Calculation of Absorbed Doses to Water Pools in Severe Accident Sequences

Received by OSTI
JAN 13 1992

Prepared by
C. F. Weber

Oak Ridge National Laboratory

Prepared for
U.S. Nuclear Regulatory Commission

IRUTION OF THIS DOCUMENT IS UN

ED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

AVAILABILITY NOTICE

Availability of Reference Materials Cited in NRC Publications

Most documents cited in NRC publications will be available from one of the following sources:

1. The NRC Public Document Room, 2120 L Street, NW., Lower Level, Washington, DC 20555
2. The Superintendent of Documents, U.S. Government Printing Office, P.O. Box 37082, Washington, DC 20013-7082
3. The National Technical Information Service, Springfield, VA 22161

Although the listing that follows represents the majority of documents cited in NRC publications, it is not intended to be exhaustive.

Referenced documents available for inspection and copying for a fee from the NRC Public Document Room include NRC correspondence and internal NRC memoranda; NRC bulletins, circulars, information notices, inspection and investigation notices; licensee event reports; vendor reports and correspondence; Commission papers; and applicant and licensee documents and correspondence.

The following documents in the NUREG series are available for purchase from the GPO Sales Program: formal NRC staff and contractor reports, NRC-sponsored conference proceedings, international agreement reports, grant publications, and NRC booklets and brochures. Also available are regulatory guides, NRC regulations in the *Code of Federal Regulations*, and *Nuclear Regulatory Commission Issuances*.

Documents available from the National Technical Information Service include NUREG-series reports and technical reports prepared by other Federal agencies and reports prepared by the Atomic Energy Commission, forerunner agency to the Nuclear Regulatory Commission.

Documents available from public and special technical libraries include all open literature items, such as books, journal articles, and transactions. *Federal Register* notices, Federal and State legislation, and congressional reports can usually be obtained from these libraries.

Documents such as theses, dissertations, foreign reports and translations, and non-NRC conference proceedings are available for purchase from the organization sponsoring the publication cited.

Single copies of NRC draft reports are available free, to the extent of supply, upon written request to the Office of Administration, Distribution and Mail Services Section, U.S. Nuclear Regulatory Commission, Washington, DC 20555.

Copies of industry codes and standards used in a substantive manner in the NRC regulatory process are maintained at the NRC Library, 7920 Norfolk Avenue, Bethesda, Maryland, for use by the public. Codes and standards are usually copyrighted and may be purchased from the originating organization or, if they are American National Standards, from the American National Standards Institute, 1430 Broadway, New York, NY 10018.

DISCLAIMER NOTICE

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability of responsibility for any third party's use, or the results of such use, of any information, apparatus, product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights.

Calculation of Absorbed Doses to Water Pools in Severe Accident Sequences

Manuscript Completed: September 1991
Date Published: December 1991

Prepared by
C. F. Weber

Oak Ridge National Laboratory
Operated by Martin Marietta Energy Systems, Inc.

Oak Ridge National Laboratory
Oak Ridge, TN 37831-6285

Prepared for
Division of Systems Research
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555
NRC FIN B0854
Under Contract No. DE-AC05-84OR21400

MASTER

JK
DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

8
10
11
12

ABSTRACT

A methodology is presented for calculating the radiation dose to a water pool from the decay of uniformly distributed nuclides in that pool. Motivated by the need to accurately model radiolysis reactions of iodine, direct application is made to fission product sources dissolved or suspended in containment sumps or pools during a severe nuclear reactor accident. Two methods of calculating gamma absorption are discussed — one based on point-kernel integration and the other based on Monte Carlo techniques. Using least-squares minimization, the computed results are used to obtain a correlation that relates absorbed dose to source energy and surface-to-volume ratio of the pool. This correlation is applied to most relevant fission product nuclides and used to actually calculate transient sump dose rate in a pressurized-water reactor (PWR) severe accident sequence.

CONTENTS

ABSTRACT	iii
CONTENTS	v
LIST OF FIGURES	vi
LIST OF TABLES	vii
ACKNOWLEDGMENTS	viii
1. INTRODUCTION	1
2. POINT-KERNEL INTEGRATION	4
3. MONTE CARLO APPROACH	8
4. COMPUTATIONAL RESULTS	10
4.1 POINT-KERNEL INTEGRATION RESULTS	10
4.2 MONTE CARLO RESULTS	12
4.3 ANALYSIS OF RESULTS	15
5. APPLICATION TO FISSION PRODUCT SOURCES IN REACTOR ACCIDENT SEQUENCES	16
6. CONCLUSIONS	21
REFERENCES	22
APPENDIX A	25
APPENDIX B	31
APPENDIX C	33
APPENDIX D	35

LIST OF FIGURES

1.	Rectangular water volumes for point kernel integration	11
2.	Absorbed fraction of energy in water pools	13
3.	Sump dose rate for Surry TMLB' sequences	20
A.1	Cartesian nodalization in Z direction	26
A.2	Subdivision of major mesh cells	28
A.3	Element pairings equal distances apart: (a) nonaligned, (b) aligned . . .	29

LIST OF TABLES

1. Penetration of radiation in water	2
2. Energy transfer (Kerma) buildup factors for an isotropic point source in water	5
3. Photon interaction (cross section) groups and energy boundaries for Monte Carlo calculations	9
4. Source energies and absorption coefficients	10
5. Geometrical data and fractional energy absorption in various pool sizes .	12
6. Monte Carlo absorbed energy calculations	14
7. ^{97}Nb decay modes	16
8. Fission product and actinide groupings	18
D.1. Mass fractions of nuclides in basic element groups	37

ACKNOWLEDGMENTS

The author is greatly indebted to several staff members of the Oak Ridge National Laboratory for assistance in this effort. As project leader, E. C. Beahm provided overall direction and motivation. Jabo Tang performed the Monte Carlo calculations using the MORSE code system. Conversations with him and Jeff Ryman regarding dose absorption were very helpful during all stages of the work. Finally, thanks are due to Lindy Norris for resurrecting an old manuscript and diligently working it into final form.

1. INTRODUCTION

A major unresolved issue in nuclear reactor safety involves the volatility of fission product iodine. This concern has been prompted by the paucity of data and reliable models to describe iodine speciation, especially in aqueous environments subject to high-radiation fields. Because most reactor safety studies suggest that considerable amounts of iodine would reside in such locations,¹ there is reason to suspect some of the results and conclusions of these studies. However, it is also important to recognize that these studies have been based on the best science available; for criticism to be legitimate and constructive, it must also include improved understanding.

Under accident conditions, iodine and other fission products can enter a variety of sumps and water pools in the containment regions of a nuclear plant. In boiling-water reactors (BWRs), the pathway may lead directly to a large tank of water called a pressure suppression pool. In pressurized-water reactors (PWRs), or in certain BWR scenarios, iodine may escape into the gas space of the containment building. If venting to the atmosphere does not occur immediately, considerable amounts would be (1) dissolved in condensing steam or water sprays, or (2) deposited on surfaces with condensing steam or settling particulate aerosols. Subsequent surface washing and spray collection would drain sizable quantities of iodine into system sumps. Thus, most accident sequences in either BWRs or PWRs involve dissolution of iodine in water pools or sumps.

Aqueous iodine is generally thought to reside in the highly soluble species I^- and IO_3^- , which remain immobile in many accident scenarios. Recent experimental work has indicated dramatically increased formation of volatile iodine (I_2 , CH_3I) in irradiated, low-pH solutions.² Because such situations are quite plausible in many accident sequences, it is important for reactor safety analyses to allow for the existence of volatiles and to model their formation accurately.

To make a quantitative calculation of volatile iodine formation in water, it is necessary to make a reliable prediction of radiation dosage, which relies on the inventory of various radioactive nuclides in water (the overwhelming source of radiation) and the tendency of the pool to absorb the radiation emitted. Determination of these quantities is the focus of the current work.

The dominant dose of radiation to containment water pools is expected to arise largely from dissolved or suspended fission products in the water (actinides may also contribute). Although contributions from the adjacent air space or settled material may be significant in certain instances, they are ignored in this work. The present justification is that there is no available accident sequence code that reliably predicts fission product inventories in such locations; therefore, the dose from radioactive sources in such locations would be highly uncertain. Thus, it is assumed that all fission products in water are uniformly distributed, and the decay of radioactive nuclides creates a uniform volumetric radiation source. The energy from this source may likely

be absorbed in the water but may also enter concrete or steel structures surrounding the water. This energy will either escape entirely or be reflected into the water, where a positive contribution to the water dose could result.

Essentially all radioactive fission products undergo β^- decay and simultaneously emit γ photons of varying energies. Actinides undergo α or β decay and also emit γ radiation. Table 1 lists several representative types and energies of radiation that may result from such decays. Also shown are the mean-free paths (i.e., the average distances traveled in water before some interaction occurs).³ Because the distances traveled by α and β^- particles are quite small, virtually all their energy is expected to be absorbed, except decays occurring in the immediate vicinity of the water boundary. For large pools, this implies that essentially all the energy from α and β^- particles will be deposited in the water, and it is hereby assumed without further discussion. This assumption obviously breaks down for puddles, very small pools, water droplets, wall surface films, and other water locations where surface-to-volume ratio is quite large.

