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NUREG-0279

DETERMINATION OF PERFORMANCE CRITERIA FOR HIGH-LEVEL SOLIDIFIED NUCLEAR WASTE

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Lawrence Livermore Laboratory
for
U. S. Nuclear Regulatory Commission

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DETERMINATION OF PERFORMANCE CRITERIA FOR HIGH-LEVEL SOLIDIFIED NUCLEAR WASTE

J. J. Cohen, et al.

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Lawrence Livermore Laboratory
University of California
Livermore, CA 94550

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Office of Nuclear Material Safety and Safeguards
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734 252

FOREWORD BY THE NUCLEAR REGULATORY COMMISSION STAFF

The NRC staff is developing a regulation specifically directed at radioactive waste management. The regulation, when first proposed, will be primarily concerned with the management of high level wastes (HLW). The staff plans to expand the regulation to include the management of low level wastes, decommissioned facilities, and other waste management areas over the next several years.

The HLW sections of the proposed regulation will set forth procedures for licensing HLW management facilities along with performance criteria for the management of HLW The latter will include site suitability criteria, facility design and performance criteria, and performance criteria for solidified HLW.

The NRC has initiated several contractor studies to develop analytical models and to expand the available technical data base for HLW management. The results from such studies will be considered by the NRC in establishing waste management regulations along with data from the literature, information provided by industry and other federal agencies, and comments and information received from the general public. The HLW sections of the waste management regulation are tentatively scheduled to be published for public comment in March 1978. Published along with the proposed regulation will be an Environmental Impact Statement (EIS) which will evaluate the impact of implementing the proposed regulation.

Following the public comment period on the proposed regulation, which could include a rulemaking hearing, the Commission will evaluate the rulemaking record, including comments received; modify the proposed regulation as appropriate based on the rulemaking record; and publish the regulation as an effective rule.

The following draft report was prepared by Lawrence Livermore Laboratory(LLL) under contract to the NRC as a technical data base for the NRC's consideration in establishing HLW solids performance criteria. Included in the study is the development of a systems analysis model which describes the behavior of HLW solids for expected conditions and for postulated accidents as a function the waste properties which affect radionuclide releases. The draft report provides general background information related to the LLL study, describes the work performed by LLL and its subcontractors through early 1977, provides some details regarding the analytical model developed, and sets forth LLL's preliminary findings and recommendations.

A supplement to this draft report is due to be submitted to the NRC in June 1977. That supplement will complete the description of the study and provide the final study results and recommendations. A final report, which will include the information from the draft and supplemental reports, will be published as a NUREG document in the summer of 1977.

The information provided in the final report, including any comments received on the draft or final reports, will be included in the rulemaking record for consideration by the Commission in establishing performance criteria for HLW solids. Persons wishing to comment on either report should mail their comments to the address provided below. Comments received prior to September 30, 1977 will be considered in the development of the proposed regulation. Comments received after that date will be considered in the final rulemaking evaluation. Persons wishing to comment on or provide information pertinent to the development of HLW regulations should mail their comments or information to: Chief, High Level and Transuranic Waste Branch, U. S. Nuclear Regulatory Commission, Washington, D. C., 20555.

734 255

CONTENTS

734 256

734 257

734 258

734 259

734 260

734 261

STUDY CONTRIBUTORS

Study Coordinator

J. J. Cohen, Lawrence Livermore Laboratory (LLL)

Authors and Principal Contributors

Executive Summary - J. J. Cohen (LLL)

