Plan* (8/24/04) (DTS 2004) [23]; DTS-SQA-006, *Radcalc Software Requirement Specification* [24]; and DTS-SQA-008, *Radcalc Software Design Description and Implementation Document* [25], document the QA requirements and technical content incorporated in Radcalc 4.0. DTS-VV-021, *Radcalc 4.0 Verification and Validation Test Plan* [26], documents the implementation process for applicable QA requirements. DTS-VV-023, *Radcalc 4.0 Verification and Validation Test Report* [27], documents verification and validation of the software in accordance with the QA program.

User Support

Radcalc is currently available free to users as a service of the DOE Office of Transportation. The software can be downloaded from the Radcalc website (http://www.radcalc.energy.gov) [28], which is maintained on DOE's Consolidated Business Center server. Software documentation is available to registered users.

The Radcalc software comes with a database viewer that allows users to access the Radcalc nuclear, transportation classification, and G value database values on an isotope-by-isotope basis.

User-funded training in the use of Radcalc's transportation and hydrogen gas modules is available periodically when sufficient students express interest. Training may be provided at facilities in Richland, Washington, or may be arranged at sites convenient to the user.

Energy*Solutions* maintains a list of known Radcalc issues on the Radcalc website and distributes user advisors regarding significant new software issues. User input is encouraged.

Direct questions by E-mail or phone to the following individuals:

Bill Willis	WLWillis@energysolutions.com	(509) 375-9532	Technical issues/training
Aaron Schatz	ALSchatz@energysolutions.com	(509) 375-9555	Distribution/web page

RADCALC 4.1

Radcalc is revised periodically to reflect changes in the regulations and customer requirements. Radcalc is in the process of being updated to correct errors, incorporate new regulatory interpretations, and implement other changes identified by DOE, users, and the technical staff.

Current plans anticipate that Radcalc 4.1 will be published in June 2008. A new web interface with enhanced security will be implemented at that time.

The upgrade to Radcalc 4.1 will incorporate the changes identified in problem report/change requests (PR/CR)-023 through -034, which include the following:

- PR/CR-023, *Radcalc 4.0 User, Technical and Database Manuals* [29]

  - Update the user, technical and database manuals as budget and schedule allows

  DTS-SQA-009.1, *Radcalc User's Manual*, Rev. 0 [30] was published in April 2005; Rev. 1 will be published concurrent with the release of Radcalc 4.1 (currently anticipated to be June 2008). Information previously presented in DFSNW-RPT-042, *Radcalc 3.0 Volume II: Technical Manual* [31] and DFSNW-RPT-042, *Radcalc 3.0 Volume IV: Database Manual* [32] are to be consolidated in the DTS-SQA-009.3 [25] when revised for Radcalc 4.1.

- PR/CR-024, *Radcalc 4.0 Unit and Fissile Excepted Errors* [33]

  - Use the International System of Units regulatory values for DOT calculations

  - Remove the "fissile excepted" flag from the low-specific activity output when no fissile material is present.

- PR/CR-025, *Radcalc 4.0 Fissile Excepted Waste Form Error* [34]

  - Allow that the "15 grams or less of fissile material" identified in 49 CFR 173.453, "Fissile materials–exceptions," paragraph (b) may be either solid or liquid

  The "at least 200 grams of solid nonfissile material" identified in 49 CFR 173.453(b) includes both the packaging and the payload.

- PR/CR-026, *Radcalc 4.0 Fissile Excepted Error Involving the Use of the Mass of Packaging* [35]

  - Require that the "at least 2000 grams of nonfissile material" identified in 49 CFR 173.453(c)(1) is limited to only the payload (excluding the packaging)

  The "at least 2000 grams nonfissile material for every gram of fissile material" is the key numerical criteria in 49 CFR 173.453(c). The 360 kg identified in 49 CFR 173.453(c)(2) is not a regulatory criterion; rather "180 grams of fissile material distributed within 360 kg of contiguous nonfissile material" is merely an example of the 2000:1 ratio. Radcalc does not evaluate the qualitative "contiguous" criterion but the user must do so.

- PR/CR-027, *Radcalc 4.0 Incorporating Revisions to WIPP FGE Requirements* [36]

  - Update Pu-239 fissile-gram equivalent (FGE) database values for U-233 and U-235.