Table 1. Penetration of radiation in water

Radiation type	Mean-free path (cm)
0.1 MeV γ	6
1.0 MeV γ	14
5.0 MeV γ	33
2.0 MeV β	0.9
5.0 MeV α	0.003

In contrast with the α and β^- particles, the γ radiation may travel some distance before an interaction; thus, noticeable energy may escape from the water pool to be absorbed in the surroundings. For this reason, the assumption of total absorption is incorrect, and a more detailed analysis is necessary to determine the water dose. The problem is calculating the total dose actually absorbed by a water pool that results from a volumetric γ source distributed uniformly in that pool. Two methods have been used to perform such calculations: (1) point-kernel integration using buildup factors, and (2) Monte Carlo computations using the MORSE code.⁴ The calculational procedures and results for each of these methods are described in later sections of this report.

The theory behind the point-kernel integration is presented in Sect. 2, where an equation for absorbed dose is derived. This equation requires numerical solution, and these procedures are described in Appendix A. The Monte Carlo approach is outlined briefly in Sect. 3; a description of the MORSE code can be found elsewhere.⁴ Results from both approaches are given in Sect. 4, where a correlation is developed linking absorbed dose to source energy and surface-to-volume ratio of the pool. This correlation is applied to many fission product nuclides in Sect. 5, resulting in useful

expressions that can be used in actual accident sequence evaluations. In addition, a few results of such applications are described briefly. Finally, Sect. 6 gives a summary of the accomplishments and applicability of this work.

2. POINT-KERNEL INTEGRATION

The dose to a small volume of water caused by incident photons can be modeled as if the water volume element were a detector. The absorbed water dose would then be analogous to the detector response. If the detector is located at a point $\mathbf{r}$, then the response R is given by³

$$R(\mathbf{r}) = \int_0^\infty \frac{\mu_k E}{\rho} \phi(\mathbf{r}, E) dE, \quad (1)$$

where

- R = photon dose rate absorbed at $\mathbf{r}$ (MeV/s·g),
- μ_k = linear energy absorption coefficient (cm^{-1}),
- E = photon energy (MeV),
- ρ = water density (g/cm^3),
- ϕ = gamma flux density ($\text{photons}/\text{cm}^2 \cdot \text{s} \cdot \text{MeV}$).

The coefficient μ_k is one of several absorption coefficients used in shielding and dosimetry. This particular one was chosen because it represents contributions from ionizing processes (Compton scattering, pair production, and photoelectric effect) corrected for bremsstrahlung losses. This choice is appropriate because the major concern of this work is the dose contribution to radiolysis effects in aqueous chemistry. For a monoenergetic source of energy E_o , the upper integration limit can be changed from ∞ to E_o . However, the integration in Eq. (1) is still necessary to account for scattered photons that will be absorbed at $\mathbf{r}$ with a reduced energy $E < E_o$. If the flux ϕ is the result of an isotropic point source and the water pool is infinite, then the response can also be represented using buildup factors by the expression

$$R(\mathbf{r}) \approx B(E_o, \mu r) \frac{\mu_k E_o}{\rho} \phi(\mathbf{r}, E_o), \quad (2)$$

where $r = |\mathbf{r}|$ and μ (cm^{-1}) is the linear attenuation coefficient, sometimes referred to as the macroscopic cross section. This quantity represents all types of interactions, and its inverse $1/\mu$ is the mean-free path (i.e., the average distance traveled before an interaction). The buildup factor $B(E_o, \mu r)$ is an empirical function that allows Eq. (2) to replace Eq. (1) in most cases. Because real pools are not infinite and real sources are not point sources, Eq. (2) is only an approximation. Use of it will obviously produce some inaccuracy in the calculations; in most situations, these inaccuracies are not substantial.

Buildup factors themselves can be obtained from experimental measurement, but this method is not a trivial process. It is difficult for experimental results to cover the range of applicability that is usually necessary, although the experimental data are quite useful in validating results calculated by theoretical methods. Concerning the

latter, the moments method has been quite popular and has been used by Chilton et al.⁵ to calculate the water buildup factors shown in Table 2.

Table 2. Energy transfer (Kerma) buildup factors
for an isotropic point source in water

μr	$E_o(\text{MeV})$					
	0.1	0.2	0.5	1	2	5
0	1.00	1.00	1.00	1.00	1.00	1.0
0.5	2.36	1.92	1.61	1.47	1.38	1.29
1	4.52	3.42	2.45	2.08	1.83	1.57
2	11.7	8.22	4.87	3.62	2.82	2.10
3	23.5	15.7	8.29	5.50	3.87	2.62
4	40.6	26.4	12.4	7.66	4.99	3.12
5	64.0	41.3	18.1	10.1	6.16	3.63
6	94.8	61.0	24.6	12.8	7.38	4.14
7	134	86.2	32.2	15.7	8.66	4.64
8	183	118	40.8	18.9	9.97	5.14
10	314	202	61.8	26.0	12.7	6.14
15	917	582	137	47.4	20.1	8.62
20	2,120	1,310	247	73.5	28.0	11.1
25	4,260	2,580	395	104	36.4	13.5
30	7,780	4,640	582	138	45.2	15.9
35	13,100	7,890	809	175	54.3	18.3
40	20,300	12,800	1,080	214	63.6	20.7

Source: A. B. Chilton, C. M. Eisenhauer, and G. L. Simmons,
Nucl. Sci. Eng. **73**(1), 97-107 (1980).

The response in Eq. (2) represents the dose rate at the point $\mathbf{r}$. To get the rate of energy absorption in the small volume $d\mathbf{r}$ (surrounding $\mathbf{r}$), Eq.(2) is multiplied by $\rho d\mathbf{r}$ to get

$$R(\mathbf{r})\rho d\mathbf{r} = \mu_k E_o B(E_o, \mu r) \phi(E_o, \mathbf{r}) d\mathbf{r} . \quad (3)$$

The total energy deposited (per unit time) in the entire region V_d is then obtained by summing (integrating) over all such incremental volumes in V_d :

$$R_d = \lim_{d\mathbf{r} \rightarrow 0} \sum R(\mathbf{r})\rho d\mathbf{r} = \iiint_{V_d} R(\mathbf{r})\rho d\mathbf{r} = \mu_k E_o \iiint_{V_d} B(E_o, \mu r) \phi(\mathbf{r}, E_o) d\mathbf{r} . \quad (4)$$

This equation represents the total response (absorbed dose) of a water volume acting as a detector. If the flux ϕ arises from a point source located at the origin, then it can be represented as⁶

$$\phi(\mathbf{r}, E_o) = S \frac{e^{-\mu r}}{4\pi r^2}, \quad (5)$$

where S is the source strength (photons/s). The exponential term in Eq. (5) represents attenuation by interactions with the medium; the term $(4\pi r^2)^{-1}$ represents geometric attenuation as the radiation spreads radially outward. Substituting Eq. (5) into Eq. (4) yields

$$R_d = \frac{\mu_k E_o S}{4\pi} \iiint_{V_d} B(E_o, \mu r) \frac{e^{-\mu r}}{r^2} dr, \quad (6)$$

which represents the total dose absorbed by the water per unit time in V_d due to a point source at the origin of strength S and energy E_o . For a source located at some point other than the origin, for example at $\mathbf{r}_s$, the response is obtained by simply substituting $|\mathbf{r} - \mathbf{r}_s|$ for r in Eq. (6).

In most applications to water pools, it is expected that the source of radiation will be uniformly distributed throughout the pool volume rather than concentrated at a single point. Thus, it is necessary to consider the absorbed dose response to a volumetric source density S_v (photons/cm³·s) distributed in the source volume V_s , which may coincide exactly with the response volume V_d .

If $d\mathbf{r}_s$ is a small volume surrounding the point $\mathbf{r}_s$ (and $d\mathbf{r}_s$ is a subset of V_s), then $S_v d\mathbf{r}_s$ behaves effectively as a point source located at $\mathbf{r}_s$. Substituting this for S in Eq. (6), the total dose response becomes

$$R_d = \frac{\mu_k E_o}{4\pi} S_v d\mathbf{r}_s \iiint_{V_d} B(E_o, \mu |\mathbf{r} - \mathbf{r}_s|) \frac{e^{-\mu |\mathbf{r} - \mathbf{r}_s|}}{|\mathbf{r} - \mathbf{r}_s|^2} d\mathbf{r}. \quad (7)$$

The total dose due to radiation from the entire source volume V_s is obtained by summing the contributions of every incremental volume $d\mathbf{r}_s$ within V_s . As each $d\mathbf{r}_s \rightarrow 0$, this results in an integral over V_s :

$$R_T = \frac{\mu_k E_o S_v}{4\pi} \iiint_{V_s} \iiint_{V_d} B(E_o, \mu |\mathbf{r} - \mathbf{r}_s|) \frac{e^{-\mu |\mathbf{r} - \mathbf{r}_s|}}{|\mathbf{r} - \mathbf{r}_s|^2} d\mathbf{r} d\mathbf{r}_s. \quad (8)$$

Thus, Eq. (8) represents the total absorbed dose rate to a water pool occupying volume V_d due to a uniformly distributed source in volume V_s with strength density S_v and energy E_o .

