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**A CALCULATIONAL TECHNIQUE TO PREDICT  
COMBUSTIBLE GAS GENERATION IN SEALED  
RADIOACTIVE WASTE CONTAINERS**

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## **ABSTRACT**

Certain forms of nuclear waste, when subjected to ionizing radiation, produce combustible mixtures of gases. The production of these gases in sealed radioactive waste containers represents a significant safety concern for the handling, shipment and storage of waste. The U.S. Nuclear Regulatory Commission (NRC) acted on this safety concern in September 1984 by publishing an information notice requiring waste generators to demonstrate, by tests or measurements, that combustible mixtures of gases are not present in radioactive waste shipments; otherwise the waste must be vented within 10 days of shipping.

A task force, formed by the Edison Electric Institute to evaluate these NRC requirements, developed a calculational method to quantify hydrogen gas generation in sealed containers. This report presents the calculational method along with comparisons to actual measured hydrogen concentrations from EPICOR II liners, vented during their preparation for shipment. As a result of this, the NRC recently altered certain waste shipment Certificates-Of-Compliance to allow calculations, as well as tests and measurements, as acceptable means of determining combustible gas concentration. This modification was due in part to work described herein.

## EXECUTIVE SUMMARY

The potential exists for the production of combustible mixtures of gasses in certain waste forms containing radioactive material (e.g., resins, binders, sludge, and wet filters). In sealed containers, radioactive waste gas production could result in pressures above atmospheric pressure. The production of combustible gases in sealed radioactive waste containers has been identified as a significant safety concern relative to the handling, shipping, and storage of radioactive waste.

At Three Mile Island (TMI-2), during preparations to ship 50 EPICOR-II liners containing ion exchange resins, it was revealed that significant concentrations of hydrogen gas were present. As a result, the U.S. Department of Energy (DOE) sponsored various research and development programs addressing gas generation and related safe management of radioactive ion exchange waste. The Nuclear Regulatory Commission (NRC) also sponsored technical studies by the Brookhaven National Laboratory (BNL) on the effects of ionizing radiation on organic ion exchange resin and other radioactive wastes. The NRC studies included a survey on resin use and radionuclide loading at operating nuclear power plants.

It was determined from the research and studies that nuclear wastes subjected to ionizing radiation produce gas through the process of radiolysis and waste degradation. The radiolytic decomposition of the residual water in the resin produces hydrogen and oxygen. In addition, the ionizing radiation acts as a catalyst for a chemical reaction between the oxygen, waste, and residual water in the container. Although hydrogen gas generation increases in an almost linear relationship with increasing curie content, pressure does not increase in a sealed container until an integrated dose is reached in which the oxygen in the air inside the container is consumed by chemical reaction. The ongoing radiolytic process continues to produce gas. Pressure will increase in a sealed container and combustible concentrations of hydrogen gas may result.

The NRC's subsequent evaluation of the hydrogen gas generation problem resulted in the issuance of new requirements (Inspection Enforcement Information Notice No. 84-72: *Clarification of Conditions for Waste Shipments Subject to Hydrogen Gas Generation*) for certain certificates of compliance related to radioactive waste shipment packages. These requirements affect most radioactive waste shipments (resins, binders, waste sludge, and wet filters) from operating nuclear power plants. The NRC requirements are summarized as follows:

- For waste containers that have the potential to radiolytically generate combustible gases, plant personnel must conduct tests and take measurements to determine the factors affecting gas generation. Tests and measurements are not required if radioactivity concentration is less than that for low specific activity (LSA) material and shipped within 10 days of preparation (sealing) or venting.

Utilities (waste generators) were concerned because compliance by conducting tests and taking measurements would be difficult (if not impossible) leaving venting prior to shipment as their only alternative. The venting option is costly financially and in terms of man-rem expenditures. The Utility Nuclear Waste Management Group of the Edison Electric Institute (EEI) formed a Hydrogen Generation Task Force to study and evaluate the new NRC requirements. The task force used direct

operational experience from GPU Nuclear's EPICOR II campaign and technical assistance from the EG&G Idaho/DOE Technical Integration Office at TMI. These efforts resulted in the development of a cost effective method (in man-rem and dollars) to accurately quantify hydrogen gas production in radioactive waste containers and aid utilities in compliance to NRC Information Notice No. 84-72.

Electric Power Research Institute (EPRI) demonstrated the use of this calculational method at TMI. Twenty-eight "non-leaking" EPICOR II liners were used for the demonstration. EPRI used a desk top computer with a spreadsheet program to compare the predicted hydrogen concentration from the EG&G Idaho calculational method to the hydrogen concentration actually measured when the EPICOR II liners were vented during their preparation for shipment. On the average, the predicted hydrogen concentration was within 15 percent of the measured values.

EEI 'Task Force' representatives met with NRC Transportation Branch personnel to discuss the use of calculations to determine hydrogen gas generation in radioactive waste containers. As the basis for discussion, EEI presented the calculational method that EG&G Idaho and DOE developed. EEI requested that calculations be recognized as a means of satisfying the requirements for determining hydrogen gas concentration in radioactive waste containers, as set forth in NRC Inspection Enforcement Information Notice No. 84-72. The NRC acknowledged the validity of the method. Affected certificates-of-compliance have been modified to allow calculations as an acceptable means of assessing gas generation, as well as tests and measurements set forth in NRC Inspection Enforcement Information Notice No. 84-72.

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## **ACRONYMS AND ABBREVIATIONS**

BNL	Brookhaven National Laboratory
DOE	United States Department of Energy
EEI	Edison Electric Institute
EPRI	Electric Power Research Institute
LSA	Low Specific Activity
NBS	National Bureau of Standards
NRC	United States Nuclear Regulatory Commission
SDS	Submerged Demineralizer System
TMI-2	Three Mile Island-Unit 2



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# A CALCULATIONAL TECHNIQUE TO PREDICT COMBUSTIBLE GAS GENERATION IN SEALED RADIOACTIVE WASTE CONTAINERS

## INTRODUCTION

### Background

Nuclear wastes subjected to ionizing radiation produce combustible mixtures of gases in certain waste forms containing radioactive material (e.g., resins, binders, sludge, and wet filters) through the processes of radiolysis and waste degradation. In sealed containers, radioactive waste gas production could result in pressures above atmospheric pressure. The production of combustible gases in sealed radioactive waste containers has been identified as a significant safety concern relative to the handling, shipping, and storage of radioactive waste.

The radiolytic decomposition of the residual water in the resin produces hydrogen and oxygen. In addition, the ionizing radiation acts as a catalyst for a chemical reaction between the oxygen, waste, and residual water in the container. Although hydrogen gas generation increases in an almost linear relationship with increasing curie content, pressure does not increase in a sealed container until an integrated dose is reached in which all the oxygen in the air inside the container is consumed by chemical reaction. As the ongoing radiolytic process continues to produce gas, pressure will increase in a sealed container and combustible concentrations of hydrogen gas may result.

Three Mile Island Unit 2 (TMI-2) used various ion exchange media systems to process contaminated accident waste water. One system, EPICOR II, used organic ion exchange resins to process more than 500,000 gal of waste water, resulting in the generation of 72 prefilter liners with curie loadings ranging from 160 to 2200 Ci. Loadings in 22 of the liners were low enough to permit their disposal in a commercial low-level waste site, while the remaining 50 liners were stored at TMI-2. Subsequent preparations to ship these 50 liners revealed the presence of significant concentrations of hydrogen gas. As a result, the U.S. Department of Energy (DOE) sponsored various research and development programs addressing gas

generation and related safe management of radioactive ion exchange waste. The Nuclear Regulatory Commission (NRC) also sponsored technical studies by the Brookhaven National Laboratory on the effects of ionizing radiation on organic ion exchange resin and other radioactive wastes. The NRC studies included a survey of operating nuclear power plants on resin use and radionuclide loading.

**Parameters of Gas Generation.** The DOE and NRC gas generation research efforts and actual TMI-2 on-site (EPICOR II) direct measurements and sampling revealed that certain conditions and specific relationships exist. These include:

- Ion exchange media subjected to ionizing radiation generate a variety of gases (hydrogen, oxygen, carbon dioxide, carbon monoxide, and methane) from radiolysis and resin degradation. This generation occurs at low curie loadings as well as at higher levels.
- Hydrogen gas generation rates in radioactive waste containers increase at a nearly linear rate with increasing curie content.
- Pressure in a sealed wet radioactive waste container initially decreases due to oxygen depletion, masking hydrogen production. The pressure will continue to decrease until the delivered dose is sufficient to deplete all of the gaseous oxygen.
- The time necessary to reach a given hydrogen gas concentration, in a sealed radioactive waste container, is predictable.
- United States commercial nuclear power generating plants typically ship radioactive wastes that are well below the present NRC permissible radionuclide loading limit (10 Ci/ft<sup>3</sup>); however, a few have exceeded this limit.

**NRC Requirements.** The NRC's subsequent evaluation of the hydrogen gas generation problem resulted in the issuance of new requirements (Inspection Enforcement Information Notice No. 84-72: *Clarification of Conditions for Waste Shipments Subject to Hydrogen Gas Generation*) for certain certificates of compliance related to radioactive waste shipment packages. The following paragraphs summarize these requirements, which address hydrogen gas generation and applicable safe storage and shipment periods.

For waste containers that have the potential to radiolytically generate combustible gases, plant personnel must conduct tests and take measurements to determine the factors affecting gas generation. Specifically, the following criteria must be observed during a period twice the expected shipment time:

1. Hydrogen gas concentration must be limited to no more than 5 percent by volume of the secondary container gas void, *or*
2. The secondary container and cask cavity must be inerted to ensure that oxygen is limited to 5 percent by volume in those portions of the waste package that could have greater than 5 percent hydrogen gas.

The above is not required if radioactivity concentration is less than that for Low Specific Activity (LSA) material and shipped within 10 days of preparation (sealing) or venting.

These NRC requirements affect most radioactive waste shipments (resins, binders, waste sludge, and wet filters) from operating nuclear power plants. Although the problem of hydrogen gas generation has been seen only in containers with greater than Type A quantities of waste (as defined in 10 CFR 61), waste generators should be aware that the same physical conditions exist in containers with Type A quantities of waste as well, even though the time to reach hazardous conditions is much longer.

Waste generators were concerned because these new NRC requirements seemed to be too conservative. Compliance by conducting tests and taking measurements would be difficult (if not impossible) for many waste generators, leaving venting prior to shipment as their only alternative. The venting option is costly financially and in terms of man-rem expenditures (where the concern is to keep dose rates as-low-as-reasonably-achievable).

**Hydrogen Generation - Calculation and Comparison.** The Utility Nuclear Waste Management Group of the Edison Electric Institute (EEI) formed a Hydrogen Generation Task Force to study and evaluate the new NRC requirements. The task force used direct operational experience from GPU Nuclear's EPICOR II campaign and technical assistance from the EG&G Idaho/DOE Technical Integration Office at TMI. This evaluation resulted in EG&G Idaho/DOE's development of a calculational method (presented in the next section of this report) to quantify hydrogen gas generation in sealed containers. The interrelation of gas production resulting from radioactive decay in the waste form, the amount of energy retained in the waste container (resulting from the decay process), and the free volume of the container are discussed, and examples given.

The Section on EPICOR Gas Measurements discusses how theoretical predictions, made with the calculational method, were checked against actual gas measurements from representative waste packages. The result of this study is discussed and clarifying appendices are presented. Appendix A gives an example of how the values from various tables and figures are used to predict combustible gas concentrations. Appendix B is a discussion of the characteristics of various waste forms that have the potential to generate combustible quantities of gas. Appendix C contains a detailed description of the computer code used to determine the absorbed dose in different container geometries with various waste forms. Appendix D provides a mathematical description of energy absorption for a spherical source and supplements the discussions presented in Appendix C.

## METHOD CALCULATION

The calculational method for predicting combustible gas generation in sealed radioactive waste containers, described in this section, is based on DOE and NRC research and General Public Utility Nuclear Corporation's (GPU Nuclear) Three Mile Island EPICOR II experience. An effort has been made to extract pertinent findings from these resources, organizing the information into a unified format. Data requirements and analytical capabilities of typical waste generators were evaluated, and the format was modified to facilitate implementation by potential users of the model. Certain parameters that require mainframe computer computation have either been tabularized or are presented in graphic form. Presented below is a complete description of the calculational method developed to quantify combustible gas generation.

### Formulae and Empirical Data

Combustible gas generation in sealed radioactive waste containers involves a combination of factors that can be addressed by considering the following:

- The production of gas in the waste form resulting from radioactive decay
- Amount of energy resulting from the decay process that is retained in the waste container and absorbed by the waste
- Free volume of the container, including interstitial voids inherent in the waste form.

These three factors and their interrelations are discussed below.

**Gas Production in the Waste Form Resulting from Radioactive Decay.** The quantity of gas produced is a function of the amount of energy absorbed by the waste form. Gas formation is due primarily to radiolysis caused by the radioactive decay energy of nuclides present in the waste. NRC studies performed at Brookhaven National Laboratory (BNL) indicate that the amounts of combustible gases formed by radiolysis are approximately linear with dose. The relationship between gas production and energy absorption is expressed as the waste's gas generation constant or G-value, which is described as the number of radicals or molecules

formed or decomposed per 100 eV absorbed. A number of G-values have been reported in the literature.<sup>1-4</sup> Those of concern to typical waste generators are the G-values for dewatered, or swollen resin, and solidified waste. These gas generation constants range from 0.1 to 0.6 molecules per 100 eV (See Table 1). G-Values for mixed resin bed systems can be approximated using the weight percent mix of the components.

**Radioactive Decay Energy Absorbed by the Waste Form.** The amount of energy absorbed by a waste form as a result of the radioactive decay process is a function of curie loading, waste properties, and container geometry. The primary decay modes for consideration by typical waste generators are beta, including other electron emissions, and gamma emission. For practical purposes, all beta and associated secondary radiation emission energies are assumed to be deposited in the waste. This is because of the relatively short travel length, or range, (as compared with gamma rays) required for the beta particle to give up its energy in the waste material, and the extremely low probability of a beta particle escaping the container. The gamma energy absorbed by the waste depends on the strength of the gamma emission, the amount of gamma ray energy absorbed by interaction (collision) with a waste particle, and the number of particles with which the gamma ray interacts. Because an interaction will either absorb or, more likely, attenuate an emission, the fraction of gamma energy absorbed depends on the number of interactions possible. Therefore, gamma energy absorption increases with increasing numbers of waste particles. For this reason, gamma absorption is a function of container waste density and geometry.

The total energy absorbed in the waste is the sum of the beta and secondary radiation decay energy and that portion of the gamma ray energy retained in the waste's matrix. In order to calculate combustible gas generation, it is necessary to determine the absorbed energy at a specific time after the waste container is loaded and sealed. This is accomplished by the use of equations that consider nuclide loading curie content; waste properties such as density, geometry, gamma ray attenuation; and absorption coefficients. The result of this exercise yields an absorbed energy dose in rads (radiation absorbed dose) for each nuclide in the waste at a time after sealing. The contributions of

**Table 1. Hydrogen gas generation constants ( $G_H$ )<sup>a</sup>**

Resin	Ionic Form	$G_H$ (Molecules/100 eV) <sup>b</sup>
DOWEX 50WX10	Li <sup>+</sup>	0.11
DOWEX 50W10	H <sup>+</sup>	0.095
ZEO-KARB 215	H <sup>+</sup>	0.12
IRN-77	H <sup>+</sup>	0.13 ± 0.02
IRN-78	OH <sup>-</sup>	0.6
IRN-150	HOH	0.5
IRN-77	Na <sup>+</sup>	0.2
IRN-78	Cl <sup>-</sup>	0.3
IRN-150	NaCl	0.3

a. The hydrogen gas generation yield for a mixed bed system is the sum of the yields of the individual components. For example, a mixed bed containing 0.40 resin A and 0.60 resin B by weight with  $A:G_H = x$  and  $B:G_H = Y$  is

$$G_H(A + B) = 0.4x + 0.6Y$$

For beds of unknown composition use

anion bed (OH <sup>-</sup> )	$G_H = 0.6$
cation bed (OH <sup>-</sup> )	$G_H = 0.13$
solidified resin/cement	$G_H = 0.24$
asphalt	$G_H = 0.41$

b. These values are the result of experimental analysis and appear as reported, for swollen (dewatered) resins from NUREG/CR-2830 and NUREG/CR-3383; cement values from NUREG/CR-2969, and values for asphalt from NUREG/CR-50617. The G-value is defined as the number of molecules formed or decomposed per 100 ev absorbed.

the beta, secondary, and gamma components of each nuclide are computed using the following equations:

$$\text{Beta absorbed dose (rads)} = D_i^\beta(t)$$

$$= \frac{AC_i}{\lambda_i} (\bar{E}_i) (1 - e^{-\lambda_i t})$$

Secondary radiation absorbed dose (rads)

$$= D_i^x(t) = \frac{AC_i}{\lambda_i} (E_i^x) (1 - e^{-\lambda_i t})$$

Gamma absorbed dose (rads) =  $D_i^\gamma(t)$

$$= \frac{AC_i}{\lambda_i} (E_i^\gamma) (1 - e^{-\lambda_i t})$$

Combining equations yields  $D_i^{\beta,x,\gamma}(t)$

$$= \frac{AC_i}{\lambda_i} (\bar{E}_i + E_i^x + E_i^\gamma) (1 - e^{-\lambda_i t})$$

where

$D_i^{\beta,x,\gamma}(t)$  = total (beta plus secondary plus gamma) absorbed dose delivered by the  $i^{\text{th}}$  nuclide at time  $t$ , in rads

$t$  = the time after sealing the container in years

$A$  = a proportionality constant equal to  $1.84 \text{ E} + 10 \text{ rad-gram/MeV-yr-Curie}$

$C_i$  = the specific activity of the  $i^{\text{th}}$  nuclide in curies/gram

- $\lambda_i$  = the decay constant of the  $i^{\text{th}}$  nuclide in years<sup>-1</sup>
- $\bar{E}_i$  = average beta energy in MeV of the  $i^{\text{th}}$  nuclide
- $E_i^x$  = the absorbed x-ray energy in MeV of the  $i^{\text{th}}$  nuclide
- $E_i^\gamma$  = the absorbed gamma ray energy in MeV of the  $i^{\text{th}}$  nuclide.

The total cumulative absorbed dose for all nuclei and decay modes at time (t) is found from

$$D_{\text{Total}} = \sum_i D_i^{\beta, x, \gamma}(t) .$$

A discussion of various absorption parameters is presented below.

**Beta Absorbed Dose**—The beta absorbed dose in the waste is calculated using the value for average beta energy ( $\bar{E}_i$ ) for a nuclide. The average beta ray energy is approximately one third of the sum of the possible beta emissions multiplied by the relative abundance of the emission. For example, Iodine-131 emits two beta particles; 85 percent of the disintegrations result in a 0.608 MeV beta, while the remaining 15 percent are 0.315 MeV beta. Therefore,

$$\bar{E}_i = 1/3(0.85 \times 0.608 + 0.15 \times 0.315)$$

$$\bar{E}_i = 0.187 \text{ MeV.}^a$$

These values have been tabularized and are available in standard health physics and nuclear handbooks. The beta absorbed dose is then determined by

$$D_i^\beta(t) = \frac{AC_i}{\lambda_i} (\bar{E}_i) (1 - e^{-\lambda_i t}) .$$

This is done for each beta emitting nuclide. Summing the beta dose for all nuclides at a time (t) yields the total beta absorbed dose for the waste.

**Absorbed Dose Due to Secondary Radiations**—Secondary radiations ( $E_i^x$ ), such as x-rays and Auger electrons, result from the transition of a nuclide from an excited state to the ground state. All emissions with energies of 100 keV or less

are considered to be totally absorbed by the waste form, and to participate in gas production.

Values for secondary radiations are available in standard handbooks<sup>5</sup>. The contribution to absorbed dose from secondary radiations for each nuclide in the waste is determined by

$$D_i^x(t) = \frac{AC_i}{\lambda_i} (E_i^x) (1 - e^{-\lambda_i t}) .$$

Summing the secondary radiation contribution for each nuclide present in the waste yields the total absorbed dose due to secondary radiations.