Section 1	Introduction - J. J. Cohen (LLL)
Section 2	HLSW Criteria - R. A. Van Konynenburg, J. J. Cohen, and T. Holdsworth (LLL), and A. Sutherland (TASC)
Section 3	Determination of Objective for Waste Management - A. J. Toy, Jr. and J. J. Cohen (LLL), and J. Boegel (U. of A.)
Section 4	High-Level Solid Waste (HLSW) and Radionuclide Release - R. A. Van Konynenburg, A. J. Toy, Jr., and A. Iantuono (LLL) and R. G. Post and M. E. Wacks (U. of A.)
Section 5	Model Development
Section 6	Results - T. Holdsworth (LLL), A. A. Sutherland, Jr., J. L. Center, B. S. Crawford, and B. Ross, The Analytic Sciences Corporation (TASC)
Section 7	Discussion - J. J. Cohen (LLL)
Appendix A:	Biologically Significant Radionuclides in High-Level Waste - A. J. Toy, Jr., (LLL)
Appendix B:	Dose Conversion Factors - H. A. Tewes and Y. C. Ng (LLL)
Appendix C	Hydrologic Model - T. Holdsworth (LLL) and J. L. Center and B. Ross (TASC)
Appendix D	Meteorology and Demography - K. R. Petersen (LLL)
Appendix E	History - N. Hilberry and R. G. Post, University of Arizona (U. of A.)

734 265

Editors

W. O. Shearer

F. N. Weld

W. T. Wilfong

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Determination of Performance Criteria for High-Level Solidified Nuclear Waste

EXECUTIVE SUMMARY

Introduction

Present federal regulations require that high-level radioactive waste liquid resulting from the reprocessing of irradiated nuclear reactor fuel be converted to a dry solid with sufficient thermal, radiological, and chemical stability such that the pressure in the canister encapsulating the waste would not exceed the safe operating pressure of the container for a period of at least 90 days following receipt of the waste at a federal repository. Insofar as the mechanical, radiological, and chemical properties of the dry solid waste form chosen could potentially affect the public health and safety during the handling, storage, transportation, and disposal of the wastes, the Nuclear Regulatory Commission (NRC) requested Lawrence Livermore Laboratory to determine what further constraints, if any, should be placed on the properties of high-level solid wastes. It is the purpose of this study to identify and recommend performance orientated criteria and/or standards which, if applied, would reduce the risk to the environment and to the public health and safety due to solid HLW management operations including final disposal. The study will result in a two-part technical report describing the study and results and in an Environmental Impact Statement which will assess the potential impacts of a rule making action to establish performance criteria for solidified high-level waste. This document (Part 1 of the technical report) describes the study approach, the calculational model developed, factors considered in the evaluation, and the preliminary results obtained to date. Part 2, which will contain the technical basis for the criteria recommended, is scheduled for completion in July 1977. A draft Environmental Impact Statement is scheduled for completion in August 1977.

Discussion

The study was conducted using a systems analysis approach which considered the various situations which could lead to potential releases of radioactive materials during the handling, storage, transportation, and disposal of high-level solid wastes (HLSW). A systems analysis approach was chosen to facilitate the identification of potential release points and mechanisms which would be limiting in establishing criteria, i.e., to identify potential points of release in the management cycle most sensitive to the waste form characteristics.

The study required the use of calculational models to predict potential release mechanisms and probabilities, radionuclide transport and dispersive mechanisms, and to evaluate pathways and doses to man.

A systems model has been developed which considers the various operations related to the management of HLSW. Failure modes, probabilities of failure, and the severity of consequences due to failure were determined for each operational step or phase. Operational sequences can be categorized as: (1) operations at the fuel reprocessing plant (FRP) where the HLSW is formed, containerized, and stored; (2) HLSW transportation operations during shipment from the FRP to the Federal Waste Repository (FWR), and (3) events at the FWR, including pre- and post-emplacement phenomena.

Event-tree methodology was used to determine potential failure mechanisms. Evaluations of failure consequences and probabilities were based on available data and the best judgement of the authors and other researchers. Population doses to man were based on assumed demographic models for the FRP, transportation routes, and FWR.