As seen in Eq. (8), it is necessary to perform two volume integrals to obtain the total dose response R_T . Even if V_s and V_d coincide exactly and represent a simple

geometrical shape, obtaining a solution is still a difficult problem and is virtually impossible to do analytically. It is therefore necessary to use numerical techniques to perform the integrations in Eq. (8). This procedure has been done to obtain the results described in Sect. 4 (shown in Appendix A).

3. MONTE CARLO APPROACH

In Monte Carlo analysis, numerous histories of randomly generated photons are followed and the resulting interactions treated as samplings of true system behavior. Each photon repeatedly travels a distance and interacts with an atom of the medium (which need not be homogeneous). Both the distance traveled and the interaction results are determined by drawing random numbers and comparing with the probabilities for various outcomes. Because every interaction causes a photon to lose strength, each photon eventually is totally absorbed unless it first escapes from the absorbing medium of interest (in this case, water). If a water medium is surrounded by concrete or steel, then photons escaping the water may interact in the surrounding regions and be reflected into the water at reduced energy.

In practice, the statistical weight of each photon is reduced by the ratio of the scattering cross section to total cross section at each collision site, and no absorption is permitted. This form of weight biasing usually leads to lower variances of the quantity being estimated (e.g., energy deposition) than would be the case for the analog transport described above. To keep the transport "game" fair, a photon history may be terminated based upon the results of a procedure called Russian roulette after its weight has dropped below some predetermined value. This form and other possible forms of transport biasing are described in the MORSE manual.⁴

It is important that a sufficient number of histories be included to ensure that the behavior of the samples will provide an accurate picture of the entire system in a statistical sense. Group structure is an approximation commonly used in radiation transport methods. Rather than consider all photon energies on a continuum between zero and the source energy E_o , the energy variable is discretized, as illustrated in Table 3. The energies E_i , $i=1,2,\dots,n$ form boundaries between groups whose behavior represents an integrated average behavior (in some sense). Thus, if a photon has energy $E \in (E_i, E_{i+1})$, it will behave as any other photon with energy in the same group. Since group structure does constitute a discrete approximation, the accuracy of calculations, obviously, depends somewhat on the fineness of the group structure used.

The absorbed energy in a given region is obtained by simply adding the energies deposited by all photons that have interacted within the region. This total absorbed energy will include contributions from photons at the source energy E_o as well as those at lower energy (partially attenuated by previous interactions).

Table 3. Photon interaction (cross-section) groups and energy boundaries for Monte Carlo calculations

Group	Energy (MeV)	Group	Energy (MeV)
1	10	10	1.33
2	8	11	1.0
3	6.5	12	0.8
4	5	13	0.6
5	4	14	0.4
6	3	15	0.3
7	2.5	16	0.2
8	2.0	17	0.1
9	1.66	18	0.05
	1.33		0.01

4. COMPUTATIONAL RESULTS

Numerous calculations have been performed using both point-kernel integration and the Monte Carlo approach. A wide range of source energies was used, and several different sized water pools were considered. All pools were shaped as rectangular solids, but ten different dimensional variations were used. In all cases, the source and detector volumes [V_s and V_d in Eq. (8)] are identical and coincide exactly with the actual pool volumes. The calculational results are used in Sect. 5 to develop a correlation that relates the fraction of source energy absorbed to the source energy itself and to the surface-to-volume ratio of the pool.

4.1 POINT-KERNEL INTEGRATION RESULTS

The energy absorption rate was determined from the numerical approximation to Eq. (8), which is described in Appendix A. The buildup factors shown in Table 2 were used in tabular form, with linear interpolation used for distances between those stated. For simplicity, the source strength S_v was assumed equal to unity (this is varied in the application to actual nuclides given in Sect. 5). The interaction coefficients μ_k and μ were obtained from Chilton et al.³ and are shown in Table 4. Also depicted in the table are the various energies at which calculations were performed. It is expected that virtually all ionizing processes will result from fission product or actinide decay modes that yield γ energies between 0.1 and 5 MeV; therefore, the energies used span the interval of principal interest for water radiolysis considerations.

Table 4. Source energies and absorption coefficients

E_o (MeV)	$\mu(\text{cm}^{-1})$	$\mu_k(\text{cm}^{-1})$
0.1	0.163	0.0244
0.2	0.133	0.0291
0.5	0.0952	0.0326
1.0	0.0697	0.0306
2.0	0.0486	0.0256
5.0	0.0271	0.0172

Source: Calculation using data taken from A. B. Chilton, J. K. Shultis, and R. E. Faw, *Principles of Radiation Shielding*, Prentice-Hall, Englewood Cliff, N.J. (1984), pp. 122, 126; and assuming water density to be $0.9858 \text{ g}\cdot\text{cm}^{-3}$.

Calculations have been performed in ten different rectangular pools, which are illustrated in Fig. 1 and listed in Table 5. These pools were chosen to represent a wide range of surface areas and volumes because they are the primary geometrical factors in leakage and absorption, respectively. All pools were assumed to be completely homogeneous and composed of pure water.

Fig. 1. Rectangular water volumes for point-kernel integration.
(Dimensions are in meters.)

For each of the source energies listed in Table 4 and repeated as headings of columns 6 through 11 in Table 5, calculations were performed to determine total absorbed energy in each of the ten pools. Dividing absorbed energy by source energy yields the absorbed fraction of energy; these values appear as entries in columns 6 through 11 of Table 5. As expected, the absorbed fractions increase as the surface-to-volume ratio S/V decreases and as source energy E_o decreases. Thus, for $E_o > 2$ MeV, less than one-half of the energy is absorbed in a cubic pool of 0.6 m on a side. On the other hand, all source energies considered produce at least 90% absorption in a cubic pool 10 m on a side. Several absorbed fractions are actually greater than 1 (a physical impossibility), due primarily to the approximation errors involved in using buildup factors. In addition to the listing in Table 5, data for $E_o = 0.1, 1$, and 5 MeV are plotted in Fig. 2, along with results from Monte Carlo calculations. As seen in the figure, the absorbed fractions are fairly linear functions of S/V over a wide range.

Table 5. Geometrical data and absorbed fraction of energy for various pool sizes

Run no.	Pool dimensions (m)				Absorbed fraction of energy for listed source energies (MeV)					
	X	Y	Z	S/V (m^{-1})	0.1	0.2	0.5	1	2	5
1	0.6	0.6	0.6	10	0.6949	0.6788	0.6090	0.5513	0.4780	0.3789
2	2.4	2.4	200	1.677	0.9633	0.9890	0.9358	0.9047	0.8682	0.8100
3	0.8	12.5	100	2.680	0.9232	0.9405	0.8814	0.8414	0.7907	0.7105
4	0.5	40	50	4.090	0.8675	0.8740	0.8087	0.7607	0.6996	0.6062
5	0.4	50	50	5.080	0.8295	0.8296	0.7620	0.7112	0.6471	0.5552
6	10	10	10	0.6	1.0047	1.0385	0.9909	0.9684	0.9469	0.9175
7	6	6	6	1	0.9893	1.0201	0.9704	0.9447	0.9176	0.8773
8	3	3	3	2	0.9517	0.9753	0.9209	0.8879	0.8484	0.7851
9	2	2	2	3	0.9154	0.9323	0.8739	0.8347	0.7849	0.7042
10	1	1	1	6	0.8139	0.8138	0.7470	0.6948	0.6259	0.5224

4.2 MONTE CARLO RESULTS

Calculations were performed using the MORSE computer code,⁴ which was developed primarily as a tool for shielding and radiation penetration analyses. All computations were done using the 18-group photon-interaction library shown in Table 3. Because of the expense of three-dimensional Monte Carlo runs, only a few energies and pool sizes were considered. These were chosen to provide maximum information and to compare with the point-kernel results. Sufficient photon histories were used to ensure that the following statistical accuracy was achieved:

$$\frac{\text{standard deviation}}{\text{mean absorbed energy}} < 0.01.$$

Fig. 2. Absorbed fraction of energy in water pools.

Because all three desired source energies ($E_o = 0.1, 1.0, 5.0$ MeV) fell on energy group boundaries, sources from both adjoining groups were chosen, weighted inversely by group ΔE , so that the average source energy $\bar{E}$ was equal to the desired value of E_o . Thus, to achieve a source strength of 1 and source energy $E_o = 1$, two actual sources were used in the calculations, one from energy group 10 with strength S_{10} and one from group 11 with strength S_{11} , where

$$S_{10} = \frac{\Delta E_{11}}{\Delta E_{10} + \Delta E_{11}} = 0.377, \quad S_{11} = \frac{\Delta E_{10}}{\Delta E_{10} + \Delta E_{11}} = 0.623. \quad (9)$$

The average source energy is then

$$\bar{E} = S_{10} \bar{E}_{10} + S_{11} \bar{E}_{11} = (0.377)(1.167) + (0.623)(0.9) = 1.00 \text{ MeV}. \quad (10)$$

Even though the average source energy is the same, this difficulty introduces a slight discrepancy that should be realized when comparing Monte Carlo and point-kernel results.