  PR/CR-027 references Revision 1 of the *Contact-Handled Transuranic Waste Authorized Methods for Payload Control (CH-TRAMPAC)* (WTS 2005) [37] as the source of FGE values; the CH-TRAMPAC [37] is currently in Revision 2, which reports the same FGE values as Revision 1. Radcalc 4.1 shall be current to Revision 2.

- PR/CR-028, *Radcalc 4.0 Incorporating Revisions to WIPP PE-Ci Requirements* [38]

  - Update Pu-239 equivalent curie (PE-Ci) database values for U-233 and Np-237

  - Add 60 other transuranic isotopes to the calculation.

  PR/CR-028 references Revision 1 of DOE/WIPP-02-3122, *Contact-Handled Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant* [39] as the source of PE-Ci values; DOE/WIPP-02-3122 is currently Revision 6, which reports the same PE-Ci values as Revision 1. Radcalc 4.1 shall be current to Revision 6. It is important to recognize that DOE/WIPP-02-3122 [39] defines "transuranic" differently than the more standard definition established in DOE Manual 435.1-1 Chg 1 [8].

- PR/CR-029, *Radcalc 4.0 Fissile Excepted Error Involving Plutonium* [40]

  - Add a cautionary note if 49 CFR 173.453(f) is used by Radcalc as the basis for identifying a payload as fissile excepted, as follows:

    *The package meets the fissile excepted criteria identified in 49 CFR 173.453(f); however, application of these criteria as written may result in a fissile-excepted designation even though large quantities of fissile material are present. The user should review the issues discussed in the Radcalc Users' Manual and obtain guidance from the competent regulatory authority.

  49 CFR 173.453(f) states: "Packages containing, individually, a total plutonium mass of not more than 1000 grams, of which not more than 20 percent by mass may consist of plutonium-239, plutonium-241, or any combination of these radionuclides." This exception, taken in the context of the introductory paragraph of 49 CFR 173.453, which states: "Fissile materials meeting the requirements of at least one of the paragraphs (a) through (f) of this section are excepted…." could be misinterpreted to allow the shipment of large quantities of fissile uranium.

  In September 2005, DOE formally requested an interpretation from DOT. In January 2006, DOT issued a response to DOE; however, the response was not sufficiently descriptive to resolve the regulatory issue in the software. Based on subsequent conversations, DOE directed that the literal reading as presented in Radcalc 4.0 should remain in the software until DOT provides a more definitive regulatory interpretation. Although a literal

interpretation may result in misclassification of shipments containing elevated quantities of fissile material as fissile excepted, there is currently no regulatory basis for an alternate interpretation. Shippers are responsible to be aware of and evaluate the issues associated with the regulation, and obtain guidance on the issue from a competent regulatory authority.

- PR/CR-030, *Radcalc 4.0 Geff Entry Window Unit Error* [41]

  - Correct a typographical error in a dropdown menu on the G_{eff} Calculation window to indicate units of "cm^3/hr"

- PR/CR-031, *Radcalc 4.0 Error Regarding Lead in Payload* [42]

  - Remove the restriction on the presence of lead in the calculation evaluating 49 CFR 173.453(d); the requirement is "that the mass of any beryllium, graphite, and hydrogenous material enriched in deuterium constitute less than 5 percent of the uranium mass"

- PR/CR-032, *Radcalc 4.0 Error Regarding Moderators and Lead in Packaging* [43]

  - Exclude any lead, beryllium, graphite, and hydrogenous material present in the packaging in determining the mass for solid nonfissile material when evaluating compliance with 49 CFR 173.453(b)

- PR/CR-033, *Radcalc 4.0 Parent/daughter issue – impact on A1/A2 calculations and shipping papers* [44]

  - Account for daughter products in the calculation of A_1/A_2 values as follows:

    1) For those nuclides listed in 49 CFR 173.435, "Table of A_1 and A_2 values for radionuclides" and flagged with footnote (a), Radcalc will exclude contributions from daughter nuclides with half lives less than 10 days to prevent double counting of their contributions; once a daughter is encountered with a half life greater than 10 days, that daughter and subsequent progeny are evaluated as part of a mixture