The overall model was derived primarily from existing models appropriately modified to provide the analytical features required for this study. Care was taken to make the model consistent with NRC staff models where appropriate. Calculational capabilities include the calculation of doses for each pathway, risks for each pathway (product of the probability times the dose), and the expected value dose for a given waste management operation (summation of the risks from the various potential pathways resulting from that operation). Where appropriate, the release pathways consider the mitigating effects of engineered safety features (e.g., fuel reprocessing plant related accidents, shipping cask accidents, etc.) and radionuclide retention by natural processes in the geologic media.

For any given pathway, one of three mechanisms will control the release of radioactive materials; volatilzation, dispersion of particulates, or leaching by

734 268

water. The performance criteria will be structured to control each of these mechanisms based on an analysis of the pathways each affects, the state of technology for controlling each mechanism, and balancing the cost of control against the benefits in terms of reduced risks to individuals and populations. Although the criteria will be derived from analytical pathway studies for various accident scenarios, the criteria will be set forth in terms of impact resistance, thermal stability, and resistance to leaching by water in order to facilitate the proof testing of potential candidate waste forms.

Research has been conducted for several years to develop methods for solidifying high-level liquid nuclear wastes. The objective of this research has been to produce stable solid waste forms in order to assure radionuclide immobility, thereby further isolating emplaced wastes from the biosphere. The characteristics of these various waste forms are summarized in Table 1.

The properties of the waste forms in Table 1 along with the information available regarding the cost of producing each waste form will be factored into the cost analysis used to derive the criteria.

Conclusions

The results obtained to date indicate that the pre-emplacement waste environs may be more limiting in establishing the waste-matrix performance criteria than the post-emplacement environs, considering both normal conditions and potential accident conditions. These results are based upon waste emplacement in a stable geologic repository but do not rely upon the repositories remaining intact over the potentially hazardous lifetime of the waste.

Our preliminary evaluations show the transportation phase of the waste management system to be potentially the most limiting due to both the variety of possible disruptive interactions and to demographic factors. These concerns may be partially, if not totally, mitigated by the large degree of protection afforded by the casks* in which the wastes will be transported. Studies in

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* Appendix F to 10 CFR Part 50 requires HLW to be transported in casks meeting the requirements set forth in 10 CFR Part 71.

734 269

progress, which take into consideration the effect of the cask, will evaluate the potential for release during the transportation phase.

A recommended format for specifying performance criteria orientated towards controlling radionuclide releases due to volatilization, particulate dispersion, and leaching has been derived and is given in Chapter 2 of the report. Numerical standards to be incorporated into the criteria will be derived in the subsequent phases of this study and published in part 2 of the technical report.

It should be noted that the results and conclusions reached to date are preliminary and are being re-examined as a part of the current studies which will be discussed in Part 2 of this report. Part 2 will include the technical bases for the parameters used in the analyses along with a sensitivity analysis which will show the relationship between the parameters considered, their uncertainty, and the results obtained.

Introduction

1.1 THE PROBLEM

Existing NRC regulations (Appendix F to 10 CFR Part 50) define high-level liquid radioactive wastes as: "those aqueous wastes resulting from the operation of the first cycle solvent extraction system, or equivalent and the concentrated wastes from subsequent extraction cycles, or equivalent, in a facility for reprocessing irradiated reactor fuels." These high level liquid wastes contain over 99% of the radioactivity produced in the nuclear fuel cycle. Appendix F to 10 CFR Part 50 further states:

> "A fuel reprocessing plant's inventory of high-level liquid radioactive wastes will be limited to that produced in the prior 5 years . . . High level liquid radioactive wastes shall be converted to a dry solid as required to comply with this inventory limitation, and placed in a sealed container prior to transfer to a Federal repository in a shipping cask meeting the requirements of 10 CFR Part 71. The dry solid shall be chemically, thermally, and radiolytically stable to the extent that the equilibrium pressure in the sealed container shall not exceed the safe operating pressure for that container during the period from canning through a minimum of 90 days after receipt (transfer of physical custody) at the Federal repository. All of these high-level radioactive wastes shall be transferred to a Federal repository no later than 10 years following separation of fission products from the irradiated fuel."