A second problem is the discretization error arising from the formation of energy groups. As seen in Table 3, any photon scattered from group 16 would have a resultant energy in group 16, 17, or 18. This division is quite coarse and could have a significant impact on calculations using $E_o = 0.1$ MeV. The problem is much less serious for $E_o = 1.0$ MeV and is probably not a factor at all for $E_o = 5.0$ MeV.

Actual results of the MORSE calculations are listed in Table 6 and are also plotted in Fig. 2. Agreement with point-kernel results is fairly good for $E_o = 1$ and 5 MeV but not as good for $E_o = 0.1$ MeV. Both methods could be at fault—Monte Carlo through the poor energy group discretization, and point kernel through higher buildup-factor inaccuracies at $E_o = 0.1$ MeV.

Table 6. Monte Carlo absorbed energy calculations

Pool size	S/V	Reflector	Absorbed fraction of energy for listed source energies (MeV)		
			0.1	1.0	5.0
1.1 m	5.45	1 m water	0.783	0.704	0.566
1.1 m	5.45	30 cm concrete	0.754	0.701	0.560
2.5 m	2.4	30 cm concrete	0.873	0.849	0.765

Finally, note that the type and size of the reflector does have some impact on the results. In actual reactor settings, reflectors would be concrete (containment sumps), steel (BWR pressure suppression pools), or some combination of the two. Physically speaking, reflection from water is not altogether different than reflection from concrete

(or even steel). Furthermore, the comparison between water and concrete reflectors (the pool with sides of 1.1 m in Table 6) shows that the results are practically indistinguishable at 1 and 5 MeV and slightly different at $E_o = 0.1$ MeV. Hence, it is a reasonable (and slightly conservative) assumption that a water reflector will have general applicability, regardless of the structural geometry in the actual containment to be studied. For this reason, the point-kernel results, which assume infinite water reflection, will be used in subsequent analyses.

4.3 ANALYSIS OF RESULTS

As mentioned previously, a significant body of data has been gathered from code calculations of absorbed energy. These results for rectangular pools are now assumed to be transferable to pools of any geometry; that is, the only geometrical factor affecting energy absorption is the surface-to-volume ratio S/V .

The data plotted in Fig. 2 are fairly linear between surface-to-volume ratios of 1 and 5 m^{-1} . For each source energy, the fraction f of absorbed energy can be fitted to a linear form using standard least-squares minimization:

$$f = a + b(S/V) .$$

When plotted, the constants a and b appear to be highly linear functions of $\sqrt{E}$. Thus, it should be possible to fit all the absorbed fraction data in Table 5 to the simple functional form

$$f = (a_1 + a_2\sqrt{E}) + (b_1 + b_2\sqrt{E})(S/V) . \quad (11)$$

This fit has been done, again by minimizing the sum of squared error

$$R = \sum_{i,j} [(a_1 + a_2\sqrt{E_i}) + (b_1 + b_2\sqrt{E_i})(S/V)_j - f_{ij}]^2 . \quad (12)$$

The actual solution procedures are detailed in Appendix B and result in the following parameters:

$$a_1 = 1.062 \quad a_2 = -0.051 \quad b_1 = -0.0355 \quad b_2 = -0.0216 .$$

These results are used in the next chapter to obtain dose estimates for actual fission product sources.

5. APPLICATION TO FISSION PRODUCT SOURCES IN REACTOR ACCIDENT SEQUENCES

Most radioactive nuclides experience many decay modes; that is, a number of γ and particle energies could result during decay. Each has its own frequency of occurrence, known as intensity, so that the decay of a nuclide is completely described by a list of possible particle and γ energies, together with the corresponding intensity for each energy. Such a list has been reproduced from ref. 7 and is depicted as Table 7. Denoting the individual γ decay energies and intensities as E_i and I_i , respectively, the average γ energy per decay is

$$\bar{E} = \sum_i E_i I_i . \quad (13)$$

Table 7. ^{97}Nb decay modes^a

β^- decay		γ decay	
Average energy (MeV)	Intensity (%)	Energy (MeV)	Intensity (%)
0.0882	0.118	0.481	0.15
0.1266	0.167	0.658	98.09
0.2176	0.206	1.025	1.08
0.3149	1.081	1.269	0.16
0.4698	98.30	1.516	0.12

^aDecay modes with intensity less than 0.1% not included.

Source: D. C. Kocher, *Radioactive Decay Data Tables*, DOE/TIC-11026, Technical Information Center, U.S. Department of Energy, Oak Ridge, Tenn. (1981).

If this decay took place in a water pool, the fraction of the energy E_i that would be deposited can be determined from Eq. (11). Thus, for a given nuclide, the average energy (MeV) absorbed in water from each γ decay is

$$E_\gamma = \sum_i [a_1 + a_2 \sqrt{E_i} + (b_1 + b_2 \sqrt{E_i})(S/V)] E_i I_i , \quad (14)$$

which can be rewritten as

$$E_\gamma = A + B(S/V) , \quad (15)$$

where

$$A = \sum_i (a_1 + a_2 \sqrt{E_i}) E_i I_i, \quad B = \sum_i (b_1 + b_2 \sqrt{E_i}) E_i I_i. \quad (16)$$

Using a nuclear data base⁸ to obtain values for E_i and I_i , the constants A and B have been calculated for most important fission product nuclides and are listed in Appendix C.

A second quantity of interest is the specific γ dose rate (absorbed dose per unit mass of nuclide), which is calculated from E_γ :

$$\dot{d}_\gamma = \frac{1.602 \times 10^{-11} E_\gamma \lambda N_a}{M m_{wat}}, \quad (17)$$

where

- $\dot{d}_\gamma$ = specific dose rate due to γ photons (rad/s.g),
- E_γ = average energy (MeV) absorbed in water per γ decay,
- λ = nuclide decay constant (s^{-1}),
- N_a = Avogadro's number,
- M = atomic weight of nuclide,
- m_{wat} = mass of water present (kg).

The absorbed energy in Eq. (17) refers only to contributions from γ radiation. If it is augmented by energy from α and β particles (assumed totally absorbed), then the resulting dose rate represents the total specific dose rate for a given nuclide:

$$\dot{d} = 1.602 \times 10^{-11} \frac{(E_\alpha + E_\beta + E_\gamma) \lambda N_a}{M m_{wat}}. \quad (18)$$

The values for E_α and E_β are averages obtained by applying Eq. (13) for these radiation types. Because no nuclide of interest undergoes both α and β decay, either E_α or E_β will always be zero.

A number of available codes are available for calculating fission product transport of radioactive species into containment water pools.⁹⁻¹¹ None of these, however, has the need or the capability of transporting nuclides individually. Rather, they are lumped together as elements or groups of elements whose chemical and physical behavior is thought (or hoped) to be similar. Several such groupings have been used in the past; Table 8 shows what has been developed recently with the VANESA code.¹²

Table 8. Fission product and actinide groupings

Group no.	Characteristic element	Member elements	Specific dose rate ^a	
			(rad/s·g) × 10 ³	(Mrad/h·g) × 10 ⁵
1	Te	Te,Se	4.34	1.56
2	I	I,Br	141.8	51.04
3	Cs	Cs,Rb	1.40	0.505
4	Ru	Ru,Tc,Rh,Pd	3.23	1.16
5	Ce	Ce,Np,Pu,Am,Cm	0.564	0.203
6	La	La,Y,Pr,Nd,Pm,Sm,Eu	8.28	2.98
7	Sr	Sr	10.98	3.95
8	Zr	Zr ^b	6.00	2.16
9	Nb	Nb	311.3	112.1
10	Mo	Mo	1.43	0.515
11	Ba	Ba	3.27	1.18
12	Sb	Sb	1,617	582.3
13	Xe	Xe,Kr	1.01	0.362

^aRadiation dose rate per unit mass of fission products.

^bIncludes only fission product nuclides.

In many accident sequences, the water inventory of each group can be determined as it varies throughout a transient calculation. Because the overall goal is to predict water dose resulting from fission product decay, it is necessary to know the decay behavior of each fission product group (which may contain many nuclides). Hence, the mass fraction x_j of nuclide j in the group must be known. These fractions are generally not constant, although they should only vary slightly among accident sequences. Output from the ORIGEN code¹³ has been used to calculate such fractions at a time of 5 h after shutdown for the Browns Ferry Plant near the end of the Cycle 6 fuel loading. Details of the procedure are described in Appendix D. These fractions are given in Table D.1 for all nuclides making significant contributions to group dose or mass. Since most short-lived nuclides have decayed away after 5 h and the production of fission products is essentially the same in most light-water reactors (LWR), these fractions may be generally applicable during transients greater than 5 h and even to sequences involving different reactor types.