**Gamma Absorbed Dose**—Determination of the gamma absorbed dose ( $E_i^\gamma$ ) is more complex than calculating the beta absorbed dose because the total energy of the gamma emission may not be deposited in the waste form. The fraction of gamma energy deposited is a function of the waste's physical characteristics and waste container geometry. The gamma energy absorbed by the waste is calculated for each nuclide present using

$$E_i^\gamma = \sum_j [(n_{ij}) (f_{ij}) (E_{ij}^\gamma)]$$

where

$E_i^\gamma$  = the gamma energy absorbed in MeV for the  $i^{\text{th}}$  nuclide.

$\sum_j$  = the summation of the fractions of the gamma ray energies absorbed for all gamma emissions of the  $i^{\text{th}}$  nuclide.

$n_{ij}$  = the abundance of the  $j^{\text{th}}$  gamma ray per decay of the  $i^{\text{th}}$  nuclide.

$f_{ij}$  = the fraction of energy, of the  $j^{\text{th}}$  gamma ray of the  $i^{\text{th}}$  nuclide that is absorbed in the waste.

$E_{ij}^\gamma$  = is the energy in MeV, of the  $j^{\text{th}}$  gamma ray of the  $i^{\text{th}}$  nuclide.

Example:

Cobalt 60 decays by emitting two gamma rays, 1.173 MeV and 1.332 MeV, both having an abundance of 1.0. The nuclide is present in waste with a density of 1 g/cm<sup>3</sup> contained in a 55 gal drum.

For the 1.173 MeV gamma ray

$$n_{ij} = 1.0, f_{ij} = 0.54, \text{ and } E_{ij}^\gamma = 1.173 \text{ MeV.}$$

a. This is an approximate value, the value used is 0.1817 MeV from Reference 5.



For the 1.332 MeV gamma ray

$$\begin{aligned} n_{ij} &= 1.0, f_{ij} = 0.53, \text{ and } E_{ij}^\gamma \\ &= 1.332 \text{ MeV} \\ (1.0)(0.54)(1.173 \text{ MeV}) &+ (1.0)(0.53)(1.332 \text{ MeV}) \\ &= 1.339 \text{ MeV} \end{aligned}$$

Therefore, of the 2.505 MeV emitted in the gamma decay of Cobalt 60, 1.339 MeV is absorbed by the waste.

Values for  $n_{ij}$  and  $E_{ij}^\gamma$  were obtained from Reference 5. Values for  $f_{ij}$  can be obtained from Figures A-1 through A-4 in Appendix A, or computed by the methods outlined in Appendices A, B, and C.

The contribution to absorbed dose from gamma ray emission for each nuclide in the waste is

$$D_i^\gamma(t) = \frac{AC_i}{\lambda_i} (E_i^\gamma) (1 - e^{-\lambda_i t})$$

Combining the beta, secondary, and gamma absorbed doses for each nuclide yields

$$\begin{aligned} D_i^{\beta,\gamma}(t) &= \frac{AC_i}{\lambda_i} (\bar{E}_i + E_i^\beta \\ &+ E_i^\gamma) (1 - e^{-\lambda_i t}) \end{aligned}$$

This is the absorbed dose in rads in the waste at time (t) due to the decay of the  $i^{\text{th}}$  nuclide. The process must be repeated for all nuclides present in the waste to arrive at the total absorbed dose at time (t) after sealing.

$$D_{\text{Total}}(t) = \sum_i D_i^{\beta,\gamma}(t)$$

**Free Volume of the Container.** The free volume of the container is the sealed internal container volume not occupied by waste.

$$\text{Free Volume} = \text{Internal Container Volume}$$

$$- \text{Waste Volume}$$

The waste itself may contain interstitial voids that can be included in the free volume. The interstitial voids can be expressed as a void fraction of the waste. The void fraction can be determined from the bulk and true densities of the waste. Values for these properties are available for organic resins from vendor supplied information.

$$\text{Void Fraction} = \frac{\text{True Density} - \text{Bulk Density}}{\text{True Density}}$$

Void fractions for mixed systems, such as cation and anion resins, can be approximated using weight percentages of each component.

$$\begin{aligned} \text{Free Volume} &= (\text{Internal Container Volume} \\ &- \text{Waste Volume}) + (\text{Waste Volume} \\ &\times \text{Void Fraction}) \end{aligned}$$

The free volume for solidified waste is the internal container free space over the solidified waste. Credit for internal voids can be taken if the porosity of the solidified mixture is known or can be calculated.

#### **Gas Generation Constants, Absorbed Dose, and Allowable Hydrogen Concentration.**

The three components of the combustible gas generation calculation can be assembled to predict hydrogen volume at a given time. Combining the equations developed in previous sections yields

$$\text{Hydrogen volume} = \text{G-factor}$$

$$\times \text{total absorbed dose} \times \text{mass of waste}$$

$$\times \text{constant}$$

This equation is expressed mathematically as

$$V_H = G_H \times D_{\text{Total}} \times M_W \times 2.33$$

$$\times 10^{-6} \text{ eV, cm}^3/\text{rad gram molecule.}$$

The proportionality constant,  $2.33 \times 10^{-6} \text{ eV, cm}^3/\text{rad gram molecule}$  is determined from

$$\frac{100 \text{ Erg/gram}}{\text{rad}} \times \frac{6.25 \times 10^{11} \text{ eV}}{\text{Erg}}$$

$$\times \frac{1 \text{ gram mole}}{6.02 \times 10^{23} \text{ molecules}}$$

$$\times \frac{22.4 \text{ liters}}{\text{gram mole}} \times \frac{1000 \text{ cm}^3}{\text{liter}}$$

$$= 2.33 \times 10^{-6} \text{ eV, cm}^3/\text{rad gram molecule.}$$

The total absorbed dose is a function of time.

NOTE: The G-factor,  $G_H$  is expressed

as  $\frac{\text{molecules}}{100 \text{ eV}}$  .

The hydrogen concentration is obtained by dividing the hydrogen volume ( $V_H$ ) by the free volume of the waste container ( $V_F$ ).

## PRACTICAL CONSIDERATIONS

Considering the number of variables and the complexity of the calculation, a shielding computer code (QAD-FN) was used to determine the absorbed dose in different container geometries with various waste forms. The code was modified to calculate the waste's internal absorbed dose. This was accomplished by locating the modeled detector points inside the container waste matrix.

The computer calculates the absorbed gamma dose by evaluating the dose received at up to 200 detector sub-volumes (as a result of irradiation by a maximum of  $1 \times 10^6$  source sub-volumes). The detector sub-volume gamma absorbed doses are summed to yield the waste's total absorbed gamma dose. Several assumptions are made:

- The container is full to the normal fill point
- The waste is of uniform density
- Gamma absorption constants are generated using energy absorption coefficients for water (see Appendix B)
- The nuclides are uniformly distributed in the waste matrix.

A more detailed description of the computer code model and its application is provided in Appendix C.

Defining many of the variables in the foregoing sections requires complex calculations and in some cases, computer analysis. Much of this work has already been done. What follows is a description of the tables, graphs, and assumptions used to categorize waste forms and a method to simplify the calculational technique into a "user friendly" format.

The attenuation and absorption characteristics of most waste materials (anion and cation resins, concrete, asphalt, and water) are similar in the energy ranges normally encountered in radioactive waste. The amount of gamma ray energy absorbed by the waste is a function of the source strength energy and waste container geometry. A study of flux to dose relationships and absorption characteristics of common waste materials was performed to determine the most appropriate means of calculating gamma absorption. A comparison of absorption profiles for these waste forms found water to be the limiting waste (See Appendix B). The gamma absorbed dose is expressed as a fraction of total gamma energy for a specific container type.

Figures A-1 through A-4 express the gamma absorbed energy fraction as a function of container geometry, waste density, and gamma ray energy. These graphs were generated using the QAD-FN shielding code mentioned earlier and described in Appendix C. Appendix A gives an example of how the values from various tables and figures, along with pertinent facility information, are used to predict combustible gas concentrations.

## EPICOR GAS MEASUREMENTS

An important element in the overall program to justify use of a calculational technique to predict radiolytic gas generation was the need to 'benchmark' theoretical predictions against actual gas measurements in representative waste packages. Although there has been considerable research in this area (nearly all published by the NRC), most of the published work in the United States has been directed toward laboratory scale investigations to determine the constants for gas generation as a function of deposited energy (e.g., the G-value), or to assess the effects of organic resin degradation under high integrated radiation doses. There has been little study of actual size and similarly configured waste packages representative of nuclear utility wastes.

The exception to this was the work at TMI-2 with the experience gained during shipment of the EPICOR II prefilters and Submerged Demineralizer System (SDS) zeolite liners. Although the hydrogen generation characteristics for each of these waste forms had been evaluated and published,<sup>1-4</sup> the applicability of the reported hydrogen generation rates were questioned due to the higher curie loading in relation to normal power plant resin waste. The SDS liners were considered to be too highly loaded with cesium and strontium for direct use in 'benchmarking' the EG&G model. However, it was felt that the EPICOR II prefilters might offer sound operational data on hydrogen generation rates for use in validating the current calculational technique.

Consequently, it was determined that an in-depth review of actual gas measurement data from TMI-2 dewatered resin water, particularly the EPICOR II prefilters, would be undertaken with the following objectives:

- Assess the reported EPICOR II hydrogen generation data
- Determine, if possible, the hydrogen generation rate in EPICOR II prefilters by methods independent of the techniques used in Reference 6.
- Assess the linearity of long-term average hydrogen generation rates as a function of deposited curies, in order to extrapolate to curie loadings more representative of utility wastes

- Summarize the available measurements of hydrogen gas in EPICOR II prefilters for use in 'benchmarking' the EG&G calculational model
- Develop a simple computer program which could accurately predict the generation of gases in representative waste containers.

### Methodology

Hydrogen gas measurement data taken during preparations to ship the initial 50 EPICOR II prefilters was obtained from the following sources:

- "Preparations to Ship EPICOR Liners"<sup>6</sup>
- Completed TMI-2 Operating Procedure No. 2104-4.137, "EPICOR Liner Inerting," for each prefilter shipped
- GPU Nuclear correspondence transmitting prefilter summary data to EG&G Idaho prior to shipment from TMI.

During the initial review of available data to support this current evaluation, it became clear that several characteristics of the EPICOR II liners (and the contained resin media) had the potential to significantly affect the hydrogen data gathered during the sampling evolutions at TMI. These characteristics were judged to have an even greater influence on the prediction of hydrogen evolution using the EG&G calculational model. Therefore, considerable effort was devoted to quantifying accurately each liner's characteristics in order to ensure that the correlations and conclusion developed during the evaluation were based upon the best data available.

Much of the information subsequently used in this evaluation had to be summarized to preclude violation of the proprietary agreement with EPICOR, Inc. Several of the areas requiring detailed consideration are discussed in the following paragraphs.

- The residual, or interstitial, moisture content of irradiated resin media directly influences the generation of hydrogen, primarily because the production mechanism involves the radiolysis of water. Since different types

of resin media were used in the EPICOR II prefilters, it became necessary to estimate the residual moisture content from liner dewatering tests performed at TMI, manufacturer's literature, the NRC, industry studies of resin dewatering, and knowledge of the actual resin material.

- Similarly, quantification of the free volume contained within an individual EPICOR II liner became necessary. This volume would directly affect not only the hydrogen concentration buildup in the liner, but also the calculation of hydrogen volume based upon measured concentration at liner opening. Accurate resin loading data was necessary for this determination also, since the free volume surrounding a specific resin is a function of the resin's size, physical and chemical form, and shrink/swell characteristics. Again, the required level of detail for this evaluation was obtained for all 50 prefilters.
- For purposes of correlating hydrogen generation against deposited energy, the most accurate method is to calculate total integrated dose, a key element of the EG&G model. The actual curie calculations used to support liner transportation and acceptance regulations by DOE for disposal, were used in this regard to calculate the total integrated dose to the resin material.
- In calculating the total hydrogen volume present within any liner when opened, pressure and temperature corrections are required to convert the gas to volume at standard temperature and pressure (STP). This correction was deemed necessary for this study to ensure that the effects of atmospheric temperature and pressure did not influence the projection of hydrogen present in the liner. Actual average atmospheric pressure for the day each liner was opened was obtained from the National Weather Service and was used as the appropriate correction factor for pressure. Although there was a wide variation in daily temperature, it was concluded that the location of the EPICOR II liners deep within their storage modules made actual temperature variations slight. Furthermore, the overall effects on gas volume at STP from temperature variations of the

magnitudes expected in this situation are much less than the effects from pressure variations. Therefore, no temperature correction was used when calculating hydrogen volume from opened liners.

- Many of the 50 EPICOR II prefilters were suspected to be unable to completely contain the buildup of internal gases, due to small leaks in gasket materials, penetration seals, etc. Although any suspected leaks were not expected to jeopardize the containment of radioactive material, hydrogen gas would certainly be able to escape from the liner due to its small molecular size. Actual data during the opening of several liners yielded no measured hydrogen with the absence of a positive (and in several instances, a negative) internal pressure, thereby confirming the existence of liner leaks. In order to differentiate between leaking and non-leaking liners for the purpose of performing hydrogen correlation studies, an opening gas pressure of 0.2 psig was used to define a non-leaking liner.

Using this definition, a total of 28 liners were considered to be "non-leaking" and therefore suitable for detailed investigation.

- The actual dates each liner was sealed when removed from process service, and then re-opened prior to inerting for shipment, were used to accurately define the storage interval. This time interval was then used to compute individual isotope decay correction for calculation of total absorbed energy.
- The hydrogen gas generation constant ( $G_H$ ) for each individual liner was determined based upon a weighted average of the resin type (e.g., cation, anion, mixed bed) multiplied by the NRC published gas constant for the specific resin type. This method is consistent with that explained in the method calculation section of this report. Since the actual EPICOR-II resin media remains proprietary information, a detailed breakdown of these calculations cannot be reported herein.

## EPICOR-II Prefilter Comparison

The concentration of hydrogen gas expected in each of the 28 non-leaking EPICOR II prefilters was calculated using the hydrogen prediction methodology developed by EG&G Idaho and discussed earlier. The purpose of this effort was to compare predicted concentrations with the concentrations actually measured when these liners were vented for shipment after nearly three years of storage.

For ease of practical application, the EG&G Idaho theoretical model was implemented on a desk top computer "electronic spreadsheet" program specifically developed by EPRI for this purpose. This spreadsheet resulted in a simple to use computer program for predicting the generation of gases in the EPICOR II liners.

EPICOR II prefilter resin and liner characteristics necessary for use with the EG&G Idaho methodology were obtained using the same data and methods described previously. Additional parameters were obtained from EG&G's calculational results summarized earlier in this report.

Table 2 summarizes the results of the calculations using the spreadsheet program. In addition to basic data (liner number, deposited curies, waste volume, specific activity, and years in storage) for each prefilter, a comparison of the terminal (i.e., after several years of sealed storage) hydrogen concentrations actually measured [percent  $H_2$  (M)] and the theoretically predicted hydrogen concentrations [percent  $H_2$  (P)] using the EG&G Idaho model is also presented. Based upon results of the previous investigation, which concluded that only 28 of the original 50 prefilters were relatively leak tight, only these 28 liners were subjected to this comparison.

On the average, the predicted hydrogen gas concentrations were within 20% of the measured values. The mean ratio of measured to predicted concentrations was 84%, with a standard deviation of 11%. The calculational model over predicted the terminal hydrogen concentration in all but one of the liners, thereby providing a consecutively safe prediction of the generation. (These averages do not consider PF-10 which was over predicted by 51%.)

**Table 2. EPICOR II prefilters**

PF Number	Ci	Waste (ft <sup>3</sup> )	Ci (ft <sup>3</sup> )	Years Storage	Percent Hydrogen		Percent Measured/ Predicted
					Measured (M)	Predicted (P)	
10	156.2	32	4.9	2.99	2.80	5.49	51
13	986.4	32	30.8	3.30	18.90	23.75	80
14	954.1	32	29.8	3.24	20.30	23.48	86
15	961.3	32	30.0	3.27	23.20	23.80	97
17	1184.0	32	37.0	3.27	18.90	27.48	69
18	1367.0	32	42.7	2.70	26.00	26.98	96
19	1342.0	33.5	40.1	2.83	25.00	27.32	92
20	1316.2	34.5	38.2	2.59	22.40	27.22	82
21	1316.2	34.5	38.2	3.21	21.50	30.89	70
22	1154.2	34.5	33.5	3.21	22.30	28.15	79
23	895.9	34	26.3	3.01	15.50	22.29	70
24	1316.2	34	38.7	3.20	21.20	28.72	47
25	1316.2	34	38.7	2.77	22.70	26.31	86
26	1316.2	34	38.7	2.73	25.60	26.03	98
29	1316.2	34	38.7	2.87	20.50	26.75	77
30	973.1	34	28.6	3.00	17.20	28.87	60
31	1109.6	34.5	32.2	3.13	22.50	27.36	82
32	1109.6	34.5	32.2	2.75	23.90	25.15	95
33	1109.5	34	32.6	2.82	23.50	23.76	99
34	1109.6	34	32.6	2.88	19.20	24.10	80
35	1109.6	34.5	32.2	2.67	26.20	24.63	106
36	1109.5	34.5	32.2	2.71	18.60	24.89	75
37	1109.6	34	32.6	2.95	20.10	27.67	73
38	1109.6	34	32.6	2.85	21.60	27.03	80
39	1109.5	34	32.6	2.77	23.90	26.52	90
41	1109.5	34	32.6	2.98	19.70	26.18	75
42	1109.5	34	32.6	2.64	21.40	34.20	88
43	1109.5	34	32.6	2.76	24.10	24.88	97
Non-Leaking: Pressure > 0.2 psig Percent Hydrogen > 15%						Average:	82
						w/o PF-10:	84
						St Dev:	13
						w/o PF-10:	11

## RESULTS

Electric Power Research Institute (EPRI) demonstrated the use of this calculational method at TMI-2. Twenty-eight "non-leaking" EPICOR II liners were used for the demonstration. EPRI used a desk top computer with a spreadsheet program to compare the predicted hydrogen concentration from the EG&G Idaho calculational method to the hydrogen concentration actually measured when the EPICOR II liners were vented during their preparation for shipment, after nearly three years of storage on site. On the average, the predicted hydrogen concentration was within 20% of the measured values.

EEI 'Task Force' representatives met with NRC Transportation Branch personnel to discuss the use

of calculations to determine hydrogen gas generation in radioactive waste containers. As the basis for discussion, EEI presented the calculational method that EG&G Idaho and DOE developed. EEI requested that calculations be recognized as a means of satisfying the requirements for determining hydrogen gas concentration in radioactive waste containers, as set forth in NRC Inspection Enforcement Information Notice No. 84-72. The NRC acknowledged the validity of the method. Affected Certificates-Of-Compliance have been modified to allow calculations as an acceptable means of assessing gas generation, as well as tests and measurements set forth in NRC Inspection Enforcement Information Notice No. 84-72.



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**APPENDIX A**  
**SAMPLE PROBLEM**

## APPENDIX A

### SAMPLE PROBLEM

#### Discussion

Worksheets A-1 and A-2 can be completed from plant records, vendor information supplied to plants, and tables and graphs in this appendix. Worksheet A-1 requires information from Table A-1 for Step 8, and vendor information for Step 6. Step 5 can be determined from vendor information or plant measurements. Worksheet A-2 values are obtained from Table A-2, Figures A-1 through A-4 (whichever figure is appropriate as to container type) and plant records. Column A of Worksheet A-2 is found by dividing the number of curies of a nuclide present by the mass of the waste (Worksheet A-1, Step 7). The calculations are admittedly tedious. Therefore, the technique should be thoroughly understood before attempting use. This method is valid for both dewatered resins and solidified wastes.

The sample problem uses data from NUREG/CR 2830 on a high level reactor cleanup resin, shipped from the Vermont Yankee Plant in 1980, for isotopic activity density values.