Although conversion of the liquid HLW to a solid form as required in Appendix F to 10 CFR Part 50 decreases the potential for radionuclide mobility, it appears reasonable that performance requirements for the high-level solid waste (HLSW) matrix and its canister beyond those included in Appendix F may be needed to reduce potential radionuclide releases to as low as reasonably achievable levels. This study was initiated to evaluate the need for establishing additional performance criteria for HLSW and to identify, if necessary, a new set of performance criteria designed to minimize risk to the public health and safety and the environment during all activities involved with management and ultimate disposal of the wastes. Since isolation in deep geologic media appears to be the most likely disposal method for high level wastes during the next decade, this disposal mode was assumed for purposes of this study.

1.2 GENERAL BACKGROUND

For several years, research has been conducted to develop methods for solidification of high-level liquid wastes. The objective of this effort has been to produce stable solid-waste forms in order to assure their immobility and, therefore, their isolation from the biosphere. Efforts to improve HLSW forms are continuing. However, it is difficult to judge the adequacy of any given type in the absence of objective criteria. Because considerable quantities of both military and commercial nuclear waste continue to accumulate, it is necessary to determine these performance criteria so that fuel reprocessors can plan their solidification operations accordingly.

1.3 PROJECTION

The 1976 National Energy Outlook of the Federal Energy Administration (FEA, 1976) predicts that nuclear power will expand from 8.6% of the total electrical power generated in the United States in 1975 to 26% in 1985. The capacity of installed nuclear generators is estimated to rise from about 42 GW in the U. S. at present (September 1976) (Nuclear News, 1976) to 160 GW by the end of 1985 and to 625 GW by the end of the year 2000 (Bown, 1976). The amounts of fuel to be reprocessed are estimated (Blomeke and Kee, 1976) to reach 3100 tonnes/yr in 1985 and nearly 12,000 tonnes/yr in the year 2000. The accumulated radioactivity of the HLSW at the FWR would be over 3500 MCi by 1995 and nearly 28 GCi by the year 2010 (see Fig. 1-1). These estimates assume that the wastes be shipped to the FWR 10 years after generation.

In view of the anticipated growth in nuclear power, the reprocessors, need for more detailed criteria, and the increased public concern about the plans for management of radioactive wastes, the NRC determined the necessity of a systematic study to derive detailed performance criteria.

1.4 METHODOLOGY

To evaluate the performance requirements for HLSW forms it is necessary to consider the potential for release of radioactive components of the waste to the biosphere from formation of the solid through disposal and decay to safe levels. A systems analysis model was developed for this purpose. The model considers potential releases for both routine and accident situations for all phases of storage, handling, transportation, and disposal in deep geologic media.

Event trees were constructed for each waste management operation to identify potential release mechanisms. Published data for failure probabilities were used wherever available. For the initial phase of the study, releases were normalized to Ci/MWe - yr and individual and population doses were calculated on an expected value basis, also normalized to per MWe - yr. Subsequent analyses will be conducted or a "per event" rather than "expected value" basis assuming nominal canister sizes and nominal radionuclide inventories in storage systems. transportation casks, and HLW repositories. The "per event" analysis will evaluate potential doses to individuals for all operational phases. Population doses will be considered primarily during the pre-emplacement phase since uncertainties in future population locations, sizes, and customs make dose calculations for future populations of questionable value.

The expected value calculations provided a means of identifying the potential "pinch points" for setting criteria on a systems basis. Although they provide information on integrated risks (expected value) they do not indicate the consequences of individual contributing events. The recommended values for the HLSW performance criteria (standards) will consider the reduction of both the integrated risks and single event risks to the public to "as low as reasonable achievable" levels.