Using the mass fractions in Appendix D and dose rates from Eq. (18), a specific dose rate can be calculated for each nuclide group as follows:

$$\dot{D} = \sum_j \dot{d}_j x_j, \quad (19)$$

where

- $\dot{D}$ = specific dose rate for element group (rad/s·g);
 $\dot{d}_j$ = specific dose rate for all radiation from nuclide j within the group (rad/s·g),
calculated from Eq. (18).

Thus, if m is the mass of a given elemental group in water, then $m\dot{D}$ is the resulting pool dose received from nuclides in that group. Total pool dosage is then determined by simply adding dose contributions from each group. Using a water mass of 1.15×10^5 kg and $S/V = 3.5 \text{ m}^{-1}$, values of $\dot{D}$ for the nuclide groups in Table 8 are listed there in the fourth column. These pool values are approximately those of the Surry reactor containment sump during a Station Blackout Accident Sequence.¹ The same dose rates are stated in different units in the fifth column.

The methodology described previously has been used in certain accident sequence calculations where iodine radiolysis reactions were significant in contributing to overall iodine volatility. The computations were performed by the TRENDS code ("TRENDS: Transport and Retention of Nuclides in Dominant Sequences," by C. F. Weber et al., to be published), a special code designed to include such reactions in fission product transport calculations. Values of $\dot{D}$ were input into the code; the group masses m in water pools were computed using aerosol transport parameters from other code calculations. As an example of the results, the dose rate to the containment sump of the Surry Plant was calculated during an entire Station Blackout Accident Sequence and is shown in Fig. 3. Analogous calculations also have been performed for Peach Bottom sequences, where dose to the pressure suppression pool was an important factor in iodine radiolysis.

Fig. 3. Sump dose rate for Surry TMLB' sequences.

6. CONCLUSIONS

The methodology developed in previous sections is capable of providing a quantitative estimate of absorbed radiation in water pools, given the mass inventories of various nuclides, elements, or element groups. In addition, certain geometric information is needed to calculate the surface-to-volume ratio (S/V) for the pool (surface meaning the boundary on all sides). This dosage estimate is of particular importance to iodine volatility calculations because radiolytic conversion of soluble I^- and IO_3^- into volatile I_2 or CH_3I depends closely on dose rate and cumulative absorbed dose.

The procedure assumes that all α and β^- radiation is totally absorbed, and calculates the leakage and absorption of γ emissions. For $S/V < 1$, absorption is over 90% for all fission product decays. However, as S/V increases past 1, absorption will decline, and highly energetic γ radiation will show considerable energy leakage for most types of boundary reflectors. In most cases, containment sumps will still absorb 70 to 80% of the radiated energy.

Although not of primary interest, the methodology could also be used to calculate heatup of water pools, air spaces, or other homogeneous mixtures. This capability might be especially useful for unusual geometries, although it would be complicated by other sources of heat gain or loss and absorption of decay energy in surroundings.

Finally, it is anticipated that this work could be extended to several other situations where effects of irradiation and fission product behavior are dependent on peculiar geometries. Doses to air and airborne water droplets are two examples of volumetric absorption. Surface irradiation from air or water volumes and surface sources formed by deposited fission products could be modeled by changing a volume integral into a surface integral in Eq. (8). Unlike the present analysis for water pools, such situations would generally absorb only a small fraction of incident radiation. They are further complicated by the need to consider leakage of α and β^- radiation since total absorption may not be a good assumption. Nevertheless, although some modifications would be necessary, the present methodology is capable of calculating absorbed doses in these situations.

REFERENCES

1. J. A. Gieseke et al., *Radionuclide Release Under Specific LWR Accident Conditions*, BMI-2104, Battelle Columbus Laboratories, Columbus, Ohio (1984).
2. S. J. Wisbey, E. C. Beahm, W. E. Shockley, and Y.-M. Wang, "Iodine Behavior in Containment Under LWR Accident Conditions," *Proceedings of the Symposium on Chemical Phenomena Associated with Radioactivity Releases During Severe Nuclear Plant Accidents*, NUREG/CP-0078, Anaheim, Calif. (September 1986).
3. A. B. Chilton, J. K. Shultis, and R. E. Faw, *Principles of Radiation Shielding*, Prentice-Hall, Englewood Cliff, N.J. (1984), pp. 122, 126.
4. M. B. Emmett, *The MORSE Monte Carlo Radiation Transport Code System*, ORNL-4972, Union Carbide Corp. Nucl. Div., Oak Ridge Natl. Lab. (1975).
5. A. B. Chilton, C. M. Eisenhauer, and G. L. Simmons, *Nucl. Sci. Eng.* **73**(1), 97-107 (1980).
6. T. Rockwell (ed.), *Reactor Shielding Design Manual*, TID-7004, D. Van Nostrand, Princeton, N.J. (1956).
7. D. C. Kocher, *Radioactive Decay Data Tables*, DOE/TIC-11026, Technical Information Center, U.S. Department of Energy, Oak Ridge, Tenn. (1981).
8. L. T. Dillman, *EDISTR — A Computer Program to Obtain a Nuclear Decay Data Base for Radiation Dosimetry*, ORNL/TM-6689, Union Carbide Corp. Nucl. Div., Oak Ridge Natl. Lab. (1980).
9. K. D. Bergeron et al., *User's Manual for CONTAIN 1.0, A Computer Code for Severe Nuclear Reactor Accident Containment Analysis*, NUREG/CR-4085 (SAND-84-1204), Sandia National Laboratories, Albuquerque, NM (May 1985).
10. *MAAP3.0 User's Manual*, IDCOR Technical Report 16.2-3, Fauske and Associates, Inc., Burr Ridge, Ill. (February 1987).
11. J. A. Gieseke et al., *Source Term Code Package: A User's Guide*, NUREG/CR-4587 (BMI-2138), Battelle Memorial Institute, Columbus, Ohio (April 1986).
12. D. A. Powers, J. E. Brockman, and A. W. Shiver, *VANESA: A Mechanistic Model of Radionuclide Release and Aerosol Generation During Core Debris Interactions with Concrete*, NUREG/CR-4308 (SAND-85-1370), Sandia National Laboratories, Albuquerque, N.M. (September 1986).
13. A. G. Croff, *ORIGEN2 — A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code*, ORNL-5621, Union Carbide Corp. Nucl. Div., Oak Ridge Natl. Lab. (July 1980).

APPENDIXES

APPENDIX A

NUMERICAL SOLUTION OF POINT-KERNEL INTEGRALS

Letting the source and detector regions be equal (i.e., $V_s = V_d \equiv V$), Eq. (8) can be rewritten as

$$R_T = \iiint_V \iiint_V I(|\mathbf{r} - \mathbf{r}_s|) d\mathbf{r} d\mathbf{r}_s, \quad (\text{A.1})$$

where $I(|\mathbf{r} - \mathbf{r}_s|)$ represents the integrand of Eq. (8). If V is now assumed to be a rectangular region, numerical procedures can be developed for three-dimensional slab geometry in a straightforward manner.

Each of the Cartesian directions x , y , and z are divided into equally spaced nodes, creating a three-dimensional mesh of small rectangular regions. Denoting the mid-points of mesh rectangles by $\mathbf{r}_{ijk}$ or $\mathbf{r}_{\ell mn}$ ($i, \ell = 1, \dots, I$; $j, m = 1, \dots, J$; $k, n = 1, \dots, K$), Eq. (A.1) can be discretized to yield

$$R_T \cong \sum_{i,j,k} \sum_{\ell,m,n} I(|\mathbf{r}_{ijk} - \mathbf{r}_{\ell mn}|) (\Delta v)^2, \quad (\text{A.2})$$

where $\Delta v = \Delta x \Delta y \Delta z$.

Consider now the two-dimensional mesh illustrated in Fig. A.1. Because the mesh spacing is uniform, the distances between nodes 1-2, 2-3, 3-4, 4-5, and 5-6 are all identical. Because Eqs. (A.1) and (A.2) depend only on relative distances between position vectors, the sums over these index combinations are all identical: $(k, n) = (1, 2), (2, 3), (3, 4), (4, 5), (5, 6)$. Furthermore, distances between 1-3, 2-4, 3-5, 4-6 are also the same, as are 1-4, 2-5, 3-6, and 1-5, 2-6; hence, identical sums also occur in these cases. The following expression uses this information to reduce the effort of summing that is present in Eq. (A.2):

$$R_T = (\Delta v)^2 \sum_{i,j} \sum_{\ell,m} \sum_{k=1}^K (K+1-k) \alpha_k I(|\mathbf{r}_{ij1} - \mathbf{r}_{\ell mk}|), \quad (\text{A.3})$$

where

$$\alpha_s = \begin{cases} 1, & s = 1 \\ 2, & s > 1. \end{cases}$$

When $k > 1$, then $\alpha_k = 2$, representing a source-receiver exchange between two disjoint mesh regions (each acts as a source for the other to receive); when $k = 1$, then $\alpha_k = 1$, because a cell is a source to itself only once.

ORNL DWG 87-19201

Fig. A.1. Cartesian nodalization in Z direction.