#### Figures A-1 Through A-4

**Gamma Fraction Absorbed.** These graphs<sup>a</sup> provide values for Worksheet A-2, column E. Find gamma ray energy(s) of nuclides in Table A-2 and place in column C of Worksheet A-2. Use this energy (column C) to find the gamma fraction absorbed by intercepting the density line closest to the waste density<sup>b</sup> from Step 5, Worksheet A-1. Read the fraction on the y axis and place this value in column E, Worksheet A-2. In the example problem, the waste density from Table A-2, Step 5, is 0.78 g/cm<sup>3</sup>—so the 0.8 g/cm<sup>3</sup> line is used. For example, a 1 MeV gamma would be 0.48 absorbed.

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a. These graphs are valid for both resin and solidified wastes.

b. If the waste density differs from the plotted values, use the density line that is greater—but closest to the actual waste density. More exact values for the gamma fraction absorbed as a function of density can be obtained using the methods outlined in Appendix C.

## Worksheet A-1

Step	Variable	Value
1.	Volume of waste container (ft <sup>3</sup> x 2.83 E + 4 cm <sup>3</sup> /ft <sup>3</sup> )	<u>1.83 x 10<sup>5</sup> cm<sup>3</sup></u>
2.	Heights (interior) of waste container	<u>72 cm</u>
3.	Radius (interior) of waste container	<u>27 cm</u>
4.	Volume of waste (ft <sup>3</sup> x 2.83 E + 4 cm <sup>3</sup> /ft <sup>3</sup> )	<u>1.65 x 10<sup>5</sup> cm<sup>3</sup></u>
5.	Bulk density of waste (lbs/ft <sup>3</sup> x 1.6 E-2 g cm <sup>3</sup> /lb/ft <sup>3</sup> )	<u>0.78 g/cm<sup>3</sup></u>
6.	True density (specified gravity) of waste	<u>1.3 g/cm<sup>3</sup></u>
7.	Mass of waste (Step 4 x Step 5)	<u>1.29 x 10<sup>5</sup> g</u>
8.	Radiolytic hydrogen generation constant, G <sub>H</sub> (See attached Table A-1)	<u>0.3<sup>a</sup> molecules/100 eV</u>
9.	Waste void space (expressed as a decimal) (Step 6 - Step 5) / Step 6	<u>0.40</u>
10.	Free volume (Step 1 - Step 4) + (Step 9 x Step 4)	<u>8.4 x 10<sup>4</sup> cm<sup>3</sup></u>
11.	Maximum allowable hydrogen volume (0.05 x Step 10)	<u>4.2 x 10<sup>3</sup> cm<sup>3</sup></u>
12.	Absorbed dose for Step 11 [Step 11/(Step 8 x Step 7 x 2.33 E-06)]	<u>4.7 x 10<sup>6</sup> rad</u>

a. 0.65% cation  
 0.35% anion  
 $G_H = (0.65 \times 0.13) + (0.35 \times 0.6)$   
 $G_H = 0.3$

**Worksheet A-2.**

Nuclide	Curies	Column							
		A <sup>a</sup> (activity density)	B <sup>b</sup> (decay constant)	C <sup>b</sup> (gamma energy)	D <sup>b</sup> (abundance)	E <sup>c</sup> (% absorption)	F <sup>d</sup> (E <sub>γ</sub> )	G <sup>b</sup> (E <sub>γ</sub> )	H <sup>b</sup> (E <sub>γ</sub> )
Mn-54	0.770	5.98 10 <sup>-6</sup>	0.809	0.835	1.0	0.51	0.430	0.0	0.0052
Co-58	0.757	5.88 10 <sup>-6</sup>	3.575	0.811	1.0	0.51	0.410	0.0290	0.0053
				1.675	0.0054	0.44	0.0040	—	—
				0.511	0.299	0.53	0.081	—	—
Co-60	3.93	3.05 E-5	0.132	1.173	1.0	0.48	0.560	0.0958	—
				1.332	1.0	0.47	0.630	—	—
Zn-65	2.61	2.03 E-5	1.036	1.115	0.508	0.48	0.270	0.0020	0.0077
				0.511	0.0283	0.53	0.0077	—	—
Cs-134	2.29	1.78 E-5	0.336	0.475	0.0146	0.53	0.0037	0.157	0.0055
				0.567	0.238	0.52	0.070	—	—
				0.605	0.976	0.52	0.307	—	—
				0.797	0.941	0.51	0.382	—	—
				1.039	0.0100	0.49	0.005	—	—
				1.168	0.0180	0.48	0.010	—	—
				1.365	0.0304	0.47	0.020	—	—
Cs-137/Ba-137	6.24	4.84 E-5	0.023	0.662	0.8998	0.52	0.310	0.171	0.0657

a. a = Ci/mass of Waste (Worksheet A-1 Step 7).

b. From Table A-1.

c. Figure A-1 through A-4, fraction of a gamma energy absorbed.

d. Column F = Column C x Column D x Column E.

**Table A-1. Hydrogen gas generation constants ( $G_H$ )<sup>a</sup>**

Resin	Ionic Form	$G_H$ (Molecules/100 eV) <sup>b</sup>
DOWEX 50WX10	$Li^+$	0.11
DOWEX 50W10	$H^+$	0.095
ZEO-KARB 215	$H^+$	0.12
IRN-77	$H^+$	$0.13 \pm 0.02$
IRN-78	$OH^-$	0.6
IRN-150	HOH	0.5
IRN-77	$Na^+$	0.2
IRN-78	$Cl^-$	0.3
IRN-150	NaCl	0.3

a. The hydrogen gas generation yield for a mixed bed system is the sum of the yields of the individual components. For example, a mixed bed containing 0.40 resin A and 0.60 resin B by weight with  $A:G_H = x$  and  $B:G_H = Y$  is

$$G_H(A + B) = 0.4x + 0.6Y$$

For beds of unknown composition use

anion bed ( $OH^-$ )	$G_H = 0.6$
cation bed ( $OH^-$ )	$G_H = 0.13$
solidified resin/cement	$G_H = 0.24$
asphalt	$G_H = 0.41$

b. These values are the result of experimental analysis and appear as reported, for swollen (dewatered) resins from NUREG/CR-2830 and NUREG/CR-3383; cement values from NUREG/CR-2969, and values for asphalt from NUREG/CR-50617. The G-value is defined as the number of molecules formed or decomposed per 100 ev absorbed.

**Table A-2. Radionuclide decay properties**

Radionuclide Constants	Half Life Years	Decay Constant	Beta Energy Average MeV	Low X-Ray Energy Average MeV	Gamma Energy MeV/Disintegrations	Intensity Abundance Fraction
H-3	1.228 E + 1	5.643 E-2	.0057	0.0	0.0	0.0
C-14	5.730 E + 3	1.209 E-4	.0495	0.0	0.0	0.0
Cr-51	7.585 E-2	9.137 E + 0	0.0	.0047	.3201	.0983
Mn-54	8.561 E-1	8.095 E-1	0.0	.0052	.8348	.9998
Fe-55	2.700 E + 0	2.567 E-1	0.0	.0056	0.0	0.0
Co-57	7.417 E-1	9.344 E-1	0.0	.0232	.1230	.9720
Co-58	1.938 E-1	3.575 E + 0	.0290	.0053	.6920	.0016
					.8110	1.0010
					1.6750	.0054
Fe-59	1.222 E-1	5.671 E + 0	.1175	0.0	.5110	.2986
					.1920	.0433
					1.0992	.5850
Ni-59	7.500 E + 4	9.240 E-6	0.0	.0067	1.2916	.4320
					0.0	0.0
					0.0	0.0
Co-60	5.271 E + 0	1.315 E-1	.0958	0.0	1.1732	1.0000
					1.3325	1.0000
Ni-63	1.001 E + 2	6.923 E-3	.0171	0.0	0.0	0.0
Zn-65	6.691 E-1	1.036 E + 0	.0020	.0077	1.1155	.5075
					.5110	.0283
Kr-85	1.072 E + 1	6.465 E-2	.2505	0.0	.5140	.0043
Sr-89	1.384 E-1	5.007 E + 0	.5830	0.0	.9091	.0002
Sr-90	2.860 E + 1	2.423 E-2	.1958	0.0	0.0	0.0
Y-90		2.423 E-2	.9348	0.0	0.0	0.0
Y-91	1.602 E-1	4.326 E + 0	.6023	0.0	1.2049	.0030
Nb-94	2.030 E + 4	3.414 E-5	.1458	.0011	.7026	1.0000
					.8711	1.0000
					.7242	.4370
Zr-95	1.753 E-1	3.954 E + 0	.1161	0.0	.7567	.5530
					.7658	.9981
					0.0	0.0
Nb-95	9.599 E-2	7.220 E + 0	.0433	.0009	0.0	0.0
Tc-99	2.130 E + 5	3.254 E-6	.0846	0.0	0.0	0.0
Ru-103	1.077 E-1	6.432 E + 0	.0674	.0032	.4970	.8940
					.6100	.0635
					.0397	.0007
Rh-103m		6.432 E + 0	0.0	.0388	0.0	0.0
Ru-106	1.008 E + 0	6.874 E-1	.0100	0.0	.5119	.2060
					.6210	.1040
					1.2510	.0055
Cd-109	1.270 E + 0	5.455 E-1	0.0	.0196	0.0	0.0
Ag-110m	6.927 E-1	1.000 E + 0	.0655	.0016	.6170	1.3540
					.8580	1.4115
					1.4520	.4260
Sn-113	3.151 E-1	2.199 E + 0	0.0	.0232	.2551	.0193
In-113m		2.199 E + 0	0.0	.1360	.3917	.6490
Sn-119	8.022 E-1	8.639 E-1	0.0	.0837	0.0	0.0
Sn-123	3.537 E-1	1.959 E + 0	.5203	0.0	1.0886	.0060
Sb-124	1.648 E-1	4.205 E + 0	.3776	.0200	.6030	.9830
					.6460	.0740
					.7240	.1580
					1.0060	.0378
					1.3540	.0545
					1.4750	.0381
					1.6910	.4900
					2.0910	.0573



Table A-2. (continued)

Radionuclide Constants	Half Life Years	Decay Constant	Beta Energy Average MeV	Low X-Ray Energy Average MeV	Gamma Energy MeV/Disintegrations	Intensity Abundance Fraction
Sb-125	2.770 E + 0	2.502 E-1	.0865	.0254	.1760 .3800 .4370 .6170	.0815 .0224 .4020 .3590
Te-125m	1.588 E-1	4.364 E + 0	0.0	.1410	.1093	.0028
I-129	1.570 E + 7	4.414 E-8	.0409	.0380	.0396	.0752
I-131	2.201 E-2	3.148 E + 1	.1817	.0124	.2840 .3640 .6370 .7230	.0696 .8140 .0777 .0180
Cs-134	2.062 E + 0	3.361 E-1	.1570	.0055	.4750 .5670 .6050 .7970 1.0390 1.1680 1.3650	.0146 .2380 .9760 .9410 .0100 .0180 .0304
Cs-136	3.656 E-2	1.896 E + 1	.1001	.0388	.0805 .1680 .2740 .3410 .5070 .8190 1.0480 1.2350	.2290 .2690 .1270 .4910 .0098 .9970 .7960 .1970
Cs-137	3.017 E + 1	2.297 E-2	.1708	0.0	0.0	0.0
Ba-137m		2.297 E-2	0.0	.0657	.6616	.8998
Ba-140	3.553 E-2	1.951 E + 1	.2720	.0384	.1630 .3050 .4300 .5370 .3290 .4840 .8160 .9250 1.5960 2.4890	.0687 .0450 .0535 .2500 .2160 .4840 .3350 .1050 .9550 .0442
La-140	4.655 E-3	1.489 E + 2	.5269	.0056	.1454 .1340 1.4890 2.1850	.4840 .1190 .0030 .0077
Ca-141	9.028 E-2	7.676 E + 0	.1447	.0316		
Ce-144	7.897 E-1	8.775 E-1	.0827	.0140	.6960 2.1860	.0148 .0097
Pr-144		8.775 E-1	1.2072	.0008		

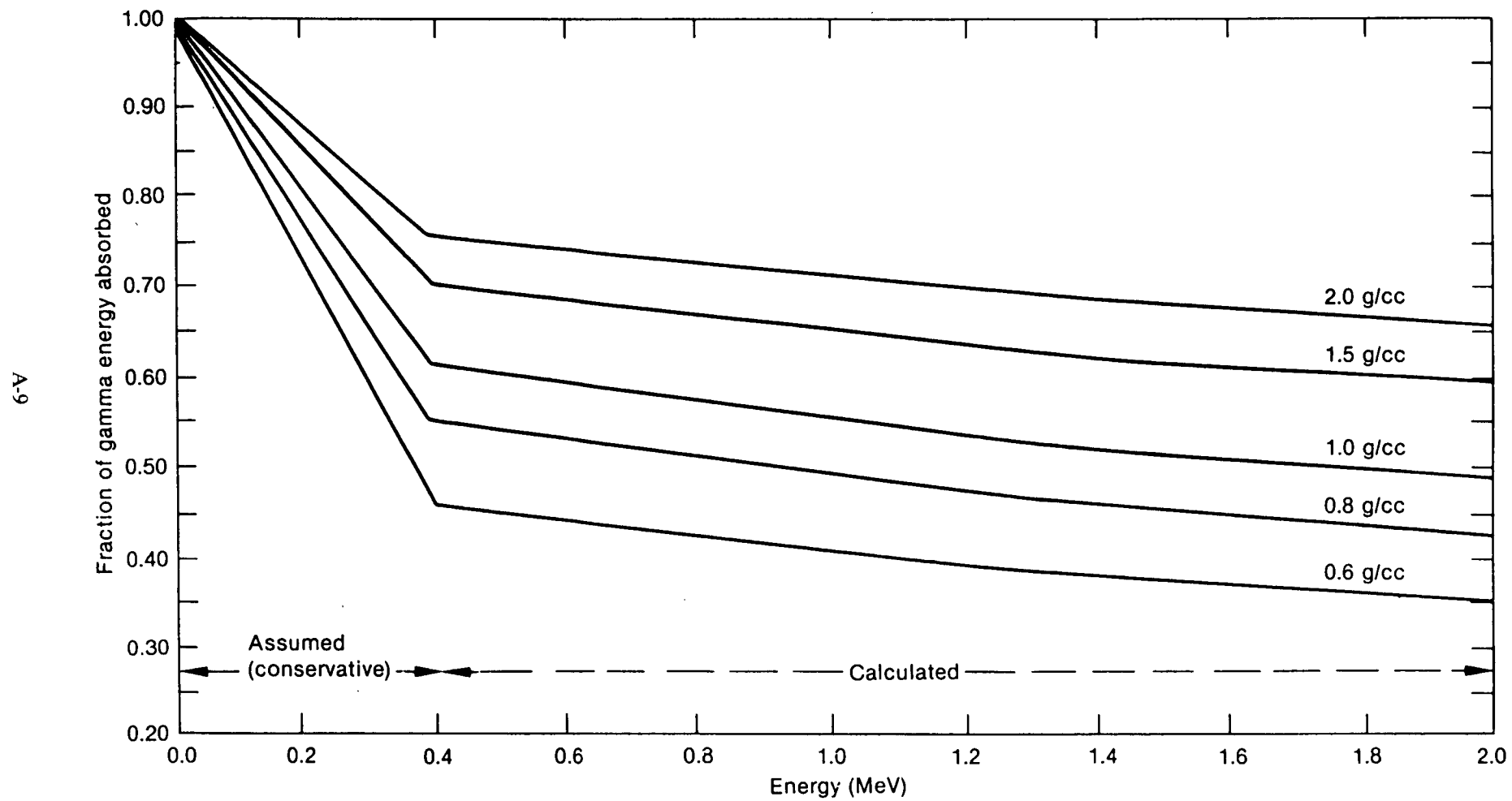
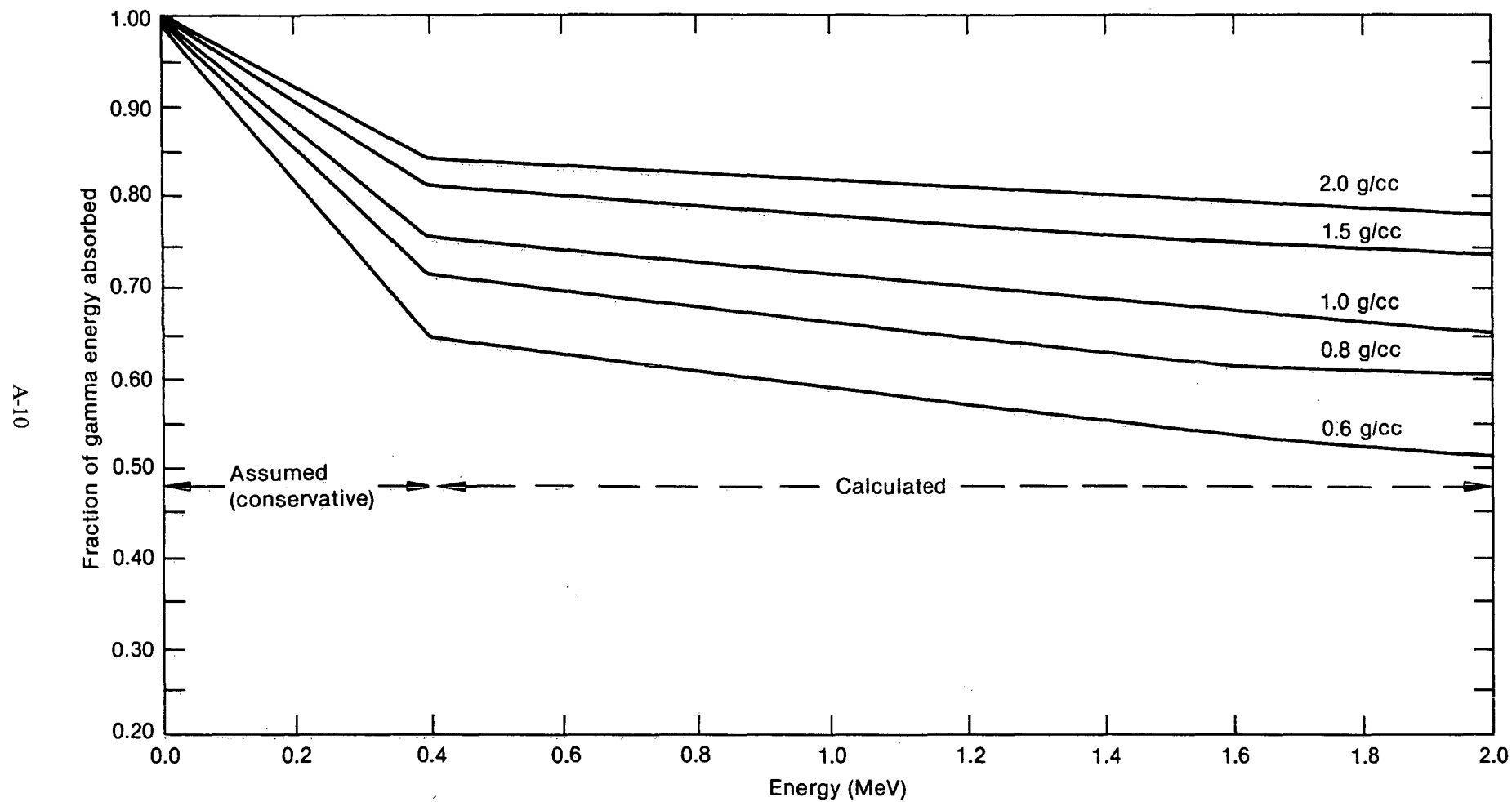
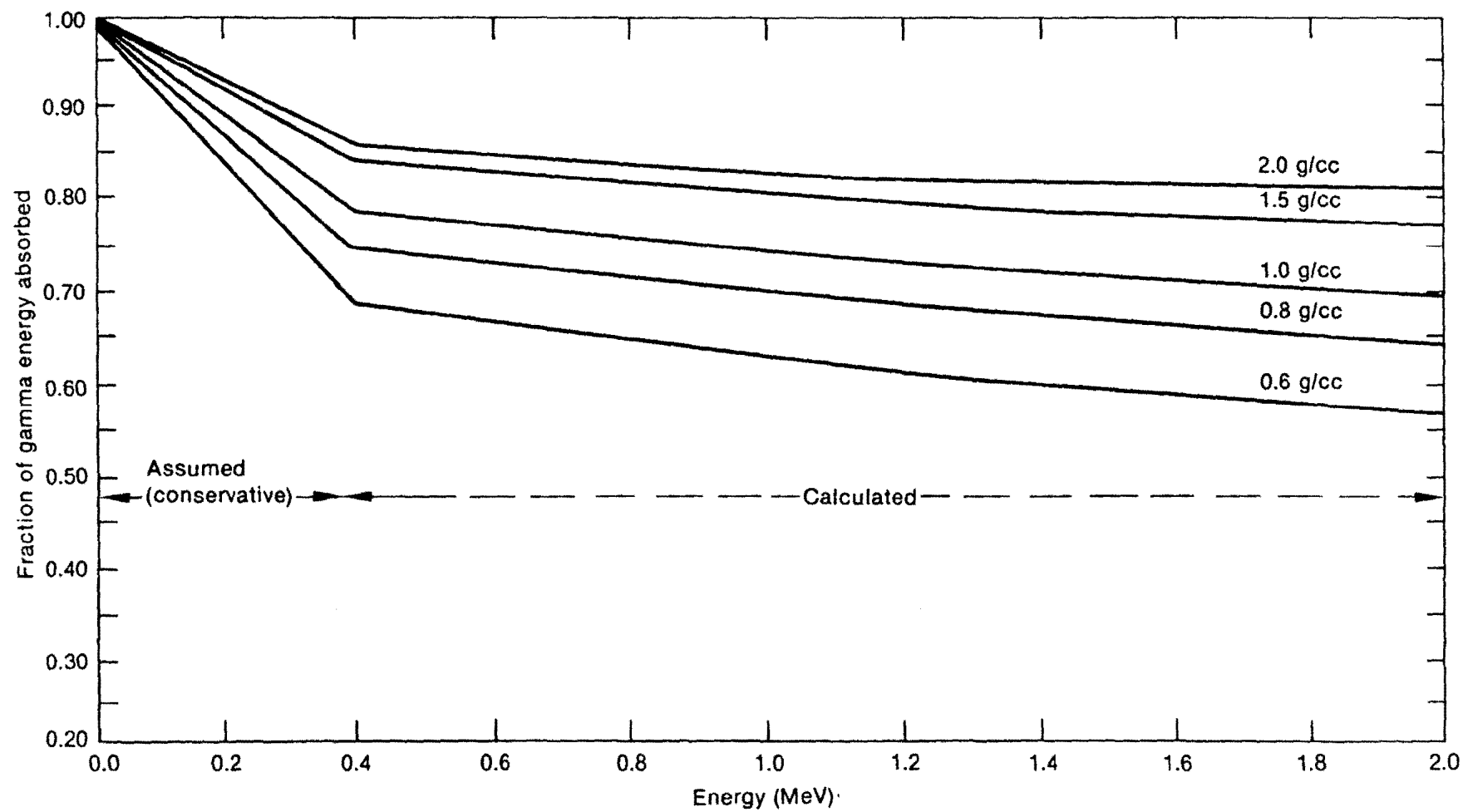