1.5 DISCUSSION

Chapters 4 and 5 discuss potential release mechanisms and the calculational model, respectively, and Chapter 6 provides the numerical results obtained to date. Although the determination of the values to be recommended for use by the NRC must await the conclusion of the second part of this study, preliminary results indicate that the pre-emplacement waste environs may be more limiting in establishing performance requirements for the HLSW than the post-emplacement environs. This conclusion is based on an analysis of potential repository failure mechanismsand resultant doses to individuals, considering transport by groundwater to be the mechanism of primary concern. (Waste volatility and particulate dispersion are not significant in the post-emplacement phase for deep geological formations.)

The criteria (format proposed in Chapter 2) will be designed to reduce the potential for radionuclide releases due to volatilization, particulate dispersion, and leaching. The criteria will set standards for resistance to chemical, mechanical, and thermal insults; the three broad categories of initiating events most likely to lead to radionuclide releases. For pre-emplacement and retrievable post-emplacement conditions the effectiveness of both the waste matrix and the canister for controlling potential releases will be considered.

1.6 REFERENCES

J. O. Blomeke and C. W. Kee, "Projections of Wastes to be Generated," in Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle, Denver, Colorado, July 11-16, 1976 (CONF-76-0701) (National Technical Information Service, Springfield, Va., 1976), p. 96.

R. W. Bown, U. S. ERDA Office of Planning and Analysis, communication of April 1976, cited in Blomeke and Kee, 1976.

U. S. Federal Energy Administration, 1976 National Energy Outlook (U. S. Government Printing Office, Washington, D. C., 1976), No. 941-018-00097-6.

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"The World List of Nuclear Power Plants," Nuclear News, 19 (10), 66 (1976).

734 274

Waste Solids

Property	Units	Salt cake	Sprav calcine	Pot calcine	Rotary kiln calcine	Fluidized bed calcine	Super calcine	Coated particles
Solution rate	$\frac{mg}{m^2 sec}$	10^5 to 10^6	10 to 100	10 to 100	10 to 100	10 to 100	10^{-5} to .016	10^{-6} to 0.01
Corrosion to clad material	nm/sec	0 to 10	0 to 10	0 to 10	0 to 10	0 to 10	0 to 10	0 to 10
Residual nitrate and water	%	0.20 to 30	≤4.0	≤0.05	≤4.0	≤.03	0.005 to 0.05	0.005 to 0.05
Maximum processing temperature	K	310 to 720	970	695	870 to 1070	670 to 1070	1170 to 1370	1670
Ruthenium volatilized at processing	%	≤1.0	≤1.0	5 to 30	≤1.0	Insignificant	≤7.0	≤10.0
Volatility	n.a.	1000 K all Ru and Cs	1200 K all Ru and Cs	1200 K all Ru and Cs	1200 K all Ru and Cs	1200 K all Ru and Cs	1500 K all Ru and Cs	1670 K all Ru and Cs
Specific volume	$\frac{m^3}{MgU}$	0.05 to 1.0	.03 to .06	0.044 to 0.058	0.045	0.032 to 0.040	0.070	0.130
Wt% fission product oxides	Max.	≤80%	≤100%	≤90%	≤100%	≤100%	50%	≤80%
	Typical	≤80%	≤100%	≤90%	≤100%	≤100%	20%	45%
Specific area	$\frac{m^2}{Kg}$	100 to 5000	10 000 to 20 000	100 to 5000	100 to 5000	100 to 5000	10 000 to 20 000	100 to 5000
Form	N/A	Monolithic	Powder	Powder or scale	Granular	Granular	Powder	Beads
Structural quality	N/A	Crumbly	Soft and crumbly	Soft and crumbly	Soft and crumbly	Soft and crumbly	Soft and crumbly	Hard
Porosity	%	0	30 to 75	40 to 85	70 to 80	45 to 80	40 to 80	≤20.0
Density	$\frac{Kg}{m^3}$	1700	1000 to 2400	1100 to 1400	1000 to 1300	2000 to 2400	4000	1700
Coefficient of linear expansion	(K-1) $\times 10^6$	0	8.3	8.3	8.3	8.3	8.3	4.5
Thermal conductivity	$\frac{W}{mK}$	0.1 to 0.5	0 2	0.35 to 1.0	0.2	0.2 to 0.3	0.6	17
Heat capacity	$\frac{J}{KgK}$	500 to 800	650	650	650	650	670	680
Liquidus temperature	K	520	1670	1670	1670	1670	1670	2570
Transition temperature	K	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

Table 1 (Cont.)