Analogous treatment of variables in the x and y directions yields

$$R_T = (\Delta v)^2 \sum_{k=1}^K \sum_{j=1}^J \sum_{i=1}^I \alpha_i \alpha_j \alpha_k (I+1-i)(J+1-j)(K+1-k) I(|\mathbf{r}_{111} - \mathbf{r}_{ijk}|), \quad (\text{A.4})$$

which can be solved with considerably less computational effort than Eq. (A.2).

Although Eq. (A.4) is more efficient than Eq. (A.2), some details of accuracy have been overlooked and require correction. In the special situation where $i = j = k = 1$, the integrand term of Eq. (A.4) is $I(\mathbf{r}_{111} - \mathbf{r}_{111}) = I(0)$. A quick glance at Eq. (8) indicates that this term is singular and must be modified. A second problem arises from the use of mesh cell midpoints in the numerical approximation, which is analogous to treating both source and receiver as points, rather than three-dimensional volumes. This approximation is good when the cells are far apart (i.e., when the distance $|\mathbf{r}_{ijk} - \mathbf{r}_{lmn}|$ is large compared with the cell dimensions $\Delta x, \Delta y$, or Δz . However, when the cells are adjacent or nearby (and especially when source and receiver are the same cell), special treatment is needed.

The corrective procedure simply involves refining the discrete mesh by further subdivision of nearby mesh cells. Consider the four mesh cells pictured in Fig. A.2 (denoted by the Roman numerals I, II, III, and IV) and the 36-node subdivisions of each.

Integrating the interactions of two large cells requires integrating the interactions of each element of one with each element of the other. Two cases are encountered, as illustrated by the x -direction integrations of a row in cell I with rows in cell II and cell IV.

The integration of a row in cell I and a row in cell IV represents any situation in which the elements are not aligned at the same positions in the direction of integration. For this situation, interelement distances occur between pairs of elements as shown in Fig. A.3a. Combinations listed in the same row have identical distances, implying identical integrals; rows are in order of decreasing distance. This example has six elements in the subdivision. For the general case of M elements, there would be $2M - 1$ different distances. If arranged in decreasing order, the i th distance would have multiplicity

$$m_x(i) = M - |M - i|. \quad (\text{A.5})$$

The integration of a row in cell I with a row in cell II represents a situation in which the elements are aligned in the direction of integration, as shown in Fig. A.2. That is, elements a and 1 have the same coordinate in the x direction, as do b and 2, c and 3, etc. Pairings for the different interelement distances are depicted in Fig. A.3b, revealing a different arrangement than in the nonaligned case. For the general case of M elements in the subdivision, there are M distances. If arranged in decreasing order, the i th distance has multiplicity

Fig. A.2. Subdivision of major mesh cells.

1F
 1E 2F
 1D 2E 3F
 1C 2D 3E 4F
 1B 2C 3D 4E 5F
 1A 2B 3C 4D 5E 6F
 2A 3B 4C 5D 6E
 3A 4B 5C 6D
 4A 5B 6C
 5A 6B
 6A

(a)

1f
 1e 2f
 1d 2e 3f
 1c 2d 3e 4f
 1b 2c 3d 4e 5f
 1a 2b 3c 4d 5e 6f
 6a
 6b 5a
 6c 5b 4a
 6d 5c 4b 3a
 6e 5d 4c 3b 2a

(b)

Fig. A.3. Element pairings equal distances apart: (a) nonaligned, (b) aligned.

$$m_x(i) = \begin{cases} 2i, & i < M \\ i, & i = M \end{cases} \quad (\text{A.6})$$

This same analysis is applicable to the y and z directions as well.

Now, let N_x , N_y , and N_z represent the number of different distances, related to the number of subdivisions as discussed above,

$$N_x = \begin{matrix} 2M_x - 1, & \text{nonaligned} \\ M_x, & \text{aligned} \end{matrix},$$

$$N_y = \begin{matrix} 2M_y - 1, & \text{nonaligned} \\ M_y, & \text{aligned} \end{matrix},$$

$$N_z = \begin{matrix} 2M_z - 1, & \text{nonaligned} \\ M_z, & \text{aligned} \end{matrix}.$$

Further, define the distance between different subdivision cells ρ_{ijk} by

$$\rho_{ijk}^2 = (\bar{x} + \Delta x - i\delta x)^2 + (\bar{y} + \Delta y - j\delta y)^2 + (\bar{z} + \Delta z - k\delta z)^2,$$

where

$\bar{x}, \bar{y}, \bar{z}$ = Cartesian distances between midpoints of major cells (I, II, III, or IV in Fig. A.2),

$\Delta x, \Delta y, \Delta z$ = dimensions of major cells,

$\delta x, \delta y, \delta z$ = dimensions of subdivision cells.

The integrations over subdivisions of two major cells can then be expressed as

$$\sum_{k=1}^{N_z} \sum_{j=1}^{N_y} \sum_{i=1}^{N_x} M_x(i) M_y(j) M_z(k) I(\rho_{ijk}) (\delta x \delta y \delta z)^2, \quad (\text{A.7})$$

In practice, two levels of subdivisions are used (i.e., the major mesh cells are subdivided when sufficiently close, and further subdivision is undertaken when these small cells are sufficiently close). This subdividing of cells is quite necessary because of the magnitude of the integrand $I(\cdot)$ at short distances [cf. Eq. (8)].

A small code has been written to perform the calculations described above. Two subroutines are used for the primary and secondary subdivision integration steps, respectively. The code has run on a CRAY-XMP, with run times varying considerably for different geometries and different nodalization parameters. None of the cases pictured in Fig. 1 had execution times longer than 5 min, and many were under 1 min. Runs with different step-size parameters indicate that all results are probably accurate to at least two significant figures.

APPENDIX B

LEAST-SQUARES FITTING OF DOSE CORRELATION

From Eq. (12), the quantity to be minimized is

$$R = \sum_{i,j} S_{ij}^2, \quad (\text{B.1a})$$

where

$$S_{ij} \equiv (a_1 + a_2 \sqrt{E_i}) + (b_1 + b_2 \sqrt{E_i})(S/V)_j - f_{ij}. \quad (\text{B.1b})$$

Since $\sqrt{E}$ and (S/V) are independent variables, the constants a_1 , a_2 , b_1 , and b_2 are independent as well. Differentiating Eq. (B.1) with respect to each parameter and setting to zero yields

$$\begin{aligned} \frac{\partial R}{\partial a_1} = 0 &= 2 \sum_{i,j} S_{ij} & \frac{\partial R}{\partial a_2} = 0 &= 2 \sum_{i,j} \sqrt{E_i} S_{ij} \\ \frac{\partial R}{\partial b_1} = 0 &= 2 \sum_{i,j} (S/V)_j S_{ij} & \frac{\partial R}{\partial b_2} = 0 &= 2 \sum_{i,j} \sqrt{E_i} (S/V)_j S_{ij}. \end{aligned}$$

If I and J are the numbers of energies E_i and ratios $(S/V)_j$, respectively, then the above can be rewritten as the set of linear equations:

$$IJ a_1 + AJ a_2 + IB b_1 + AB b_2 = F_1$$

$$AJ a_1 + CJ a_2 + AB b_1 + CB b_2 = F_2$$

$$IB a_1 + AB a_2 + ID b_1 + AD b_2 = F_3$$

$$AB a_1 + BC a_2 + AD b_1 + CD b_2 = F_4,$$

where

$$A = \sum_{i=1}^I \sqrt{E_i}, \quad B = \sum_{j=1}^J (S/V)_j; \quad C \equiv \sum_{i=1}^I E_i, \quad D = \sum_{j=1}^J (S/V)_j^2,$$

$$F_1 = \sum_{i,j} f_{ij}, \quad F_2 = \sum_{i,j} \sqrt{E_i} f_{ij},$$

$$F_3 = \sum_{i,j} (S/V)_j f_{ij}, \quad F_4 = \sum_{i,j} \sqrt{E_i} (S/V)_j f_{ij} .$$

Performing Gaussian elimination by hand, it can be determined that the unknowns are

$$b_2 = \frac{IJF_4 - AJF_3 - BIF_2 + ABF_1}{(IC - A^2)(JD - B^2)} ,$$

$$b_1 = \frac{JF_3 - BF_1}{I(DJ - B_2) - A Ia_4} ,$$

$$a_2 = \frac{IF_2 - AF_1}{J(CI - A^2) - BJa_4} ,$$

$$a_1 = (F_1 - AJa_2 - B Ia_3 - ABa_4)/IJ .$$

Using the data in Table 5, the constants a_1 , a_2 , b_1 , and b_2 are calculated by the above procedure to yield the values stated below Eq. (12).