Figure A-1. Gamma absorption in a 55 gal., 7.5 ft<sup>3</sup> drum.



6 6755

Figure A-2. Gamma absorption in a 4 x 4 ft, 50 ft<sup>3</sup> liner.



6 6752

Figure A-3. Gamma absorption in a 5 x 5 ft, 98 ft<sup>3</sup> liner.

A-12

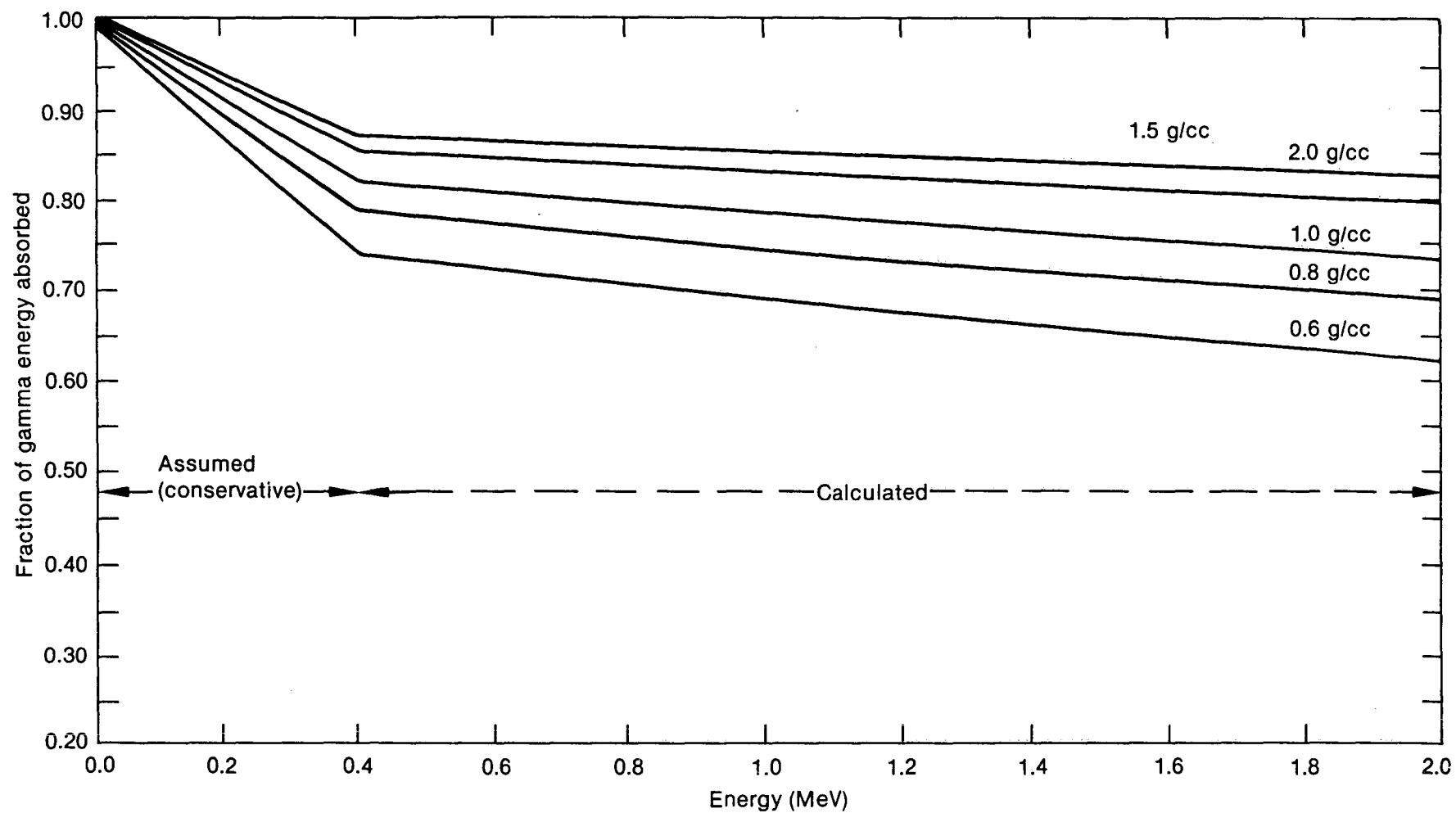


Figure A-4. Gamma absorption in a 6 x 6 ft, 170 ft<sup>3</sup> liner.

## ABSORBED DOSE CALCULATION

The absorbed dose is calculated for each nuclide (individually) using the information from Worksheets A-1 and A-2. The calculation is performed for several chosen times after the container is sealed. The dose to the waste from each nuclide, at a chosen time, is then summed to give a total absorbed dose at that time. For low-level waste, good choices for time intervals are  $t = 1, 5, 10$ , and 20 years. If the calculated total dose for a time interval exceeds the value of step 12, Worksheet A-1, the 5% hydrogen concentration level has been exceeded.

For each nuclide, the absorbed dose equals

$$\frac{AC_i}{\lambda} (\bar{E}_i + E_i^x + E_i^y) (1 - e^{-\lambda_i t})$$

In the columnar notation of Worksheet A-2

$$\frac{Aa}{\lambda_i} (g + h + f) (1 - e^{-bt})$$

where the constant A

$$= 1.86 \text{ E} + 10 \frac{\text{rad gram}}{\text{MeV yr Curie}}$$

$$\begin{aligned} \text{from } & \frac{\text{rad}}{(100 \text{ erg/g})} \times \frac{1.6 \text{ E-12 erg}}{(\text{eV})} \\ & \times \frac{\text{E} + 6 \text{ eV}}{(\text{MeV})} \times \frac{3.7 \text{ E} + 10 \text{ D/sec}}{(\text{Curie})} \\ & \times \frac{3.15 \text{ E} + 7 \text{ sec}}{(\text{yr})} \end{aligned}$$

Solving for  $t = 1$  year

$$\begin{aligned} \text{Mn-54: } D_{\text{abs}} &= (1.86 \text{ E} + 10) \\ & (5.98 \text{ E-6})(0.0052 + 0.430) \\ & (1 - \exp(-0.809))/0.809 \\ &= 3.32 \text{ E} + 4 \text{ rad} \end{aligned}$$

$$\begin{aligned} \text{Co-58: } D_{\text{abs}} &= (1.86 \text{ E} + 10)(5.88 \text{ E-6}) \\ & [0.029 + 0.0053 + \\ & (0.410 + 0.004 + 0.081)] \\ & (1 - \exp(-3.575))/3.575 \\ &= 1.57 \text{ E} + 4 \text{ rad} \end{aligned}$$

$$\begin{aligned} \text{Co-60: } D_{\text{abs}} &= (1.86 \text{ E} + 10)(3.05 \text{ E-5}) \\ & (0.0958 + 0.560 + 0.630) \end{aligned}$$

$$\begin{aligned} & (1 - \exp(-0.1315))/0.1315 \\ &= 6.84 \text{ E} + 5 \text{ rad} \\ \text{Zn-65: } D_{\text{abs}} &= (1.86 \text{ E} + 10)(2.03 \text{ E-5}) \\ & (0.0020 + 0.0077 + 0.270 \\ & + 0.0077) \\ & (1 - \exp(-1.036))/1.036 \\ &= 6.75 \text{ E} + 4 \text{ rad} \\ \text{Cs-134: } D_{\text{abs}} &= (1.86 \text{ E} + 10)(1.78 \text{ E-5})(0.157 \\ & + 0.0055 + 0.0037 + 0.070 \\ & + 0.307 + 0.382 + 0.005 \\ & + 0.010 + 0.020) \\ & (1 - \exp(-0.336))/0.336 \\ &= 2.70 \text{ E} + 5 \text{ rad} \end{aligned}$$

$$\begin{aligned} \text{Cs-137/} \\ \text{Ba-137: } D_{\text{abs}} &= (1.86 \text{ E} + 10)(4.84 \text{ E-5}) \\ & (0.1708 + 0.0657 + 0.31) \\ & (1 - \exp(-0.023))/0.023 \\ &= 4.86 \text{ E} + 5 \text{ rad} \end{aligned}$$

The total absorbed dose at 1 year after sealing is

$$\begin{aligned} D(1 \text{ yr}) \text{ Total} &= 3.32 \text{ E} + 4 + 1.57 \text{ E} + 4 \\ & + 6.84 \text{ E} + 5 + 6.75 \text{ E} + 5 + 2.70 \text{ E} + 5 \\ & + 4.86 \text{ E} + 5 = 1.55 \text{ E} + 6 \text{ rad} \end{aligned}$$

Solving in the same manner for  $t = 5$  years

$$\begin{aligned} \text{Mn-54:} & 5.88 \text{ E} + 4 \text{ rad} \\ \text{Co-58:} & 1.62 \text{ E} + 4 \text{ rad} \\ \text{Co-60:} & 2.67 \text{ E} + 6 \text{ rad} \\ \text{Zn-65:} & 1.04 \text{ E} + 5 \text{ rad} \\ \text{Cs-134:} & 7.70 \text{ E} + 5 \text{ rad} \\ \text{Cs-137/Ba-137:} & 2.32 \text{ E} + 6 \text{ rad} \end{aligned}$$

The total absorbed dose 5 years after sealing is

$$D_{\text{Total}}(5 \text{ yrs}) 5.94 \text{ E} + 6 \text{ rad}$$

To find the approximate time when 5% hydrogen concentration occurs, a graphical technique using Worksheet A-1 and the calculated absorbed dose is used.

## Hydrogen Concentration Graphical Solution

Hydrogen concentration is graphed versus time using the 1, 5, and 10 year doses with the values from the steps in Worksheet A-1.

NOTE: Because the five year dose exceeds the value of Step 12 on Worksheet A-1, the 10 year dose need not be calculated.

$$H_2 = D_{\text{Total}} \times \text{Step 7} \times \text{Step 8} \times 2.33 \text{ E-6} \\ \times 100 \text{ percent} / \text{Step 10}$$

$$\text{For 1 year: percent } H_2 = (1.56 \text{ E} + 6)(1.29 \text{ E} + 5)(0.3/100)(2.33 \text{ E-6}) (100 \text{ percent} / 8.4 \text{ E} + 4) \\ = 1.67\%$$

$$\text{For 5 years: percent } H_2 = (5.93 \text{ E} + 6)(1.29 \text{ E} + 5)(0.3/100)(2.33 \text{ E-6})(100 \text{ percent})/8.4 \text{ E} + 4 \\ = 6.38\%$$

These values are plotted as shown in Figure A-5. The graph predicts 3.7 years to produce 5% hydrogen. To verify, set  $t = 3.7$  years and calculate  $D_{\text{Total}}$ . The value is found to be  $4.73 \text{ E} + 6$  rads, slightly higher than the total Step 12 value of  $4.7 \text{ E} + 6$  rads.

Calculating the percent of  $H_2$

$$(4.73 \text{ E} + 6)(1.29 \text{ E} + 5)(0.3/100)(2.33 \text{ E-6}) \\ (100 \text{ percent})/8.4 \text{ E} + 4 = 5.08 \text{ percent}$$

The time to generate 5% hydrogen is approximately 4.0 years by calculation. The shipping window is then half this time, or 2 years.

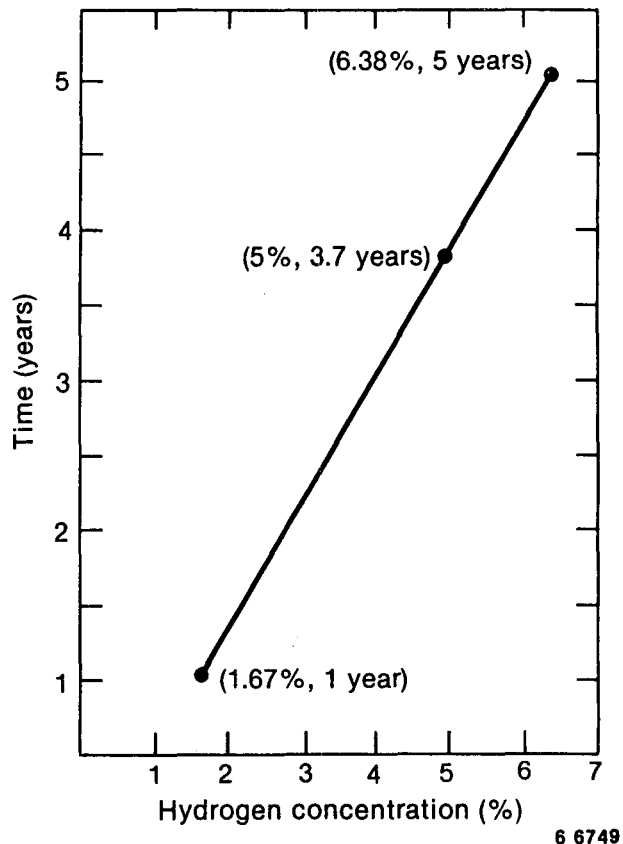


Figure A-5. Hydrogen concentration values plotted against time.

## Comment

The most desirable approach to solving this type of problem is through the use of computers. The information from Worksheet A-2 is placed in the computer memory along with the waste density (Step 5, Worksheet A-2), the container type, and constant  $A = 2.33\text{E-6}$ . The computer iterates the equation

$$\text{Absorbed dose} = \frac{Aa}{b} (g + h + f) (1 - e^{-bt})$$

for values of time ( $t$ ), until the absorbed dose of Step 12, Worksheet A-1 is reached. This yields the time to produce a 5% hydrogen concentration.

**APPENDIX B**  
**WASTE MATERIAL STUDY**



## APPENDIX B

### WASTE MATERIAL STUDY

Investigation of the characteristics of commonly encountered radioactive waste materials led to the selection of water as a generically representative substance on which calculations could be based. Calculations based on water encompass the widest spectrum of nuclear wastes generated. This choice produces gamma absorbed energy fractions that are slightly conservative (when compared to other waste materials) but more accurate than previously used techniques which have their basis in health physics considerations.

Attenuation, absorption coefficients, and buildup factors are assumed to be constant for a variety of waste forms, but are varied by the changes of waste density and gamma energy.

#### Waste Forms

The wastes to be discussed here are those radioactive forms which have the potential to generate combustible quantities of gas. Included are ion exchange resins, filter aids, evaporator concentrates, and solidified products. Waste solidification is usually done using cement, polystyrene, polyethylene, or asphalt. Most of these materials, including solidification agents, are composed of low atomic number elements; they are expected to show similar radiological characteristics in attenuation and absorption of gamma radiation. Some important properties of these materials are listed in Table B-1. National Bureau of Standards (NBS) concrete parameters are used for cement. Portland Cement's radiological behavior will be similar to that published by NBS due to its nearly identical electron density, as shown in Table B-2. Asphalt is considered as a mixture of polystyrene, polyethylene and cellulose. Filter aid is simplified as 100 percent cellulose, and resin is represented by a mixture of polystyrene and sodium sulfate or boric acid.

Gamma radiation absorption for low atomic number elements is nearly proportional to electron density. This is because for gamma rays with an energy range of 0.2, approximately 2.0 MeV lose their energy by scattering the electrons (compton effect) in the media where they travel.

#### Attenuation and Absorption Coefficients, and Buildup Factors

Attenuation and absorption coefficients, and buildup factors for the majority of radioactive wastes are largely a function of the material's electron density. An evaluation of the variance of these properties was performed for water and concrete. Inspection of Table B-1 shows that waste material electron densities fall between the values reported for water and concrete (solidifying agents are combined with materials of lower electron density), so that these substances bound the waste material evaluation. Attenuation and absorption coefficients for water and concrete are listed in Table B-3; buildup factors are listed in Table B-4 and B-5. The values for attenuation and absorption coefficients for water and concrete vary by approximately 10 percent, in the energy range of interest. The actual absorption behavior for water and concrete is similar because the combination of absorption and attenuation coefficients for the individual materials (water or concrete) tends to reduce the difference between them. Energy absorption buildup factors for water and concrete also show small differences. The buildup factor variance can become significant with increasing mean free paths; however, more than 99 percent of the gamma ray's energy is absorbed within ten mean free paths, where the variance is small. Table B-6 lists buildup factor coefficients fitted for 0 to 10 mean free paths for water and concrete. Combined effects of these are illustrated by Figures B-1 and B-2 which show the absorption characteristics for concrete and water as a function of the radius of a sphere, with the source at the center. Two different source strengths, 0.4 and 1.5 MeV, were chosen to represent typical values encountered in radioactive waste. Inspection of these curves show gamma energy absorption in water to be slightly higher than absorption in concrete.

Despite the differences in absorption and attenuation coefficients, absorbed energy fractions are quite close in water and other waste materials; the water medium cases show slightly (2 to 3%) higher percents of gamma energy absorption. This overestimation is reduced when averaged over several gamma ray travel lengths.