Waste Solids							
Phosphate glass	Borosilicate glass	Sintered process	Glass ceramic	Nepheline syenite	Metal matrix	Ion exchange	Rock melt
10^{-5} to 0.68	10^{-5} to 0.01	10^{-5} to 0.001	10^{-5} to 10^{-3}	1 × 10^{-5}	10^{-5} to 10^{-2}	0.001 to 0.01	10^{-7} to 0.001
0 to 10	0 to 10	0 to 10	0 to 10	0 to 10	0 to 10	0 to 10	n.a.
0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	≤10.0
1170	1270 to 1670	1235 to 1370	1270	1620	1470	1670	1000
3 to 15	≤2.0	≤10.0	≤10.0	≤10.0	≤10.0	≤10.0	≤5.0
<1500 K all Ru and Cs	<1500 K all Ru and Cs	<1370 all Ru and Cs	<1325 K all Ru and Cs	<1620 K all Ru and Cs	<1525 all Ru and Cs	<1670 K all Ru and Cs	n.a.
0.036 to 0.078	0.04 to 0.1	0.03 to 0.055	0.077	0.08	0.08	0.050	4 to 40
≤25%	≤50%	≤50%	30%	50%	≤65%	≤80%	10%
20%	20-35%	45%	25%	20%	33%	25%	5%
0.005 to 0.05	0.005 to 0.05	0.1 to 1.0	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.05 to 0.05	n.a.
Fractured Monolith	Fractured Monolith	Pellets	Monolithic	Monolithic	Monolithic	Mono lithic	*In situ* melt
Very hard and brittle	Very hard and brittle	Very hard	Hard and tough	Very hard and brittle	Hard and ductile	Hard and brittle	Hard
≤5.0	≤1.0	8 to 25	≤10.0	≤5.0	≤10.0	≤3.0	≤5.0
2700 to 3000	3000 to 3600	2400 to 3300	2850	3000	2400 to 5500	≤4500	2700
8 to 10	8 to 10	8.0	9.0 to 1.3	10	29	4.2	9.0 to 4.0 × 10^{-6}
0.8 to 1.3	0.9 to 1.3	0.7	2.2	1.2	5 to 35	1.2	1.0 to 1.5
1100 to 1200	750	1100 to 1200	1100 to 1200	1100 to 1200	350	550	1000 to 1600
820 to 1020	800 to 1500	970 to 1500	1070	850	600	2290	1320
770	870 to 970	n.a.	n.a.	900	n.a.	n.a.	n.a.