APPENDIX C

LINEAR FORM CONSTANTS FOR NUCLIDE DOSES

Listing of constants A and B appearing in Eq. (16)

Nuclides	A	B	Nuclides	A	B
^{85m}Kr	1.644E-01	7.091E-03	^{132}I	2.316E+00	1.255E-01
^{87}Kr	7.918E-01	4.938E-02	^{132}Te	2.427E-01	1.051E-02
^{88}Kr	1.937E+00	1.276E-01	^{133}I	6.202E-01	3.187E-02
^{88}Rb	6.250E-01	4.022E-02	^{133m}Te	2.341E-01	1.304E-01
^{89}Sr	8.565E-05	4.743E-06	^{133}Xe	4.836E-02	1.886E-03
^{90}Y	1.785E-06	6.472E-08	^{134}Cs	1.584E+00	8.365E-02
^{91}Sr	7.066E-01	3.911E-02	^{134}I	2.657E+00	1.482E-01
^{91m}Y	5.427E-01	2.737E-02	^{135}I	1.582E+00	9.483E-02
^{91}Y	3.635E-01	2.140E-04	^{135m}Xe	4.397E-01	2.191E-02
^{92}Sr	1.342E+00	8.101E-02	^{135}Xe	2.573E-01	1.162E-02
^{92}Y	2.542E-01	1.443E-02	^{136}Cs	2.197E+00	1.203E-01
^{93}Y	8.954E-02	5.218E-03	^{137m}Ba	6.086E-01	3.163E-02
^{95}Nb	7.788E-01	4.167E-02	^{137}Cs	0.0	0.0
^{95}Zr	7.519E-01	3.998E-02	^{139}Ba	4.433E-02	1.966E-03
^{97m}Nb	7.410E-01	3.941E-02	^{140}Ba	1.877E-01	9.148E-03
^{97}Nb	6.686E-01	3.480E-02	^{140}La	2.323E+00	1.394E-01
^{97}Zr	1.813E-01	1.020E-02	^{141}Ce	7.949E-02	3.309E-03
^{99}Mo	1.532E-01	7.916E-03	^{141}La	4.298E-02	2.633E-03
^{99m}Tc	1.317E-01	5.505E-03	^{142}La	2.726E+00	1.807E-01
^{103m}Rh	1.847E-03	6.753E-05	^{142}Pr	5.828E-02	3.656E-03
^{103}Ru	4.806E-01	2.383E-02	^{143}Ce	2.909E-01	1.381E-02
^{105}Rh	8.023E-02	3.701E-03	^{143}Pr	9.063E-09	4.818E-10
^{105}Ru	8.005E-01	4.138E-02	^{144}Ce	2.164E-02	8.795E-14
^{106}Rh	2.091E-01	1.082E-02	^{144}Pr	3.185E-02	1.980E-03
^{109m}Ag	1.173E-02	4.422E-04	^{145}Pr	1.336E-02	7.254E-04
^{109}Pd	1.238E-02	4.751E-04	^{147}Nd	1.448E-01	6.701E-03
^{111}Ag	2.722E-02	1.264E-03	^{148m}Pr	2.041E+00	1.058E-01
^{112}Ag	6.621E-01	3.835E-02	^{148}Pr	5.792E-01	3.343E-02
^{127}Sn	1.923E+00	1.120E-01	^{149}Nd	3.963E-01	1.852E-02
^{127}Te	5.015E-03	2.393E-04	^{149}Pr	1.101E-02	5.187E-04
^{129}Te	6.106E-02	2.996E-03	^{151}Pr	3.301E-01	1.570E-02
^{131}I	3.928E-01	1.874E-02	^{153}Sm	6.493E-02	2.570E-03
^{131m}Te	1.445E+00	7.936E-02	^{156}Eu	1.332E+00	8.091E-02
^{131}Te	4.304E-01	2.168E-02			

APPENDIX D

DETERMINATION OF FISSION-PRODUCT NUCLIDE INVENTORIES

Information has been supplied by Tennessee Valley Authority (TVA) on the loading pattern of the Cycle 6 core at Browns Ferry Unit 1. Six different fuel assembly types were involved, and each type had its own enrichment and gadolinium content. In addition, the power history was known for each assembly in the core. Defining P_ℓ^k = power history of the ℓ^{th} assembly of type k , then the highest and lowest power in each fuel type are

$$\bar{P}^k = \max_\ell (P_\ell^k) \text{ and } \underline{P}^k = \min_\ell (P_\ell^k) .$$

For each assembly type ($k=1,\dots,6$), ORIGEN¹³ calculations were performed using $\bar{P}^k$ and $\underline{P}^k$, yielding the following quantities at various times after shutdown:

$\bar{n}_j^k$ = inventory of nuclide j (g-atom) in the assembly of type k with the highest power history,
 $\underline{n}_j^k$ = inventory of nuclide j (g-atom) in the assembly of type k with the lowest power history.

Picking the values at 5 h after shutdown, nuclide inventories in all other assemblies were then determined by linear interpolation:

$$n_{j\ell}^k = \underline{n}_j^k + (\bar{n}_j^k - \underline{n}_j^k) \frac{(P_\ell^k - \underline{P}^k)}{(\bar{P}^k - \underline{P}^k)} , \quad (\text{D.1})$$

where $n_{j\ell}^k$ = inventory (g-atom) of nuclide j in the ℓ^{th} assembly of fuel type k (whose power history is P_ℓ^k). The whole core inventory of nuclide i is then obtained by summation:

$$N_j = \sum_{k=1}^6 \sum_{\ell} n_{j\ell}^k . \quad (\text{D.2})$$

The total mass of nuclides in element group p (cf. Table 8) is then

$$m_p = \sum_{j \in \text{group } p} \frac{N_j}{M_j} ,$$

where M_j = atomic weight of nuclide j , and the mass fraction of nuclide j within group p is

$$x_j^p = \frac{N_j/M_j}{m_p}, \quad j \in \text{group } p.$$

Group mass fractions for many fission-product nuclides are listed in Table D.1. Only nuclides whose mass or dose contributions are significant at five hours after shutdown have been included.

In some situations, it may be useful to calculate nuclide inventories in various regions, rather than over the entire core. If A_m are axial power factors for the core, and

$$\bar{A} = \max_m (A_m) \quad \underline{A} = \min_m (A_m),$$

then the nodes of highest and lowest power in assembly type k are $\bar{A}\bar{P}^k$ and $\underline{A}\underline{P}^k$, respectively. Interpolations analogous to Eq. (D.1) can then be done over each axial region, and summing analogous to Eq. (D.2) can be done only over individual radial regions. This yields local nuclide inventories in axial and radial nodal regions.

Table D.1. Mass fractions of nuclides in basic element groups

Based on ORIGEN¹³ calculations for Browns Ferry
Cycle 6, 5 h after shutdown

Group	Nuclide	Group mass fraction	Group	Nuclide	Group mass fraction
1	⁷⁷ Se	1.946E-03 ^a	5	¹⁴¹ Ce	4.827E-03
1	⁷⁸ Se	4.592E-03	5	¹⁴² Ce	9.469E-02
1	⁷⁹ Se	1.119E-02	5	¹⁴³ Ce	1.725E-04
1	⁸⁰ Se	2.567E-02	5	¹⁴⁴ Ce	3.080E-02
1	⁸² Se	6.475E-02	5	²³⁹ Np	7.272E-03
1	¹²⁵ Te	8.698E-03	5	²³⁸ Pu	9.397E-03
1	¹²⁸ Te	2.054E-01	5	²³⁹ Pu	5.542E-01
1	¹²⁹ Te	1.787E-05	5	²⁴⁰ Pu	1.937E-01
1	¹³⁰ Te	6.690E-01	5	²⁴¹ Pu	9.882E-02
1	^{131m} Te	3.071E-04	5	²⁴¹ Am	2.923E-03
1	¹³² Te	8.409E-03	5	²⁴² Cm	9.556E-04
			5	²⁴⁴ Cm	2.296E-03
2	⁸¹ Br	8.470E-02			
2	¹²⁷ I	1.997E-01	6	⁸⁹ Y	5.802E-02
2	¹²⁹ I	6.777E-01	6	⁹¹ Y	6.626E-03
2	¹³¹ I	3.028E-02	6	⁹¹ Y	1.658E-06
2	¹³² I	5.143E-04	6	⁹² Y	1.223E-05
2	¹³³ I	5.919E-03	6	⁹³ Y	4.244E-04
2	¹³⁴ I	0.0	6	¹³⁹ La	1.640E-01
2	¹³⁵ I	1.221E-03	6	¹⁴⁰ La	4.273E-04
			6	¹⁴¹ La	1.736E-05
3	⁸⁵ Rb	3.113E-02	6	¹⁴² La	1.766E-06
3	⁸⁷ Rh	7.773E-02	6	¹⁴¹ Pr	1.432E-01
3	⁸⁸ Rb	5.960E-07	6	¹⁴³ Pr	2.956E-03
3	¹³³ Cs	3.576E-01	6	¹⁴⁴ Pr	2.084E-06
3	¹³⁴ Cs	3.100E-02	6	¹⁴³ Nd	1.087E-01
3	¹³⁵ Cs	1.280E-01	6	¹⁴⁴ Nd	1.226E-01
3	¹³⁶ Cs	1.846E-04	6	¹⁴⁵ Nd	9.209E-02
3	¹³⁷ Cs	3.743E-01	6	¹⁴⁶ Nd	9.046E-02
			6	¹⁴⁷ Nd	1.074E-03
4	⁹⁹ Tc	2.206E-01	6	¹⁴⁸ Nd	4.940E-02
4	^{99m} Tc	8.383E-05	6	¹⁵⁰ Nd	2.339E-02
4	¹⁰⁰ Ru	2.348E-02	6	¹⁴⁷ Pm	2.199E-02
4	¹⁰¹ Ru	2.167E-01	6	¹⁴⁸ Pm	1.838E-04
4	¹⁰² Ru	2.136E-01	6	^{148m} Pm	1.922E-04
4	¹⁰³ Ru	1.315E-02	6	¹⁴⁹ Pm	1.736E-04
4	¹⁰⁴ Ru	1.481E-01	6	¹⁵¹ Pm	3.153E-05
4	¹⁰⁵ Ru	2.051E-05	6	¹⁴⁷ Sm	9.197E-03
4	¹⁰⁶ Ru	4.215E-02	6	¹⁴⁸ Sm	2.021E-02
4	¹⁰³ Rh	1.218E-01	6	¹⁵⁰ Sm	3.982E-02
4	¹⁰⁵ Rh	3.139E-04	6	¹⁵¹ Sm	2.300E-03
4	¹⁰⁶ Rh	3.963E-08	6	¹⁵² Sm	1.799E-02