**Table B-1. Electron and gravimetric density of waste materials**

Components	Electron Density <sup>a</sup> ( $10^{-23} \frac{\text{electrons}}{\text{gram}}$ )	Gravimetric Density (g/cm <sup>3</sup> )	Note
Sodium Sulfate	2.97	1.49 <sup>b</sup>	Concentrate
Concrete	3.01 <sup>c</sup>	2.35 <sup>c</sup>	Solidification Agent
Boric Acid	3.12	1.46	Concentrate
Cellulose	3.20	1.0	Filter Aid
Polystyrene	3.24	1.05 ~ 1.07	Skeleton of Resin
Water	3.37	1.0	—
Polystyrene	3.44	~1.0	Solidification Agent
Asphalt	3.20 ~ 3.44 <sup>d</sup>	1.05 ~ 1.07	Solidification Agent

a. Electron Densities are obtained from Table 3-11, EPRI NP-3223

b. Ten Hydrate

c. NBS Concrete, Nuclear Science and Engineering, 263, 56 (1975)

d. Expected Range

The choice of water as a representative material for use in the general calculation yields results for gamma

absorbed energy that are accurate and slightly conservative when considering materials other than water.

**Table B-2. Comparison for elementary composition of NBS Concrete and Portland Cement element**

Element	Electron Density (e/g · E-23)	Portland Cement <sup>a</sup>		NBS Concrete <sup>b</sup>	
		Composition (percent)	Electron Density (e/g · E-23)	Composition (percent)	Electron Density (e/g · E-23)
H	5.97	—	0.00	0.56	0.03
O	3.01	35.76	1.08	49.83	1.50
Na	2.87	—	0.00	1.71	0.05
Mg	2.97	1.21	0.04	0.24	0.01
Al	2.90	4.23	0.12	4.56	0.13
Si	3.00	10.75	0.32	31.58	0.95
S	3.00	—	0.00	0.12	0.00
K	2.93	—	0.00	1.92	0.06
Ca	3.00	45.25	1.36	8.26	0.25
Fe	2.80	2.80	0.08	1.22	0.03
		100.00	2.99	100.00	3.01

a. CRC Handbook of Health Physics.

b. Nuclear Science and Engineering, 263, 56 (1975).

**Table B-3. Comparison of attenuation/absorption coefficients for concrete and water**

Photon Energy (MeV)	Attenuation <sup>a</sup> /Absorption <sup>b</sup> Coefficient (cm <sup>2</sup> /g)	
	Water	Concrete
0.1	0.171/0.0256	0.179/0.424
0.15	0.151/0.0277	0.144/0.290
0.2	0.137/0.0297	0.127/0.290
0.3	0.119/0.0319	0.108/0.295
0.4	0.106/0.0328	0.0963/0.298
0.5	0.0968/0.0330	0.0877/0.300
0.6	0.0896/0.0329	0.0810/0.0297
0.8	0.0786/0.0321	0.0709/0.0289
1.0	0.0707/0.0309	0.0637/0.0279
1.5	0.0575/0.0282	0.0519/0.0254
2.0	0.0494/0.0260	0.0448/0.0235

a. Health Physics Handbook

b. CRC Handbook of Radiation Measurement and Protection, Section A, CRC Press Inc., 1978.

**Table B-4. Buildup factors for water<sup>a</sup>**

Mean Free Path	Energy (MeV)										
	2.000	1.500	1.000	0.800	0.600	0.500	0.400	0.300	0.200	0.150	0.100
0.0	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
0.5	1.38	1.42	1.47	1.51	1.56	1.61	1.66	1.75	1.92 <sup>a</sup>	2.07 <sup>a</sup>	2.36
1.0	1.83	1.93	2.08	2.18	2.34	2.45	2.60	2.84	3.42 <sup>a</sup>	3.91	4.52
2.0	2.82	3.11	3.62	3.96	4.48	4.87	5.42	6.25	8.22	9.36	11.7
3.0	3.87	4.44	5.50	6.24	7.40	8.29	9.56	11.5	15.7	18.6	23.5
4.0	4.99	5.90	7.66	8.96	11.1	12.7	15.1	19.0	26.4	32.5	40.6
5.0	6.16	7.47	10.1	12.1	15.4	18.1	22.2	28.8	41.3	52.0	64.0
6.0	7.38	9.14	12.8	15.6	20.6	24.6	30.8	41.2	61.0	77.9	94.8
7.0	8.66	10.9	15.7	19.6	26.4	32.2	41.1	56.5	86.2	111	134
8.0	9.97	12.8	18.9	24.0	33.0	40.8	53.2	75.0	118	153	183
10.0	12.7	16.8	26.0	33.9	48.7	61.8	83.2	122	202	268	314
15.0	20.1	27.9	47.4	65.6	102	137	197	318	582	805	917
20.0	28.0	40.4	73.5	106	176	247	377	656	1 310	1 890	2 120
25.0	36.4	54.1	104	156	272	395	632	1180	2 580	3 840	4 260
30.0	45.2	68.8	138	213	388	582	972	1930	4 640	7 050	7 780
35.0	54.3	84.4	175	277	525	809	1400	2950	7 890	12 100	13 100
40.0	63.6	101	214	349	683	1080	1940	4280	12 800	19 600	20 300

a. A. B. Chilton et. al., Nuclear Science and Engineering, 97, 73, (1980).

Table B-5. Buildup factors for concrete<sup>a</sup>

Mean Free Path	Energy (MeV)										
	2.000	1.500	1.000	0.800	0.600	0.500	0.400	0.300	0.200	0.150	0.100
0.0	1.00+00	1.00+00	1.00+00	1.00+00	1.00+00	1.00+00	1.00+00	1.00+00	1.00+00	1.00+00	1.00+00
0.5	1.39+00	1.42+00	1.49+00	1.53+00	1.60+00	1.66+00	1.73+00	1.86+00	2.11+00	2.32+00	2.39+00
1.0	1.83+00	1.93+00	2.11+00	2.22+00	2.41+00	2.55+00	2.74+00	3.06+00	3.65+00	4.04+00	3.89+00
2.0	2.80+00	3.08+00	3.59+00	3.94+00	4.48+00	4.89+00	5.46+00	6.32+00	7.69+00	8.29+00	7.06+00
3.0	3.82+00	4.36+00	5.35+00	6.03+00	7.10+00	7.89+00	9.00+00	1.06+00	1.29+01	1.35+01	1.04+01
4.0	4.92+00	5.77+00	7.35+00	8.48+00	1.03+01	1.16+01	1.34+01	1.60+01	1.93+01	1.96+01	1.41+01
5.0	6.07+00	7.28+00	9.61+00	1.13+01	1.40+01	1.60+01	1.87+01	2.25+01	2.70+01	2.67+01	1.79+01
6.0	7.28+00	8.90+00	1.21+01	1.45+01	1.82+01	2.11+01	2.50+01	3.03+01	3.60+01	3.48+01	2.21+01
7.0	8.55+00	1.06+01	1.48+01	1.80+01	2.31+01	2.70+01	3.23+01	3.95+01	4.66+01	4.40+01	2.66+01
8.0	9.86+00	1.24+01	1.78+01	2.19+01	2.86+01	3.37+01	4.07+01	5.01+01	5.88+01	5.45+01	3.15+01
10.0	1.26+01	1.63+01	2.43+01	3.07+01	4.13+01	4.96+01	6.06+01	7.62+01	8.85+01	7.91+01	4.21+01
15.0	2.00+01	2.71+01	4.40+01	5.85+01	8.40+01	1.05+02	1.34+02	1.74+02	1.99+02	1.65+02	7.47+01
20.0	2.81+01	3.93+01	6.79+01	9.41+01	1.42+02	1.83+02	2.43+02	3.26+02	3.72+02	2.92+02	1.16+02
25.0	3.67+01	5.26+01	9.55+01	1.37+02	2.16+02	2.86+02	3.92+02	5.43+02	6.20+02	4.63+02	1.65+02
30.0	4.57+01	6.70+01	1.26+02	1.86+02	3.05+02	4.14+02	5.83+02	8.34+02	9.58+02	6.86+02	2.22+02
35.0	5.50+01	8.21+01	1.60+02	2.42+02	4.08+02	5.67+02	8.20+02	1.21+03	1.40+03	9.63+02	2.86+02
40.0	6.46+01	9.80+01	1.97+02	3.03+02	5.27+02	7.47+02	1.10+03	1.67+03	1.94+03	1.30+03	3.58+02

a. C. M. Eisenhauer, G. L. Simmons, Nuclear Science and Engineering, 263, 56, (1975).

**Table B-6. Buildup factor coefficient<sup>a,b,c</sup>**

Water Media Coefficient				
Energy (MeV)	2.0000 E-0	1.5000 E+0	1.0000 E+0	8.0000 E-1
B0	1.0000 E+0	1.0000 E+0	1.0000 E+0	1.0000 E+0
B1	7.5284 E-1	8.1045 E-1	8.6259 E-1	8.9863 E-1
B2	7.3047 E-2	1.1920 E-1	2.2233 E-1	2.9110 E-1
B3	-3.2540 E-3	-4.3743 E-3	-6.0645 E-3	-5.3580 E-3
Energy (MeV)	6.0000 E-1	4.0000 E-1	2.0000 E-1	1.0000 E-1
B0	1.0000 E+0	1.0000 E+0	1.0000 E+0	1.0000 E+0
B1	9.4167 E-1	1.0282 E+0	1.5335 E+0	2.0926 E+0
B2	3.9744 E-1	5.3743 E-1	7.6967 E-1	1.2865 E+0
B3	1.6241 E-3	1.5738 E-2	1.0831 E-1	1.6278 E-1
Concrete Media Coefficient				
Energy (MeV)	2.0000 E+0	1.5000 E+0	1.0000 E+0	8.0000 E-1
B0	1.0000 E+0	1.0000 E+0	1.0000 E+0	1.0000 E+0
B1	7.6801 E-1	8.1785 E-1	9.2308 E-1	9.7877 E-1
B2	6.1447 E-2	1.0697 E-1	1.8204 E-1	2.3777 E-1
B3	-2.3030 E-3	-3.7178 E-3	-4.2724 E-3	-4.0301 E-3
Energy (MeV)	6.0000 E-1	4.0000 E-1	2.0000 E-1	1.0000 E-1
B0	1.0000 E+0	1.0000 E+0	1.0000 E+0	1.0000 E+0
B1	1.0880 E+0	1.2960 E+0	2.0200 E+0	2.7531 E+0
B2	3.1374 E-1	4.3558 E-1	6.0804 E-1	1.2137 E-1
B3	-2.1601 E-3	-3.1103 E-3	5.9321 E-3	1.3481 E-3

a. Fitted Over 0 to 10 mean free paths.

b. Buildup Factor =  $B_0 + B_1X + B_2X^2 + B_3X^3$  X: mean free path length.

c. All constants are calculated using the least square method described in EPRI NP-3223 and fitted to the buildup factors listed in Table B-4.

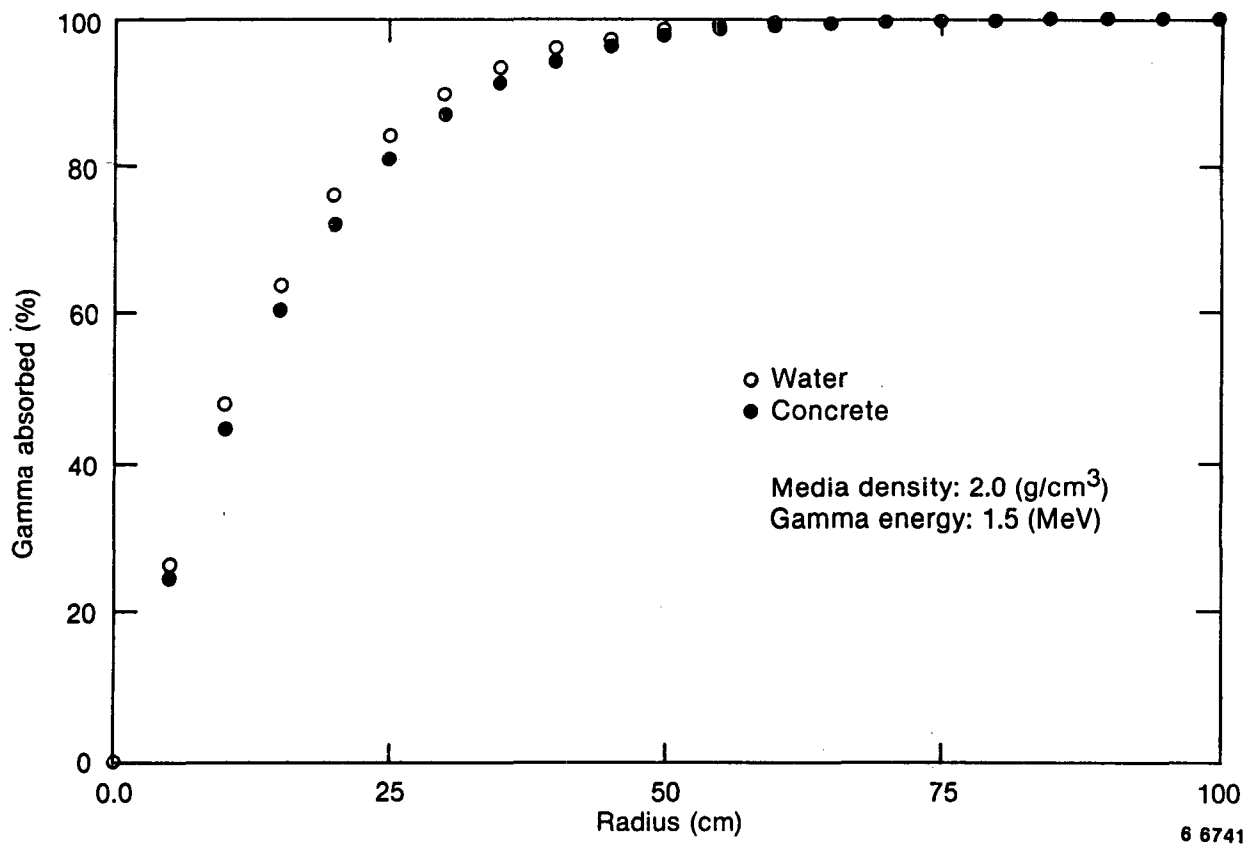


Figure B-1. Gamma absorption in spheric media.



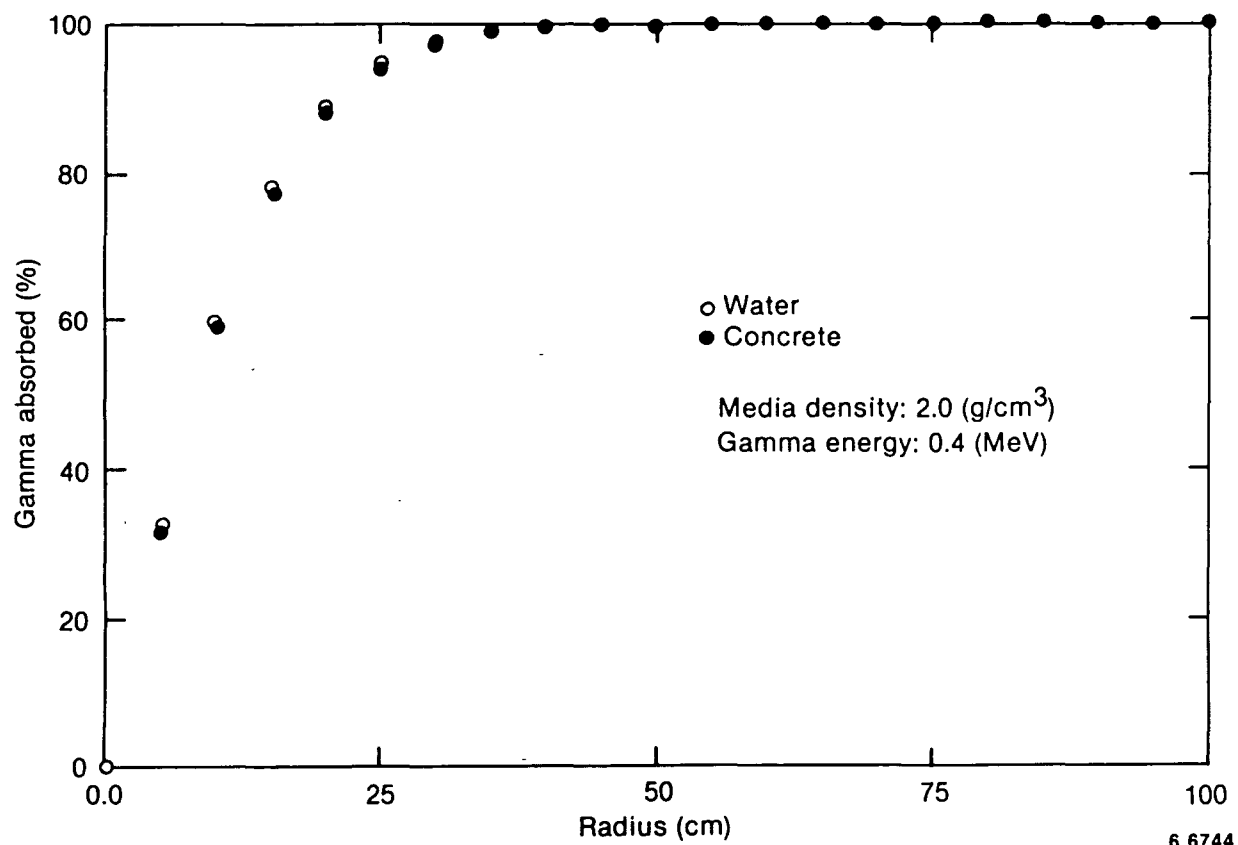


Figure B-2. Gamma absorption in spheric media.

**APPENDIX C**  
**COMPUTER APPLICATION**

## APPENDIX C

### COMPUTER APPLICATION

The gamma absorbed dose was calculated for standard waste container geometries using a point-kernal computer code. The code, QAD-FN, a version of the QAD code developed by R. E. Malenfant, is in current use at the Idaho National Engineering Laboratory.<sup>C-1</sup> Modifications were made to increase the number of source and detection points so that larger container geometries could be calculated. The attenuation, absorption, and buildup factors of the generic waste form discussed in Appendix B were used to define the gamma absorption characteristics of standard waste forms normally encountered in the nuclear power industry.

#### Basic Equations

Radiation absorption is based on an equation for point-to-point relations of radiation attenuation and absorption. For a discrete gamma ray, the energy flux,  $[\phi(r)]$ , at a distance (r) can be expressed as

$$\phi(r) = \frac{I_0 \exp(-\mu_e r) B(\mu_e r)}{4\pi r^2} dV_S \quad (C-1)$$

where

$\phi(r)$  = energy flux at distance r, (MeV/sec · cm<sup>3</sup>)

$I_0$  = source intensity, (MeV/sec · cm<sup>3</sup>)

$\mu_e$  = liner attenuation coefficient, (1/cm)

r = distance from a source, (cm)

$dV_S$  = source volume, (cm<sup>3</sup>)

The energy absorbed at target with small volume,  $dV_R$ , is

$$E_{abs}(r) = \mu_a \phi(r) dV_R \quad (C-2)$$

where

$E_{abs}(r)$  = energy absorbed at distance r, (MeV/s)

$\mu_a$  = liner energy absorption coefficient, (1/cm)

$dV_R$  = Target volume, (cm<sup>3</sup>)

Combining Equations (C-1) and (C-2),

$$E_{abs}(r) = \mu_a \frac{I_0 \exp(-\mu_e r) B(\mu_e r)}{4\pi r^2} dV_S dV_R \quad (C-3)$$

The total amount of energy absorbed in the media can be calculated by integrating Equation (C-3) over the source and irradiated media as

$$E_{abs}^{Total} = \mu_a I_0 \int_{V_S} \int_{V_R} \frac{\exp(-\mu_e r) B(\mu_e r)}{4\pi r^2} dV_S dV_R \quad (C-4)$$

where

$E_{abs}^{Total}$  is the total absorbed energy, (MeV/s), in an irradiated media.