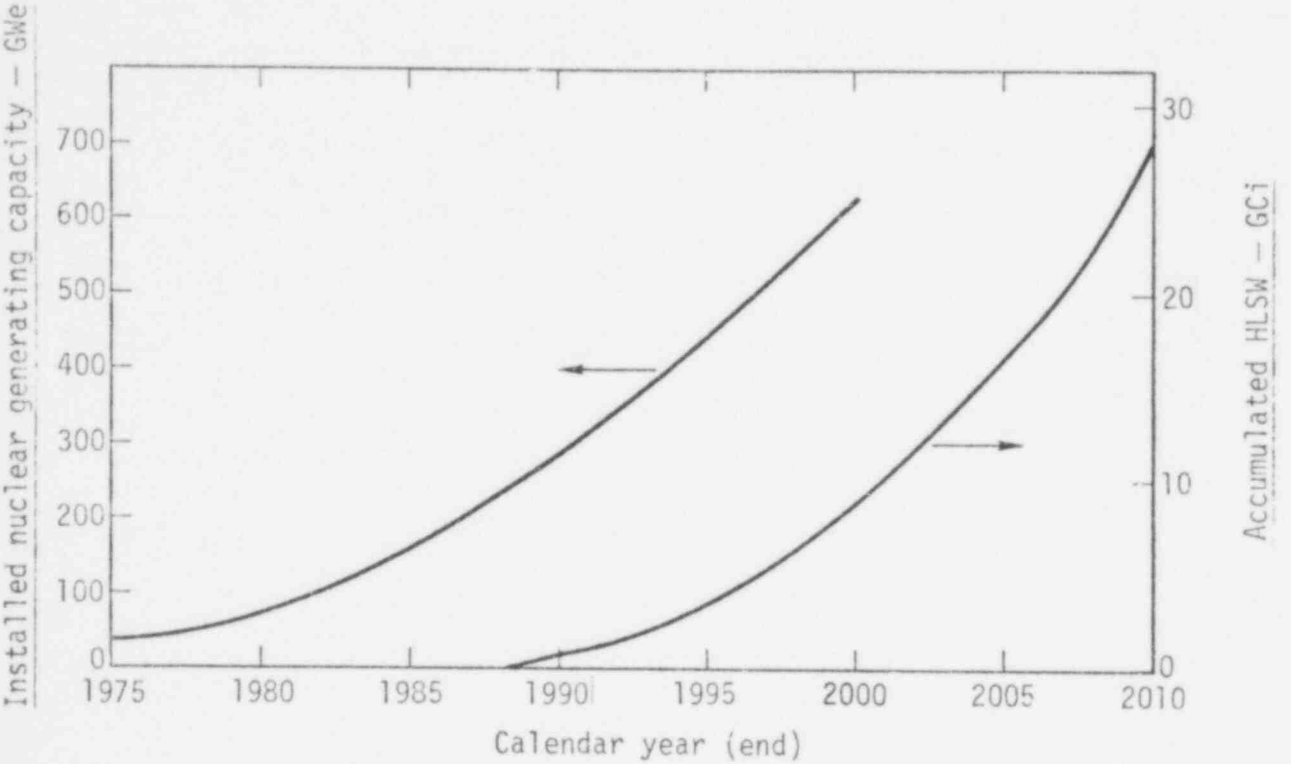

Fig. 1-1. Projected U.S. nuclear generating capacity and accumulated HLSW at the federal repository (data from Blomeke and Kee, 1976).

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2 PROPOSED FORMAT FOR HIGH-LEVEL SOLID WASTE PERFORMANCE CRITERIA

In the development of performance criteria for HLSW, consideration must be given to population doses and individual doses (both occupational and non-occupational) for the total waste-management system.

The first phase of the effort has dealt primarily with expected values of population dose. The second phase, now in progress, emphasizes individual doses in accident situations along with population doses. Relationships between HLSW properties and radioactive releases for particular accident situations will be derived from this analysis. This will lead to formulation of cost-benefit relationships which will provide insight into the dose levels that are "as low as reasonably achievable." When this phase is completed, quantitative values for HLSW performance will be recommended for use in the proposed criteria.

The following are potential formats for expressing the HLSW performance criteria. The principal release mechanisms which the performance criteria will be designed to control are volatilization of gases, particulate dispersion, and leaching by water. Volatilization requires a thermal driving force, particulate dispersion could be initiated either thermally or mechanically, and leaching requires immersion in water. The proposed criteria are structured to control these release mechanisms by placing performance limits on the response of the HLSW and/or the HLSW/canister combination to thermal, mechanical, or chemical forces. Work presently underway will develop performance tests which may be used to determine compliance with the performance criteria. Except where noted, the criteria apply to the combination of the HLSW matrix and the canister in which it is sealed.