    2) For those nuclides listed in 49 CFR 173.435 and not flagged with footnote (a), Radcalc will treat the parent and all daughter nuclides as a mixture, and therefore, will include all contributions from daughter nuclides even if the daughters have half lives less than 10 days

    3) For those nuclides not listed in 49 CFR 173.435, the Radcalc database will be flagged to identify those decay chains that meet the criteria in 49 CFR 173.433, "Requirements for determining basic radionuclide values, and for the listing of radionuclides on shipping papers and labels," paragraph (c)(2). Specifically, "a single radioactive decay chain in which the radionuclides are present in their naturally-occurring proportions, and in which no daughter nuclide has a half life either longer than 10 days or longer than that of the parent nuclide, will be considered as a single

radionuclide," and daughter isotopes in these decay chains will be excluded from A_1/A_2 calculations. All other isotopes will be evaluated as part of a mixture and included in A_1/A_2 calculations.

Radcalc can achieve consistency with the regulatory values, in part, by updating the nuclear database using the, *Joint Evaluated Fusion and Fission File* (JEFF-3.1), "Radioactive Decay Data File – Feb 2005" (NEA 2005) [45]. The radionuclide half-lives listed in JEFF-3.1 [45] are generally consistent with the source document (ICRP 38, *Radionuclide Transformations: Energy and Intensity of Emissions* [46]) used in developing the regulatory limits.

The issues identified in evaluating daughter products extend to interpretation of 49 CFR 173.436, "Exempt material activity concentrations and exempt consignment activity limits for radionuclides." Radcalc ignores a daughter in the activity concentration for exempt material and the activity limit for an exempt consignment only if footnote (b) to the table specifically identifies the parent-daughter relationship. Radcalc evaluates all other daughters as part of a mixture.

- PR/CR-034, *Radcalc 4.0 Updates, Enhancements and Minor Modifications* [not yet published]

  - Install Radcalc as a Domain User rather than Power User to simplify installation
  - Add warning regarding significant digits and truncation when importing data
  - Allow data import in grams using either g or gm
  - Expand allowable isotope import naming conventions to include: Barium-137m, Barium 137m, Barium137m, Ba 137m, 137mBa
  - Allow direct source input and import in Ci, μCi, pCi, Bq, TBq
  - Default to user-selected data entry unit
  - Allow keyboard entry for isotope update button (tab from value entry to "Add")
  - Move gamma absorption curve to the G value window
  - Add option to undo/redo actions
  - Add decay-only mode
  - Allow keyboard entry to move from "Checked by" to "Comments" fields
  - Move comment field to top of output page

- Provide a single user advisory that Radcalc does not evaluate all criteria – it is the user's responsibility to do so

- Evaluate A_1/A_2 using 49 CFR 173.435 footnotes (h) and (i), assuming domestic use

- Report gross weight on printed output and make output units consistent with input units

- Add validation report and validation failure flag to software requirement specification

- Update atomic mass and nuclear decay databases, and other physical constants; see PR/CR-033 [44] for discussion of the update of the nuclear database

- Base calculations on Bq rather than atoms to eliminate rounding errors

- Add a unit conversion tool for the convenience of the user.

Future Upgrades

Future initiatives may include:

- Migrating to a web-based application to facilitate more timely updates

- Developing web-based training modules

- Updating the G-value database or revising the G-value methodology consistent with the *CH-TRU Payload Appendices* (DOE 2005) [47]

- Developing means to integrate Radcalc functionality with other software applications, such as the Automated Transportation Management System, Solid Waste Information and Tracking System at the Hanford Site, and RH-TRUCON Maintenance Application at the Waste Isolation Pilot Plant

- Incorporating updates consistent with changes to source documents and regulations including regular harmonization with International Atomic Energy Agency regulations

- Adding capabilities to perform calculations for conveyances in addition to a single package.

CONCLUSIONS

Radcalc is a validated and cost-effective tool to assist in assuring consistency, accuracy, reproducibility, timeliness, quality, compliance, and appropriate documentation to shippers of radioactive materials and waste at DOE facilities nationwide. Hundreds of shippers and engineers throughout the DOE Complex routinely use this software to automate various determinations and to validate compliance with the regulations.

ACKNOWLEDGMENTS

The authors would like to recognize the many who have contributed to development of the Radcalc software and its predecessors over the years and the dedicated technical staff who continue to support and improve the application.

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