Table D.1 (continued)

Group	Nuclide	Group mass fraction	Group	Nuclide	Group mass fraction
6	¹⁵³ Sm	1.072E-04	11	¹³⁴ Ba	2.702E-02
6	¹⁵⁴ Sm	4.823E-03	11	¹³⁶ Ba	1.247E-02
6	¹⁵⁶ Sm	1.013E-06	11	¹³⁷ Ba	2.936E-02
6	¹⁵³ Eu	1.320E-02	11	^{137^m} Ba	0.0
6	¹⁵⁴ Eu	4.536E-03	11	¹³⁸ Ba	9.144E-01
6	¹⁵⁵ Eu	1.750E-03	11	¹³⁹ Ba	7.122E-06
6	¹⁵⁶ Eu	3.631E-04	11	¹⁴⁰ Ba	1.670E-02
7	⁸⁸ Sr	3.821E-01	12	¹²¹ Sb	2.539E-01
7	⁸⁹ Sr	3.471E-02	12	¹²³ Sb	3.009E-01
7	⁹⁰ Sr	5.829E-01	12	¹²⁵ Sb	4.313E-01
7	⁹¹ Sr	2.402E-04	12	¹²⁷ Sb	1.304E-02
7	⁹² Sr	3.008E-05	12	¹²⁹ Sb	8.699E-04
8	⁹¹ Zr	2.747E-01	13	⁸³ Kr	7.495E-03
8	⁹² Zr	3.232E-01	13	⁸⁴ Kr	1.943E-02
8	⁹³ Zr	3.633E-01	13	⁸⁵ Kr	4.152E-02
8	⁹⁵ Zr	3.839E-02	13	^{86^m} Kr	2.516E-06
8	⁹⁷ Zr	3.491E-04	13	⁸⁶ Kr	3.332E-02
9	⁹⁵ Nb	9.987E-01	13	⁸⁷ Kr	1.981E-07
9	⁹⁷ Nb	1.247E-03	13	⁸⁸ Kr	2.805E-06
9	^{97^m} Nb	1.535E-05	13	¹³¹ Xe	7.617E-02
10	⁹⁵ Mo	2.025E-01	13	¹³² Xe	1.811E-01
10	⁹⁸ Mo	1.126E-03	13	¹³³ Xe	1.826E-03
10	⁹⁷ Mo	2.485E-01	13	^{133^m} Xe	0.0
10	⁹⁹ Mo	1.126E-03	13	¹³⁶ Xe	3.849E-01
10	¹⁰⁰ Mo	2.916E-01	13	¹³⁴ Xe	2.542E-01
			13	¹³⁵ Xe	5.700E-05

^aRead as 1.946×10^{-3} .

INTERNAL DISTRIBUTION

1. W. S. Aaron	26. C. V. Parks
2. C. W. Alexander	27. D. J. Pruett
3. F. Berrera	28. C. E. Pugh
4. E. C. Beahm	29. C. S. Robinson
5. J. T. Bell	30. J. C. Ryman
6. M. L. Brown	31-33. C. H. Shappert
7. C. H. Byers	34. M. G. Stewart
8. D. O. Campbell	35. R. P. Taleyarkhan
9. J. L. Collins	36. J. S. Tang
10. D. H. Cook	37. J. R. Travis
11. W. A. Gabbard	38. J. S. Watson
12. R. K. Genung	39-43. C. F. Weber
13. S. R. Greene	44. R. M. Westfall
14. P. A. Haas	45. G. E. Whitesides/R. P. Leinius
15. J. R. Hightower	46. R. P. Wichner
16. S. A. Hodge	47. A. Zucker
17. E. K. Johnson	48-49. Laboratory Records Department
18. M. J. Kania	50. Laboratory Records, ORNL-RC
19. T. S. Kress	51. Document Reference Section
20. R. A. Lorenz	52. Central Research Library
21. J. C. Mailen	53. ORNL Patent Section
22. L. Norris	
23. M. F. Osborne	
24. L. J. Ott	
25. G. W. Parker	

EXTERNAL DISTRIBUTION

- 54. Office of Assistant Manager for Energy Research and Development, DOE Field Office, Oak Ridge, P.O. Box 2001, Oak Ridge, TN 37831
- 55. Director, Division of Reactor Safety Research, U.S. Nuclear Regulatory Commission, Washington, DC 20555
- 56-57. Office of Scientific and Technical Information, P.O. Box 2001, Oak Ridge, TN 37831
- 58. Division of Technical Information and Document Control, U.S. Nuclear Regulatory Commission, Washington, DC 20555
- 59. R. Y. Lee, Accident Evaluation Branch, U.S. Nuclear Regulatory Commission, NLN-353, Washington, DC 20555
- 60. R. O. Meyer, Accident Evaluation Branch, U.S. Nuclear Regulatory Commission, NLN-344, Washington, DC 20555
- 61. F. Eltawila, Accident Evaluation Branch, U.S. Nuclear Regulatory Commission, NLN-344, Washington, DC 20555
- 62. T. J. Walker, Accident Evaluation Branch, U.S. Nuclear Regulatory Commission, NLN-344, Washington, DC 20555
- 63. B. S. Burson, Accident Evaluation Branch, U.S. Nuclear Regulatory Commission, NLN-344, Washington, DC 20555

64. K. S. Norwood, 8 Appleford Drive, Abingdon, Oxon OX14 ORA, United Kingdom
65. S. J. Wisbey, B.220, AERE Harwell, Didcot, Oxon OX11 ORA, United Kingdom
66. T. Yamashita, Nuclear Fuel Chemistry Laboratory, Department of Chemistry, Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken, 319-11, Japan

BIBLIOGRAPHIC DATA SHEET

(See instructions on the reverse)

1. REPORT NUMBER
(Assigned by NRC. Add Vol., Supp., Rev.,
and Addendum Numbers, if any.)

NUREG/CR-5808
ORNL/TM-11970

2. TITLE AND SUBTITLE

Calculation of Absorbed Doses to Water Pools in Severe
Accident Sequences

3. DATE REPORT PUBLISHED

MONTH | YEAR
December | 1991

4. FIN OR GRANT NUMBER

B0854

5. AUTHOR(S)

C. F. Weber

6. TYPE OF REPORT

7. PERIOD COVERED (Inclusive Dates)

8. PERFORMING ORGANIZATION - NAME AND ADDRESS (If NRC, provide Division, Office or Region, U.S. Nuclear Regulatory Commission, and mailing address; if contractor, provide name and mailing address.)

Oak Ridge National Laboratory
Oak Ridge, TN 37831-6370

9. SPONSORING ORGANIZATION - NAME AND ADDRESS (If NRC, type "Same as above"; if contractor, provide NRC Division, Office or Region, U.S. Nuclear Regulatory Commission, and mailing address.)

Division of Systems Research
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555

10. SUPPLEMENTARY NOTES

11. ABSTRACT (200 words or less)

A methodology is presented for calculating the radiation dose to a water pool from the decay of uniformly distributed nuclides in that pool. Motivated by the need to accurately model radiolysis reactions of iodine, direct application is made to fission product sources dissolved or suspended in containment sumps or pools during a severe nuclear reactor accident. Two methods of calculating gamma absorption are discussed - one based on point-kernel integration and the other based on Monte Carlo techniques. Using least-squares minimization, the computed results are used to obtain a correlation that relates absorbed dose to source energy and surface-to-volume ratio of the pool. This correlation is applied to most relevant fission product nuclides and used to actually calculate transient sump dose rate in a pressurized-water-reactor (PWR) severe accident sequence.

12. KEY WORDS/DESCRIPTORS (List words or phrases that will assist researchers in locating the report.)

radiation dose
severe accident
iodine radiolysis
fission product transport
water pools
gamma absorption

13. AVAILABILITY STATEMENT

Unlimited

14. SECURITY CLASSIFICATION

(This Page)

Unclassified

(This Report)

Unclassified

15. NUMBER OF PAGES

16. PRICE

THIS DOCUMENT WAS PRINTED USING RECYCLED PAPER