For cylindrical geometry with homogeneous source distribution, Equation (C-4) is

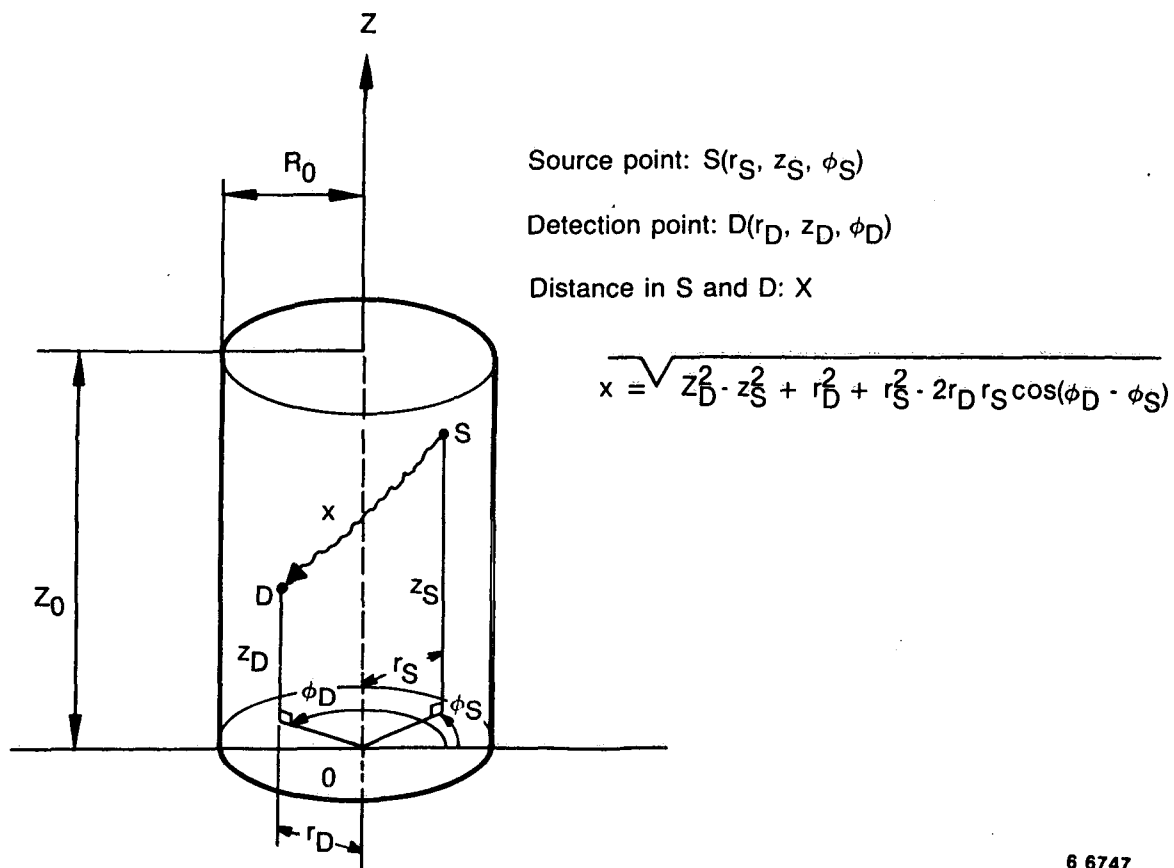
$$E_{abs}^{Total} = \mu_a I_0 \int_{\phi_D} \int_{z_D} \int_{r_D} \int_{\phi_S} \int_{z_S} \int_{r_S} \frac{\exp(-\mu_e X) B(\mu_e X)}{4\pi X^2} r_D r_S d\phi_S dz_S dr_S d\phi_D dz_D dr_D \quad (C-5)$$

and

$$X \equiv \sqrt{z_D^2 - z_S^2 + r_D^2 + r_S^2 - 2r_D r_S \cos(\phi_S - \phi_D)} \quad (C-6)$$

where  $\phi$ ,  $z$ ,  $r$  are the coordinates of a cylinder, and subscripts D and S are detection and source points, respectively. Distance, X, is defined in Equation (C-6). A schematic of this coordinate system is shown in Figure C-1. Then, Equation (C-5) can be rewritten with integration ranges as

$$E_{abs}^{Total} = 4\pi \mu_a I_0 \int_{z_1}^Z \int_0^R \left[ 2 \int_0^x \int_0^Z \int_0^R \frac{\exp(-\mu_e X) B(\mu_e X)}{4\pi X^2} r_D r_S d\phi_S dz_S dr_S \right] dz_D dr_D \quad (C-7)$$



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Figure C-1. Source and detection points in relation to the cylindrical coordinate system.

In actual computation, the integrals are substituted by the sum of point-to-point radiation absorption

$$E_{\text{abs}}^{\text{Total}} = 2\mu_a I_0 \sum_{i=1}^n \left[ 2 \sum_{j=1}^m \frac{\exp(-\mu_e X_{ij}) B(\mu_e X_{ij})}{4\pi X_{ij}^2} \Delta V_{S,j} \right] \Delta V_{D,i} \quad (\text{C-8})$$

with

$$\Delta V_{S,j} = r_S \Delta r_S \Delta \phi_S \Delta z_S$$

$$\Delta V_{D,i} = 2\pi r_D \Delta r_D \Delta z_D \quad (\text{C-9})$$

where,  $V_{S,j}$  and  $V_{D,i}$  are the  $j^{\text{th}}$  source and the  $i^{\text{th}}$  detector sub-volumes, with the source point assumed at the center of each source sub-volume. The sign  $\Delta$ , with  $\phi$ ,  $z$ , and  $r$ , are subdivisions of

each direction, and  $m$  and  $n$  are the number of sources and detectors, respectively.

Determination of source and detector points networks are discussed in the following sections.

**Source Point Networks.** Source point networks were examined by the following steps. Standard arrangements were selected for the generic calculations.

- Comparison with the analytical solution was made to verify the numerical method described in the previous section, using the analytical solution for a sphere.
- The source point networks for cylindrical configurations were developed and various arrangements examined to verify the standard setting of source point networks.

Model comparisons between analytically obtained energy absorption (see Appendix D) and numerical solutions have been done for waste densities of,  $0.6 \sim 2.0 \text{ g/cm}^3$ , and energy levels,

0.4 and 1.5 MeV. The radiological constants used are defined in Appendix B. The numerical method is principally the same as the  $j^{\text{th}}$  sum of Equation (C-8). Because of spherical symmetry, subdivisions are considered in the container radius direction with the source at the center of each subdivision. The modeled configuration and equations used are shown in Figure C-2.

Tables C-1 and C-2 show the comparisons of two methods (analytical and numerical). Table C-1 is the contribution from 0 to 10 cm in radius; Table C-2 is the contribution from 10 to 20 cm. The deviations of numerical results from analytically obtained values are decreased by increasing the number of subdivisions. Deviations are less than 1 percent for subdivisions of widths smaller than 5 cm in all cases. These results suggest that the source subdivisions surrounding detection points should be arranged such that the width is less than 5 cm.

Based on these sample calculations, absorbed energy was estimated for dewatered resin packed in 55 gal drums to determine a source point network for a cylindrical configuration. A standard source subdivision arrangement is illustrated with the relation between detection plane and source volume shown in Figure C-3. The source is assumed to be homogeneously distributed over the waste volume, and is divided into sub-volumes along three axis, Z (Height), R (Radius), and  $\phi$  (Radial), according to the mesh generating capability of the QAD-FN code. The source region, where the detectors are included, was divided into 1-cm width subdivisions in Z and R directions, and  $\pi/30$  in the radial direction. The largest width of a source subdivision is approximately 2.7 cm at the edge of a cylinder in the radial direction. To calculate the total absorbed energy, 48 detection points are placed on the detection plane in the symmetrical center plane with half-height of the waste media, according to the detection point networks shown in Figure C-4. The details of the detection point arrangement will be discussed in the next section.

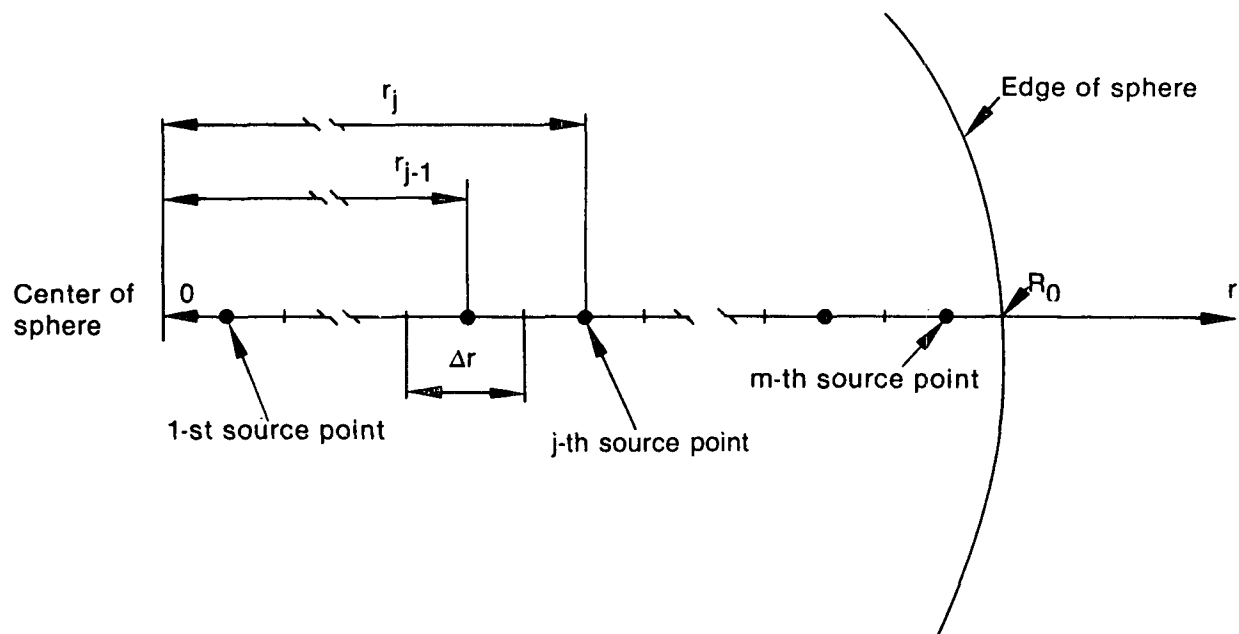
The major valuables are summarized in Table C-3. The result is shown in Table C-4 with Case No. 2 (standard). Results with different source point arrangements are listed to illustrate the effects of source point network changes. The number of source subdivisions were decreased in Case 1 for every direction; the number was increased in Cases 3 and 4 for Z direction, in Case 5 for R direction, and Cases 6 and 7 for  $\phi$  direction.

These results show that the deviations of absorbed energies with various source point arrangements are usually small. Comparison of Cases 1 and 2, indicates that the source subdivisions should not be decreased from the standard. Although the relatively high deviation in Case 6 (subdivision increased in  $\phi$  direction) is observed, further increments in this direction (Case 7) causes only a 0.17% increase from Case 6. Changes in absorbed energies are almost in equilibrium with the standard case, with deviations in the order of 1%. The standard setting of the source point arrangement discussed is used for the generic calculation, with basic source point arrangements maintained for the larger containers.

**Detection Point Networks.** A detection point is designed to represent a certain sub-volume of waste media (i.e., it is assumed that a detection point will show the average absorbed energy in the sub-volume where it is located). The internal dose profiles for dewatered resin in a 55 gal drum are shown in Figure C-5, with the relation between detection points and representative ranges illustrated. The dose profiles of the center ( $z = 42$  cm) and edge ( $z = 83$  cm) are plotted against the radius (center to edge). The detection point at 7 cm on Figure C-5 is representative of the absorbed dose from 3 to 11 cm. The error associated with the choice of the representative detection point can be evaluated by calculating the upper and lower average absorbed dose as shown in Figure C-6. The upper average is defined by the midpoint dose of a range in radius or height ( $X_{i-1}$  to  $X_{i+1}$ ) as an average value. The lower average is the average of all applicable data points in the range of interest. When dose profiles are monotonously decreasing with a convex shape as shown in Figure C-5, the upper average will always exceed the true dose profiles but the lower average will not.

Four different lines of dose profiles are examined for two energy levels (0.4 MeV and 1.5 MeV) to estimate the possible error associated with this method.

Tables C-5 (height) and C-6 (radius) show the estimated error ranges of sample calculations. The error ranges are expressed by percent deviations. As shown in the tables, the percent deviations are small, so that the overall error is expected to be well below 1%. The detection point networks discussed are applied to generic calculations. The same approach has been used to calculate energy absorption for larger containers.



#### Numerical solution for sphere

$$E_{\text{abs}}^{\text{center}} = \mu_a I_0 \sum_{j=1}^m \frac{\exp(-\mu_e r_j) B(\mu_e r_j)}{4\pi r_j^2} \Delta V_{S,j}, \quad (\text{MeV/cm}^3/\text{s})$$

$$\Delta V_{S,j} \equiv 4\pi r_j \Delta r, \quad \Delta r \equiv R_0/m$$

#### Analytical solution for sphere (with build-up factor)

$$E_{\text{abs}}^{\text{center}} = \mu_a I_0 \left\{ \sum_{k=0}^3 \frac{1}{\mu_e^{k+1}} \left[ B^{(k)}(0) - \exp(-\mu_e R_0) B^{(k)}(\mu_e R_0) \right] \right\}, (\text{MeV/cm}^3/\text{s})$$

$B^{(k)}(\mu_e r)$ : k-th derivative of  $B(\mu_e r)$

$$B(\mu_e r) \equiv \sum_{i=0}^3 B_i(\mu_e r)^i$$

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Figure C-2. Source points arrangement and energy absorption equations for spherical medium.

**Table C-1. Comparison of analytical and numerical method—radius of sphere = 0 ~ 10 cm**

Number of Subdivisions (m)	Width of Subdivisions (cm)	Percent Deviations from Analytical Solutions					
		Gamma Energy 0.4 (MeV)			Gamma Energy 1.5 (MeV)		
		$\rho = 0.6^a$	$\rho = 1.0$	$\rho = 2.0$	$\rho = 0.6$	$\rho = 1.0$	$\rho = 2.0$
1	10	0.28	1.02	3.81	0.14	0.32	0.63
2	5	0.066	0.22	0.81	0.037	0.08	0.18
4	2.5	0.015	0.05	0.19	0.0061	0.019	0.046
5	2	0.010	0.034	0.12	0	0.011	0.032
8	1.25	0.005	0.012	0.045	0	0.004	0.013
10	1.0	0.005	0.009	0.028	0	0.004	0.008
15	0.67	0	0.003	0.012	0	0	0.004
20	0.5	0	0.003	0.007	0	0	0.002
Analytical Solution	$E_{\text{abs}}^{\text{Center}^b}/I_0$	0.198	0.326	0.597	0.163	0.262	0.478

a. Density g/cm<sup>3</sup>.

b. Defined in Figure 2.

**Container Geometry.** The energy absorption characteristics of several standard waste containers have been investigated. To define the standard configurations for generic calculation, the relations between internal radius and effective height are plotted in Figure C-7, the data shown as squares were obtained from an EPRI waste study<sup>C-2</sup>.

All modeled containers except 55 gal drums are assumed to be right circular cylinders. Naturally, actual containers show some deviations from right circular cylinders so the magnitude of deviations are plotted against the internal radius in Figure C-8. The deviations from right circular cylinders in larger containers can vary by 50%. A discussion of these variances follows in a later section.

**Radiation Absorption in Generic Waste.** Calculational conditions for gamma radiation absorption in waste assumed as generic are summarized in Table C-7. The source energy range was determined

by evaluating waste shipment information and 10 CFR 61 requirements. Media characteristics are presented in Appendix B.

The numbers of source and detection points taken are summarized below; detail arrangements for a 55 gallon drum are shown in Figures C-9 and C-10.

Radius/Height (cm)	Source/Detection Points	Note
30/90	67,500/48	55 gal drum
60/120	270,000/81	4 x 4 liner
70/140	350,000/100	5 x 5 liner
90/180	810,000/121	6 x 6 liner

The results are listed in Table C-8, and are expressed by absorption percentage with three energy levels (0.4 to 2.0 MeV), five different densities (0.6 to

**Table C-2. Comparison of analytical and numerical method—radius of sphere = 10 ~ 20 cm**

Number of Subdivisions (m)	Width of Subdivisions (cm)	Percent Deviations from Analytical Solutions					
		Gamma Energy 0.4 (MeV)			Gamma Energy 1.5 (MeV)		
		$\rho = 0.6^a$	$\rho = 1.0$	$\rho = 2.0$	$\rho = 0.6$	$\rho = 1.0$	$\rho = 2.0$
1	10	0.38	0.55	3.80	0.068	0.014	1.10
2	5	0.090	0.14	0.64	0.021	0.004	0.30
4	2.5	0.021	0.033	0.15	0.007	0.004	0.10
5	2	0.011	0.022	0.090	0.007	0.004	0.043
8	1.25	0.005	0.007	0.037	0.007	0.004	0.019
10	1.0	0	0.004	0.024	0.007	0.004	0.012
15	0.67	0	0	0.010	0.007	0.004	0.007
20	0.5	0	0	0.007	0	0.004	0.005
Analytical Solution	$E_{\text{abs}}^{\text{Center}^b} / I_0$	0.189	0.274	0.292	0.146	0.212	0.280

a. Density g/cm<sup>3</sup>.

b. Defined in Figure 2.

2.0 g/cm<sup>3</sup>), and four kinds of containers. The absorption percentage is defined as the absorbed percentage of total emitted energy and is expressed by combining

Source intensity  $I_0$ , (MeV/s · cm<sup>3</sup>), total absorbed energy,  $E_{\text{abs}}^{\text{Total}}$  (MeV/s), and waste volume  $V_s$  (cm<sup>3</sup>), as

$$\text{Absorption Percentage} = \frac{E_{\text{abs}}^{\text{Total}}}{I_0 V_s} \times 100$$

The actual absorbed energy can be calculated by multiplying  $I_0 \times V_s$  by the Absorption Percentage. For example, the absorbed energy for 1.0 MeV gamma ray of 1 $\mu$ Ci/cm<sup>3</sup> in a 4 x 4 liner with 1.0 g/cm<sup>3</sup> density is calculated as

$$E_{\text{abs}}^{\text{Total}} = \frac{70.3911}{100 \text{ percent}} \times 1.0 [\text{MeV/d}] \times 3.7 \times 10^4 \text{ d/s/cm}^3 \times V_s$$

where

$$V_s = \pi r^2 h = \pi \times 60^2 \times 120 [\text{cm}^3]$$

and

$$1\mu\text{Ci/cm}^3 = 3.7 \times 10^4 \frac{\text{decays}}{\text{sec cm}^3}$$

Then

$$E_{\text{abs}}^{\text{Total}} = 3.53 \times 10^{10} (\text{MeV/s})$$

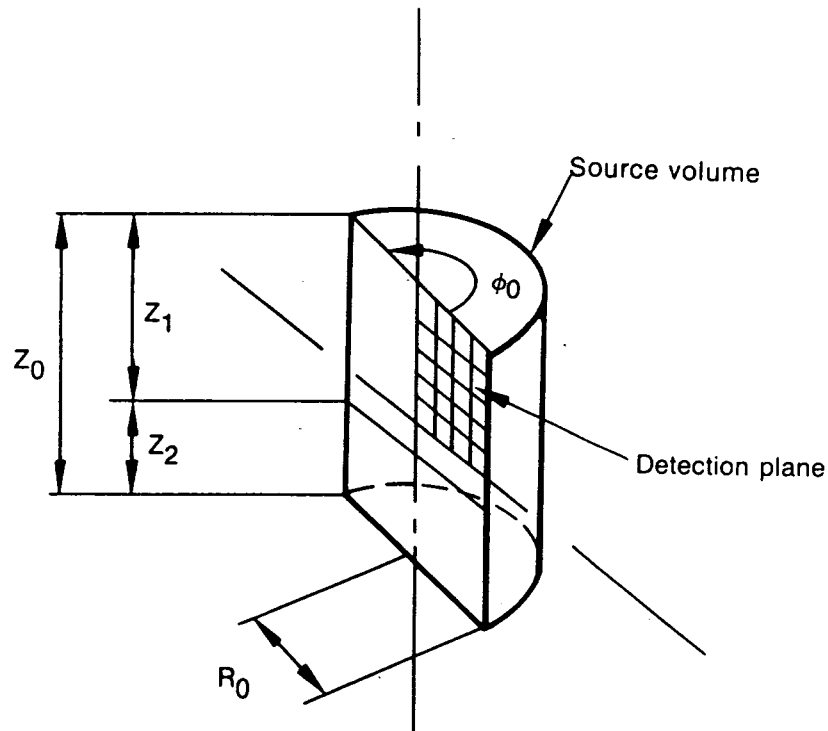
or

$$E_{\text{abs}}^{\text{Total}} = 5.66 \times 10^4 (\text{erg/s})$$

The average dose rate is obtained by using the conversion factor for rad, gram and erg, then

$$\text{Average Dose Rate} = 5.66 \times 10^4 (\text{erg/s}) \times 3600 (\text{s/h}) \times$$





$Z_0$ : Height 84 cm, 70 subdivisions

$Z_1$ : Height 56 cm (including detection plane),  $\Delta z = 1$  cm

$Z_2$ : Height 28 cm (out of detection plane,  $\Delta z = 2$  cm

$R_0$ : Radius 27 cm,  $\Delta r = 1$  cm, 27 subdivisions

$\phi_0$ : Radial  $\pi$ ,  $\Delta\phi = \pi/30$ , 30 subdivisions

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Figure C-3. Source subdivisions for U.S. Department of Transportation 55 gal. Drum.

$$\times \frac{1}{100} \text{ (rad g/erg)}/1.36 \times 10^4 \text{ (g)}$$

$$= 1.50 \text{ (rad/h)}$$

where

$f(xy)$  = absorbed portion of gamma radiation energy

$x$  = density of waste media,  $\text{g/cm}^3$

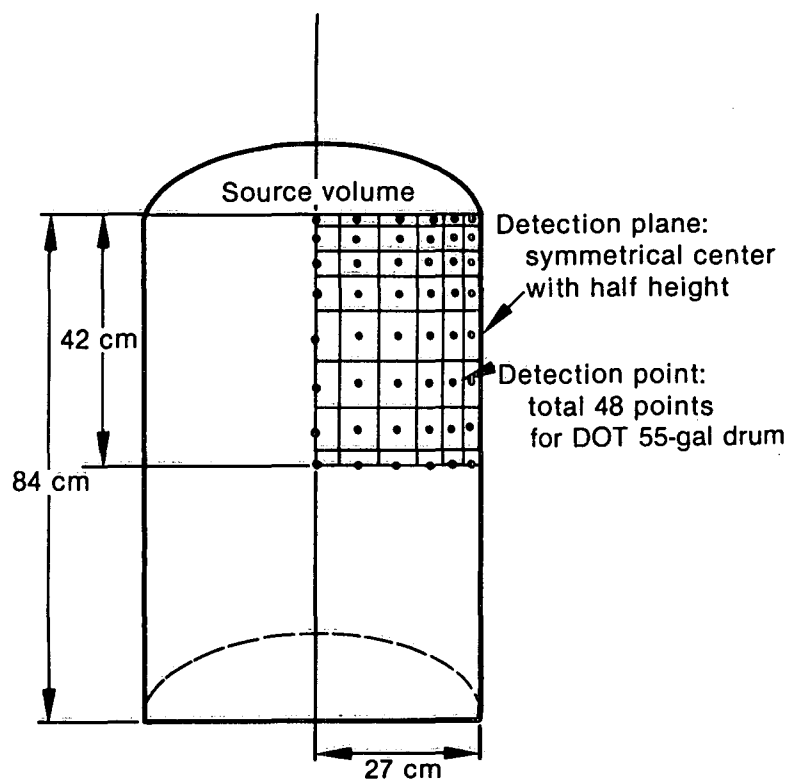
$y$  = gamma radiation energy, MeV

To apply these results to actual nuclide and waste media, data has been interpolated by the least square fitting function as

$$\begin{aligned} f(xy) = & (a_1 xy) + (a_2 x\sqrt{y}) + (a_3 \sqrt{xy}) \\ & + (a_4 x) + (a_5 \sqrt{xy}) + (a_6 y) \\ & + (a_7 \sqrt{x}) + (a_8 \sqrt{y}) + (a_9) \end{aligned}$$

Constants  $a_1$  to  $a_9$  for different containers are summarized in Table C-9. By using this function, the absorbed portion of gamma radiation energy can be calculated for various waste media and nuclides.

As an example, the absorbed energy for Co-60 is estimated below,



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Figure C-4. Detection points network for U.S. Department of Transportation 55 gal. drum.

**Table C-3. Setting of valuables for sample calculation**

Valuables	Characteristics	Note
Waste material	Dewatered resin	1:1 mixture of cation and anion
Density	0.78 g/cm <sup>3</sup>	Catalog value
Container	DOT 55 gal drum dia. 54 cm height 90 cm	
Gamma ray energy	0.4 MeV and 1.5 MeV	
Attenuation coefficient	Water <sup>a</sup>	
Absorption coefficient	Water <sup>a</sup>	
Build-up factor	Dose buildup factor <sup>b</sup>	

a. The coefficients used here are shown in Appendix B.

b. The same equation used in EPRI study was taken only in this calculation.

**Table C-4. Absorbed energy with various source point arrangements (dewatered resin pack in 55 gallon drum)**

Case Number	Numbers of Subdivisions			Average Dose (rad/h)	Deviations from Standard Case (%)
	Z	R	Y		
1	56	18	20	9.3834	-1.9
2 <sup>a</sup>	70	27	30	9.5073	—
3	84	27	30	9.5070	-0.003
4	112	27	30	9.5112	+0.04
5	70	54	30	9.5054	-0.02
6	70	27	60	9.6204	+1.19
7	70	27	90	9.6370	+1.36

a. Standard case

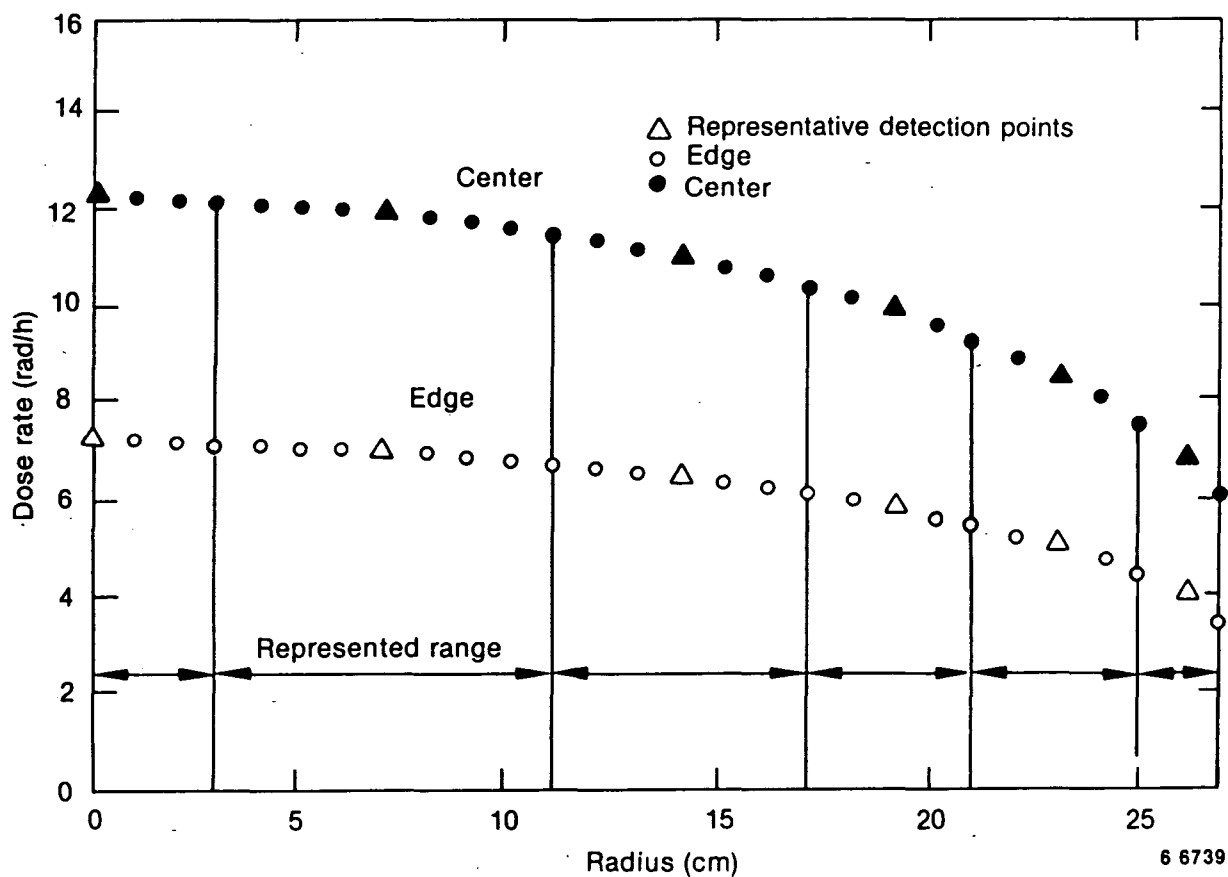
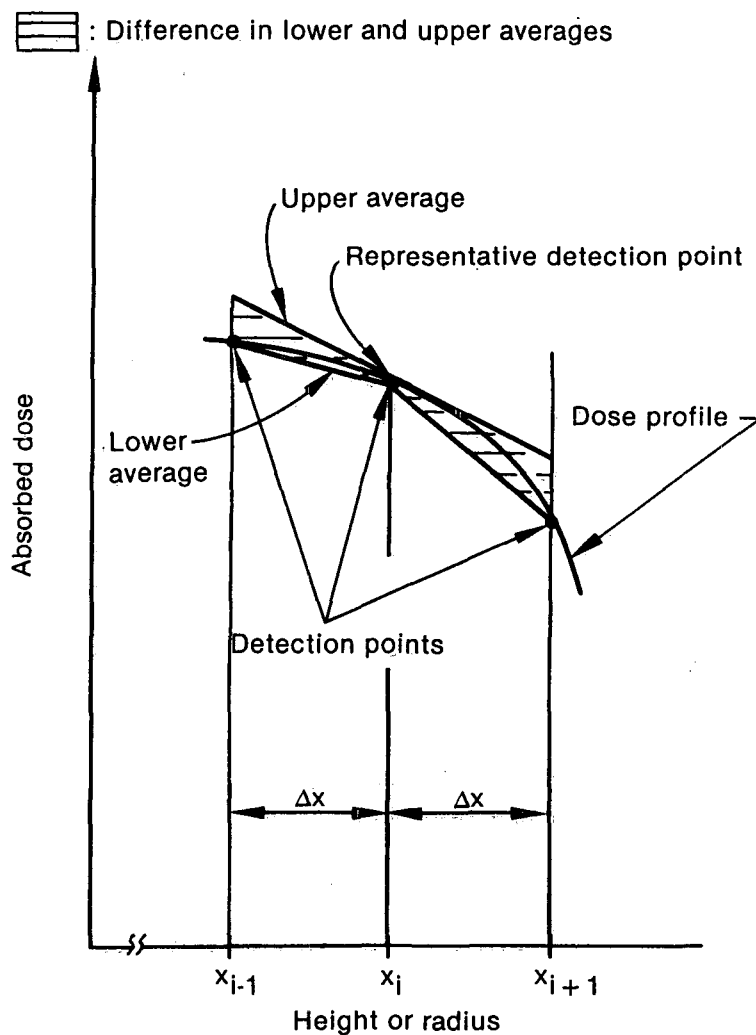


Figure C-5. Internal dose profile of resin, using 1.5 MeV gamma ray.



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Figure C-6. Error estimation for representative detection points.

- Media: Dewatered Resin (0.865 g/cm<sup>3</sup>)
- Energy: 1.17 MeV - 100 percent abundance  
1.33 MeV - 100 percent abundance
- Container: 55 gal drum.

Taking the fitting constants for a 55 gal drum, X and Y are set as 0.865 and 1.17 or 1.33. Then,

$$f(x = 0.865, y = 1.17) = 0.493$$

$$f(x = 0.865, y = 1.33) = 0.482$$

Considering the abundance of gamma ray, then the  $f(xy)$  for <sup>60</sup>Co is,

$$f(\text{Co-60}) = \frac{1.17 \times 0.493 \times 1.0 + 1.33 \times 0.482 \times 1.0}{1.17 \times 1.0 + 1.33 \times 1.0} = 0.488$$

In this case, 48.8 percent of <sup>60</sup>Co gamma energy is absorbed in waste media.

**Consideration for Extreme Cases.** As described in previous sections, generic waste forms are assumed to have

- Homogeneous media and source distributions
- Right circular cylindrical configuration for larger size containers.

**Table C-5. Estimate error of representing detection points (in height)**

Detector Coordinates (cm)			1.5 MeV		0.4 MeV	
Number	R	Z	Dose (rad/h)	Deviation (%)	Dose (rad/h)	Deviation (%)
1 <sup>a</sup>	0.00	83.00	7.736 E + 0	0.4285	2.3942 E + 0	0.9559
2	0.00	80.00	8.7621 E + 0	0.4915	2.9034 E + 0	0.4830
3	0.00	76.00	9.9476 E + 0	0.1926	3.3149 E + 0	0.1956
4	0.00	71.00	1.0845 E + 1	0.1950	3.6334 E + 0	0.2072
5	0.00	66.00	1.1417 E + 1	0.0595	3.8301 E + 0	0.0644
6	0.00	58.00	1.1926 E + 1	0.1041	4.0041 E + 0	0.1095
7	0.00	50.00	1.2163 E + 1	0.0628	4.0821 E + 0	0.0617
8	0.00	42.00	1.2233 E + 1	0.0506	4.1046 E + 0	0.0458
9 <sup>b</sup>	26.00	83.00	4.0803 E + 0	0.6486	1.3047 E + 0	0.6492
10	26.00	80.00	4.9329 E + 0	0.5243	1.5896 E + 0	0.5287
11	26.00	76.00	5.4742 E + 0	0.3063	1.8087 E + 0	0.1905
12	26.00	71.00	6.0681 E + 0	0.2568	1.9782 E + 0	0.1938
13	26.00	66.00	6.3843 E + 0	0.0560	2.0855 E + 0	0.0615
14	26.00	58.00	6.6843 E + 1	0.1076	2.1841 E + 0	0.1091
15	26.00	50.00	6.8309 E + 0	0.0688	2.2303 E + 0	0.0791
16	26.00	42.00	6.8747 E + 0	0.0578	2.2440 E + 0	0.0531

a. Center

b. Edge

Deviations from standard waste configurations are examined in this section. Figure C-11 lists deviations from standard configurations. Height changes from a right circular cylinder and nonhomogeneous source distributions are characterized.

- Changes in Height

The height of waste media may be varied by container shape and filling level of waste. Generally, the absorption of gamma radiation energy increases with height. These increments (or decrements) are not significant.

For a 6 x 6 liner, the energy absorption deviation ranges from 1 to 3% with a 50% increase in height<sup>a</sup> from right circular cylinder, and 2 to 8% decrease with 50% decrease in height<sup>b</sup>. As shown in Figure C-8, height

a. Waste Configuration (270-cm height x 90-cm radius).

b. Waste Configuration (90-cm height x 90-cm radius).

deviations from a right circular cylinder are bounded within 20 percent for most containers, these changes will not cause significant effect on energy absorption calculations.

- Source Distribution Changes

The generic calculations assume a homogeneous source distribution over the entire waste media. There can be two types of source distributions other than homogeneous distribution.

- a. One is source accumulated at the top of the waste media. It might be seen in spent cartridge type ion exchangers or filters, and is illustrated in Figure C-11 as deviation 3. When the source is accumulated in the top 50%, the energy absorption will be the same as to homogenous distribution, because the energy escaping

**Table C-6. Estimated error of representing detection points (in radius)**

Detector Coordinates (cm)			1.5 MeV		0.4 MeV	
Number	R	Z	Dose (rad/h)	Deviation (%)	Dose (rad/h)	Deviation (%)
1	0.00	83.00	7.1736 E + 0	0.3639	2.3942 E + 0	1.2703
2	7.00	83.00	7.0032 E + 0	0.2906	2.3085 E + 0	0.3057
3	14.00	83.00	6.5013 E + 0	0.2371	2.1324 E + 0	0.2500
4	19.00	83.00	5.8511 E + 0	0.1947	1.9594 E + 0	0.2008
5	23.00	83.00	5.0538 E + 0	0.5179	1.6316 E + 0	0.5192
6	26.00	83.00	4.0803 E + 0	0.5836	1.3047 E + 0	0.5726
7	0.00	42.00	1.2233 E + 1	0.8672	4.1046 E + 0	0.8526
8	7.00	42.00	1.1859 E + 1	0.2873	3.9745 E + 0	0.3016
9	14.00	42.00	1.0991 E + 0	0.2443	3.6662 E + 0	0.2527
10	19.00	42.00	9.8669 E + 0	0.2019	3.2696 E + 0	0.2527
11	23.00	42.00	8.4488 E + 0	0.4853	2.7906 E + 0	0.4779
12	26.00	42.00	6.8757 E + 0	0.4279	2.2440 E + 0	0.4123

Numbers 1-6: Edge

Numbers 7-12: Center

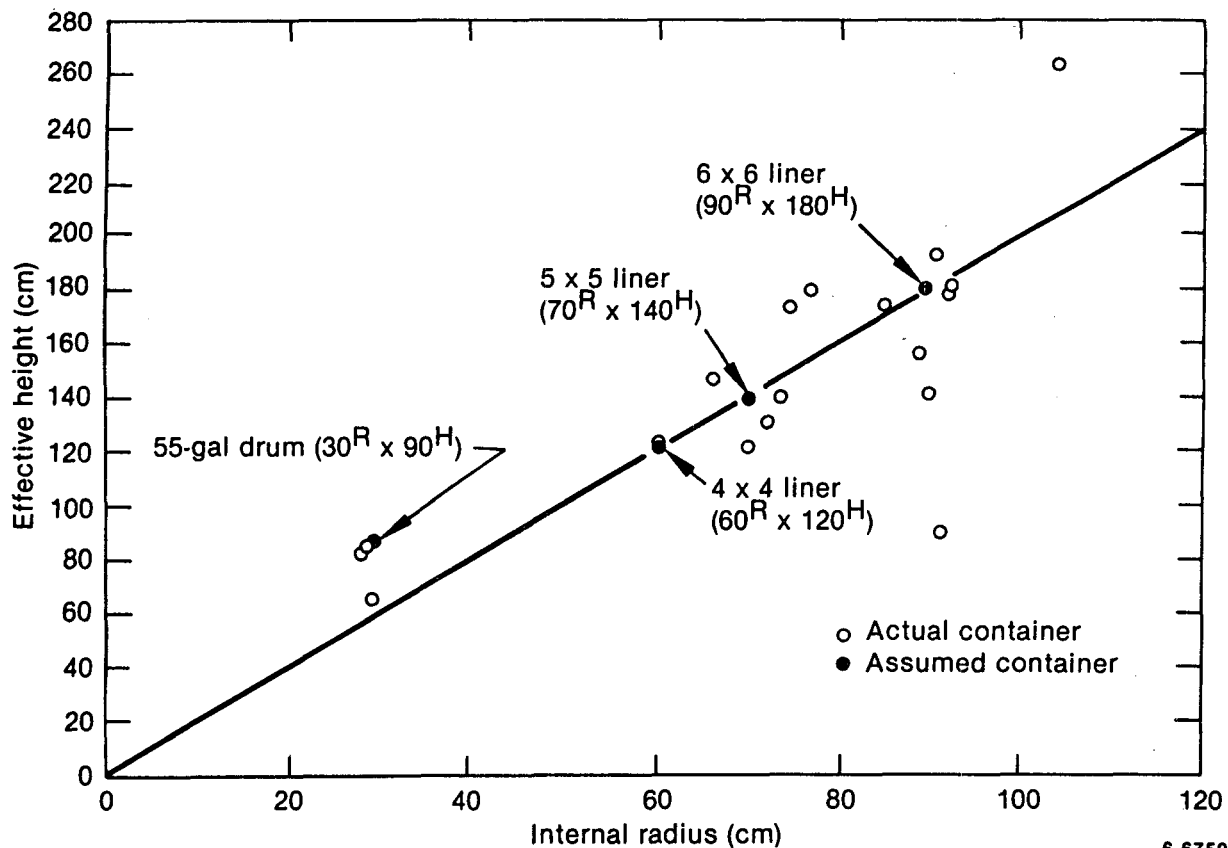


Figure C-7. Radius and height relationships for various waste containers.

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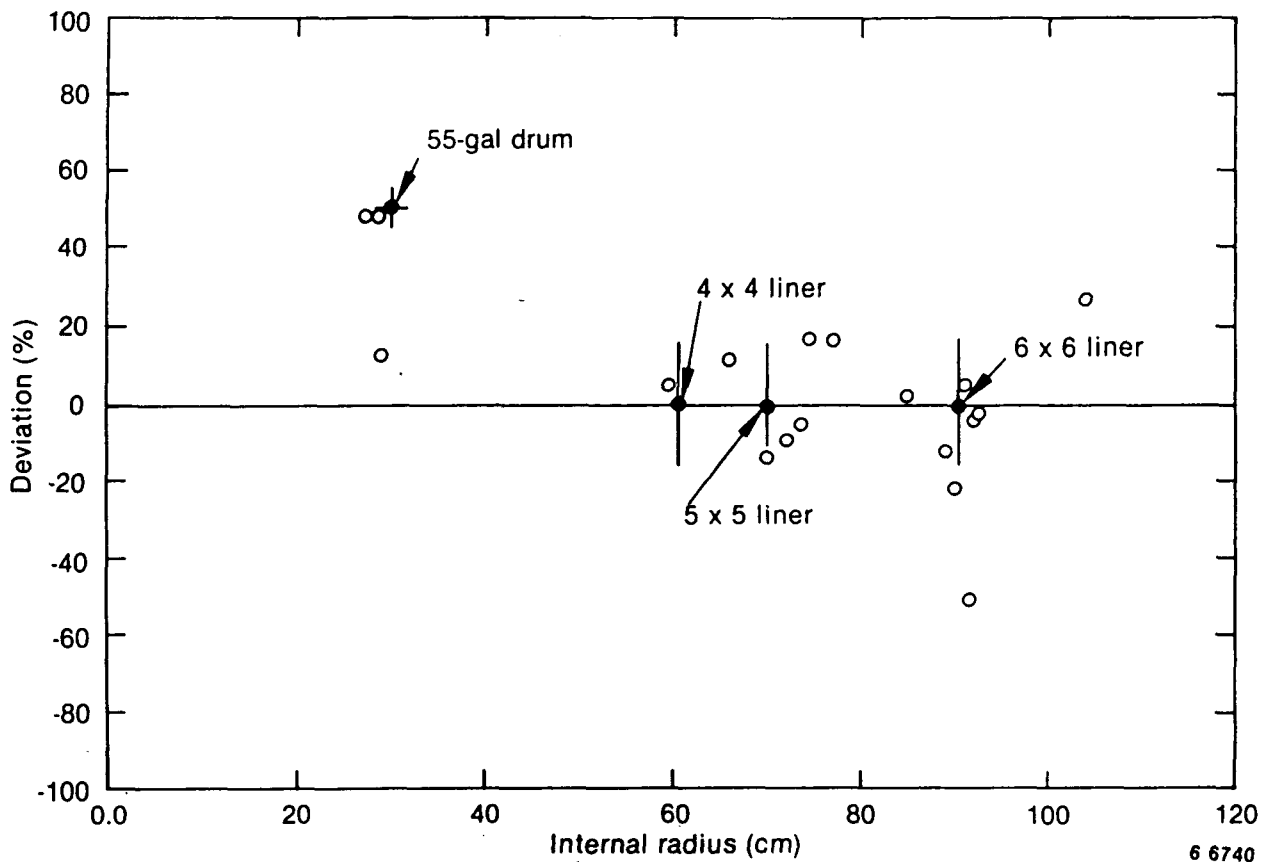


Figure C-8. Deviations from right cylinder for various waste containers.

from the top region to above is compensated by absorption in lower media. Practically, if the source region is limited to less than 50% in height at the top, the results of the generic calculation are applicable.

- b. The second type of distribution considers source accumulation at the central region of the contain-

ers. This occurs when radioactive materials such as cartridge filters are placed in the center of a container and the void is filled with binders to immobilize the waste. This is shown in Figure C-11 as deviation 4. When the source has a cylindrical shape, the absorbed energy inside the source can be calculated by using the generic method.

**Table C-7. Summary of calculational conditions for generic waste**

Variables	Range Considered
1. Source	
Energy Level	0.4 MeV to 2.0 MeV
Distribution	Homogeneous
2. Media	
Density	0.6 to 2.0 g/cm <sup>3</sup>
Material	Water <sup>a</sup>
3. Containers	
Configuration	Circular cylinder
Size	55 gal drum <sup>b</sup>
	4 ft x 4 ft liner <sup>c</sup>
	5 ft x 5 ft liner <sup>d</sup>
	6 ft x 6 ft liner <sup>e</sup>

a. The constants used are summarized in Appendix B.

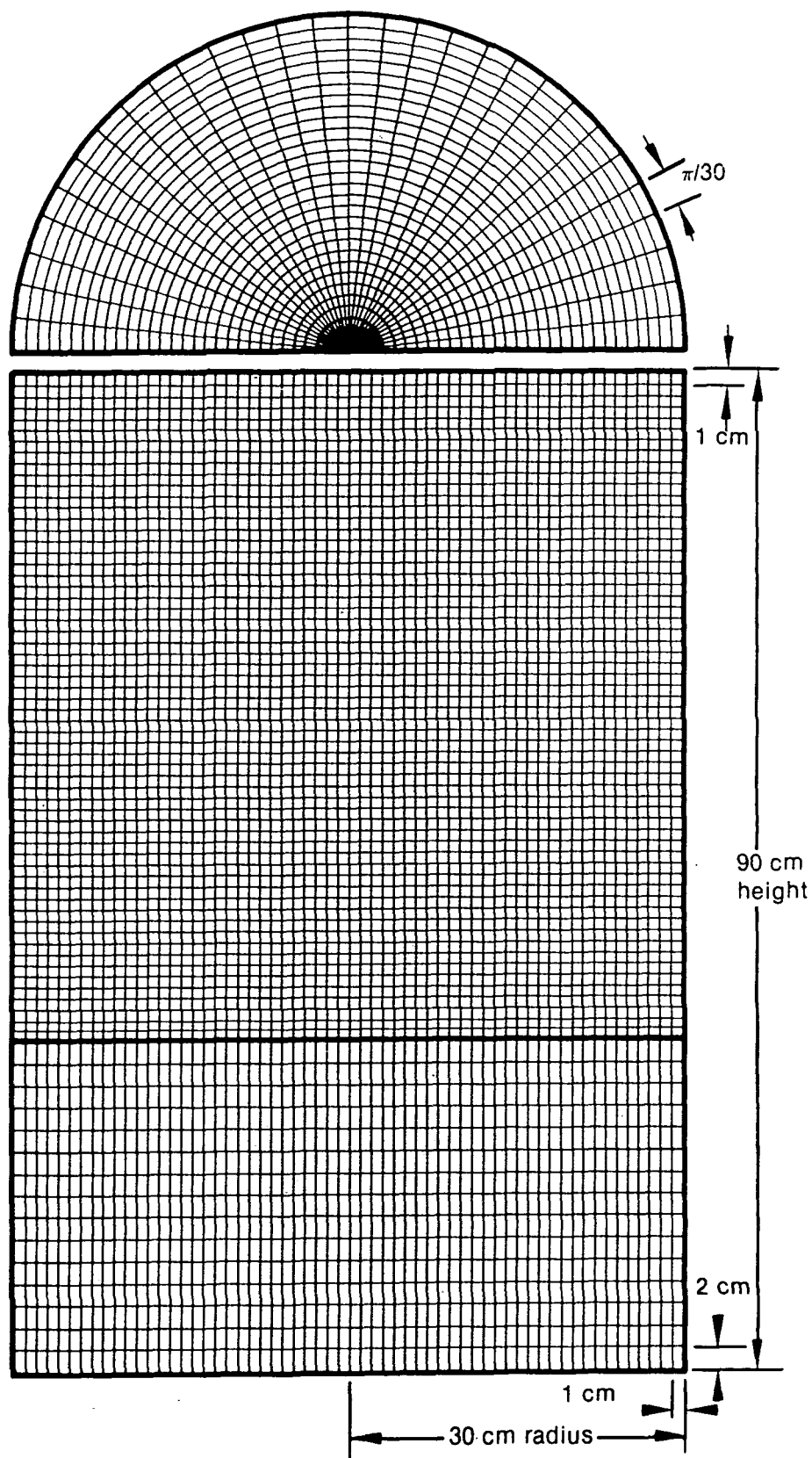
b. 30-cm radius x 90-cm height

c. 60-cm radius x 120-cm height

d. 70-cm radius x 140-cm height

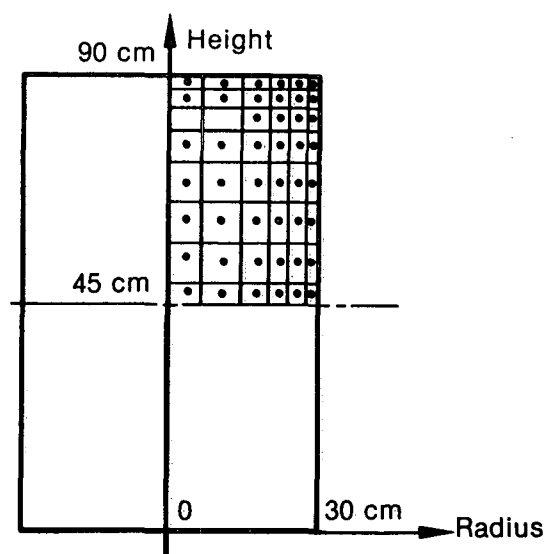
e. 90-cm radius x 180-cm height





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Figure C-9. Source points network for 55 gal. drum.



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Figure C-10. Detection point network for 55 gal. drum.

**Table C-8. Gamma energy absorption percentage**

Container <sup>a</sup>	Density <sup>b</sup>	Energy <sup>c</sup>			Note
		0.4	1.0	2.0	
30/90	6.0000E-01	45.6894	40.0910	34.3298	55 gal drum
	8.0000E-01	54.5134	48.3317	43.0109	
	1.0000E + 00	60.9414	54.6901	48.2414	
	1.5000E + 00	70.6475	65.0612	49.2058	
	2.0000E + 00	75.5871	70.8238	65.9195	
60/120	6.0000E-01	63.9283	57.5887	51.1064	4 x 4 liner
	8.0000E-01	71.0530	65.3157	59.1550	
	1.0000E + 00	75.5488	70.3911	64.8325	
	1.5000E + 00	81.4383	77.4160	73.3375	
	2.0000E + 00	83.9640	80.6775	77.6518	
70/140	6.0000E-01	68.1213	62.0894	55.6456	5 x 5 liner
	8.0000E-01	74.5177	69.1646	63.3853	
	1.0000E + 00	78.4773	73.7308	63.6713	
	1.5000E + 00	83.9372	79.8475	67.2470	
	2.0000E + 00	85.5449	82.2778	79.9512	
90/180	6.0000E-01	74.1570	68.6829	62.7335	6 x 6 liner
	8.0000E-01	79.3225	74.6983	69.6728	
	1.0000E + 00	82.5386	78.4175	74.1956	
	1.5000E + 00	86.3314	83.2331	80.2763	
	2.0000E + 00	87.6969	85.1385	83.1447	

a. Radius/Height, cm.

b. gram/cm<sup>3</sup>.

c. Million electron volt.

**Table C-9. Fitted constants for gamma radiation absorption**

Fitting Constant	Container Radius/Height (cm)			
	30/90	60/120	70/140	90/180
$a_1$	0.388	-0.0430	-0.0977	-0.321
$a_2$	-0.579	0.106	0.197	0.591
$a_3$	-0.871	0.117	0.276	0.748
$a_4$	-0.245	-0.518	-0.543	-0.634
$a_5$	1.354	-0.134	-0.423	-1.252
$a_6$	0.485	0.0547	-0.158	-0.402
$a_7$	0.954	1.371	1.428	1.549
$a_8$	-0.937	-0.137	0.0557	0.502
$a_9$	$3.302 \times 10^{-6}$	$-9.973 \times 10^{-7}$	$-1.554 \times 10^{-6}$	$-2.947 \times 10^{-6}$

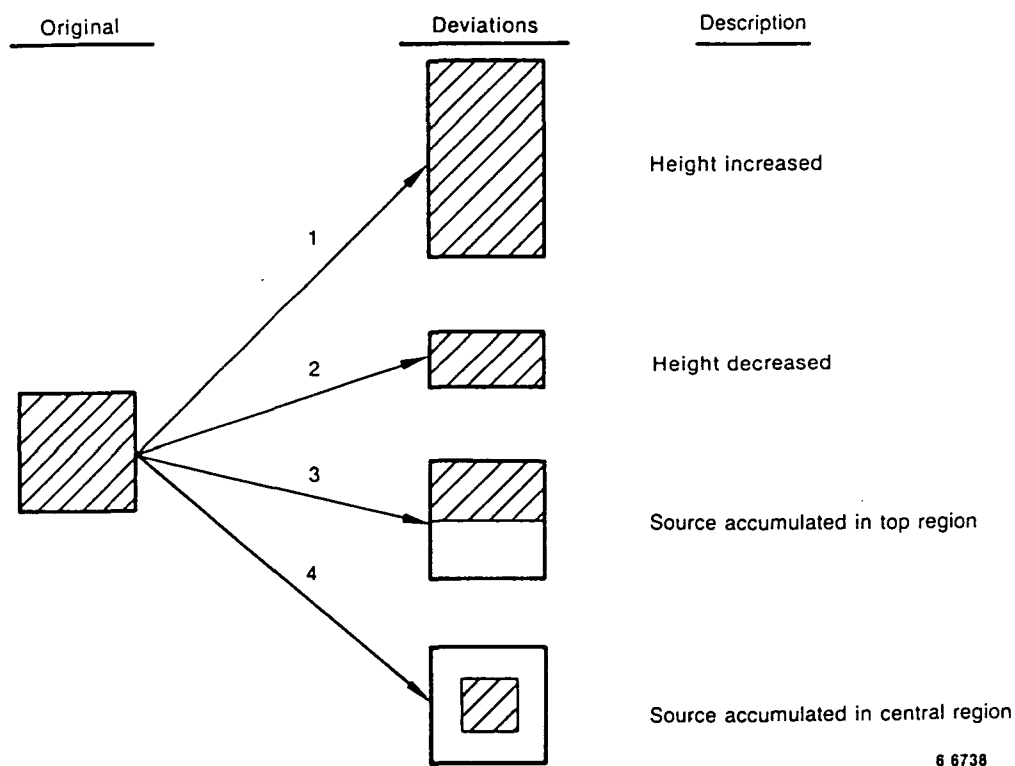


Figure C-11. Possible deviations from generic configuration.

## REFERENCES FOR APPENDIX C

- C-1. C. W. Neilson and E. E. Hochhalter, *QAD-FN, A General Purpose Shielding Design Code with Simplified Input*, Exxon Nuclear Idaho Company, Inc., March 1981.
- C-2. H. R. Helmholtz and J. K. Ryo, *Determination of Waste Container Curie Content from Dose Rate Measurements*, EPRI NP-3223, July 1983.

## **APPENDIX D**

### **DEVIATION OF ANALYTICAL SOLUTION FOR ENERGY ABSORPTION AT THE CENTER OF SPHERICAL SOURCE**

## APPENDIX D

### DEVIATION OF ANALYTICAL SOLUTION FOR ENERGY ABSORPTION AT THE CENTER OF SPHERICAL SOURCE

The energy absorption at the center of the sphere has been solved analytically without build-up factors<sup>D-1</sup>. It is modified with the same notation as Equation (4) in Appendix C,

- With integral

$$E_{abs/P}^{Center} = \mu_a \int_0^{R_0} \exp(-\mu_e r) dr \quad (D-1)$$

- Solution

$$E_{abs/P}^{Center} = I_0 \mu_a / \mu_e \{1 - \exp(-\mu_e R_0)\} \quad (D-2)$$

where,  $R_0$  is the radius of the sphere,  $E_{abs/P}^{Center}$  is the absorbed energy without buildup factor at the center of the sphere and has dimensions of MeV/s  $\cdot$  cm<sup>3</sup>. Other valuables are defined in Appendix C.

Equation (D-1) can be rewritten by combining the build-up factor as

$$E_{abs/B}^{Center} = \mu_a I_0 \int_0^{R_0} \exp(-\mu_e r) B(\mu_e r) dr \quad (D-3)$$

$$B(\mu_e r) \equiv \sum_{i=0}^3 B_i (\mu_e r)^i \quad (D-4)$$

where,  $E_{abs/B}^{Center}$  is the absorbed energy including build-up factor,  $B(\mu_e r)$ . Build-up factor is defined as a third order polynominal of mean free paths,  $\mu_e r$ , and  $B_i$  are the fitting constants.

Equation (D-3) can be solved by partial integration and becomes

$$E_{abs/B}^{Center} = \mu_a I_0 \left[ -1/\mu_e \exp(-\mu_e r) \right.$$

$$\left. B(\mu_e r) - 1/\mu_e^2 \exp(-\mu_e r) \right]$$

$$B'(\mu_e r) - 1/\mu_e^3 \exp(-\mu_e r)$$

$$B''(\mu_e r) - 1/\mu_e^4 \exp(-\mu_e r)$$

$$B'''(\mu_e r) \Big|_0^{R_0} \quad (D-5)$$

where

$$B'(\mu_e r) = B_1 \mu_e + 2B_2 \mu_e^2 r + 3B_3 \mu_e^3 r^2$$

$$B''(\mu_e r) = 2B_2 \mu_e^2 + 6B_3 \mu_e^3 r \quad (D-6)$$

$$B'''(\mu_e r) = 6B_3 \mu_e^3$$

Place Equation D-6 into Equation D-5 and evaluate at  $R_0$  and 0, then

$$E_{abs/B}^{Center} = \mu_a I_0 \left\{ \frac{1}{\mu_e} \left[ B_0 + B_1 + 2B_2 + 6B_3 \right] \right. \\ \left. - \exp(-\mu_e R_0) \left[ (B_0 + B_1 + 2B_2 + 6B_3)/\mu_e \right. \right. \\ \left. \left. + (B_1 + 2B_2 + 6B_3)R_0 + \mu_e (B_2 + 3B_3)R_0^2 + \mu_e^2 B_3 R_0^3 \right] \right\} \quad (D-7)$$

A more generic form is

$$E_{abs/B}^{Center} = \mu_a I_0 \left\{ \sum_{k=0}^3 \frac{1}{\mu_e^k} \right. \\ \left. + 1 \left[ B^{(k)}(0) - \exp(-\mu_e R_0) B^{(k)}(\mu_e R_0) \right] \right\} \quad (D-8)$$

where

$$B^{(K)}(\mu_e r) : k\text{-th derivative of } B(\mu_e r) \quad (D-9)$$

## REFERENCES FOR APPENDIX D

1. Herman Cember, *Introduction to Health Physics*, Pergamon Press, 1969, p. 168.