For these criteria, the HLSW system shall consist of the combination of the HLSW and its canister.[1]

2.1 LOSS OF COOLANT CRITERIA

Loss of Coolant - Volatility Criterion - The HLSW system shall not release to air as volatile gases a fraction of the activity of the radio-isotopes of cesium, ruthenium, and tellurium greater than ________________ _______ ____ under the thermal conditions that would be present if the interim storage cooling were lost.

Loss of Coolant - Particulate Dispersibility Criterion - The HLSW system shall not release to air a fraction of the total activity greater than ____________________ _________ in the form of particles smaller than 10 micrometers (aerodynamic diameter) under the thermal conditions that would be present if the interim storage coolant were lost.

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[1] Numerical values will result from present work scheduled for completion in mid-1977.

2.2 PRESSURIZATION CRITERIA

The combination of the HLSW form and its canister shall have properties and design such that the equilibrium pressure in the sealed canister will not exceed the safe operating pressure for that canister during the period from sealing through a time after receipt (transfer of physical custody) at the FWR sufficient to insure that emplacement and backfilling can be completed. These properties and design must take account of radiation effects on the HLSW form and the temperatures it will normally experience during the period of time from sealing to emplacement and backfilling.

2.3 TRANSPORTATION CRITERIA

For these criteria, the HLSW system shall consist of the combination of the HLSW, its canister, and its transportation cask. These criteria shall apply at the time the HLSW system is shipped.

Impact - Particulate Dispersibility Criterion - The HLSW system shall not release to air a fraction of the total activity greater than ________ ________ in the form of particulates smaller than 10 micrometers (aerodynamic diameter) when subjected to a free drop through a distance producing an impact velocity of ___ mph (see Table E-1) onto a flat, essentially unyielding horizontal surface, striking the surface in an orientation for which maximum damage is expected.

Table E-1. Environmental and accident conditions for transportation criteria.

	Truck	Train
Impact - particulate dispersibility		
Impact velocity (mph)	60	50
Puncture - particulate dispersibility		
Impact velocity (mph)	40	40
Fire - particulate dispersibility		
Temperature (°F)	2000	2000
Time (min)	80	45
Fire - volatility		
Temperature (°F)	2000	2000
Time (min)	80	240
Impact - dissolution		
Impact velocity (mph)	60	50
Immersion depth (ft)	1	1
Immersion time (hr)	24	24
Puncture - dissolution		
Impact velocity (mph)	40	40
Immersion depth (ft)	1	1
Immersion time (hr)	24	24
Fire - dissolution		
Temperature (°F)	2000	2000
Time (min)	80	240
Immersion depth (ft)	1	1
Immersion time (hr)	24	24

Puncture - Particulate Dispersibility Criterion - The HLSW system shall not release to air a fraction of the total activity greater than ________ ______ in the form of particulates smaller than 10 micrometers (aerodynamic diameter) when subjected to a free drop through a distance producing an impact velocity of __ mph (see Table E-1) striking in an orientation for which maximum damage is expected, the top end of a vertical, cylindrical, mild steel bar mounted on an essentially unyielding horizontal surface. The bar shall have a 6-inch diameter, with the top horizontal and its edge rounded to a radius of not more than 0.25 inch, and it shall be of such a length as to cause maximum damage to the system, but not less than 3 inches. The long axis of the bar shall be perpendicular to the unyielding horizontal surface.

Fire - Particulate Dispersibility Criterion - The HLSW system shall not release to air a fraction of the total activity greater than ________ ________ in the form of particulates smaller than 10 micrometers (aerodynamic diameter) when subjected to a thermal environment in which the heat input to the system and the air velocity are no less than those that would result from exposure to a fire under conditions specified in Table E-1.

Fire Volatility Criterion - the HLSW system shall not release to air as volatile gases a fraction of the activity of the radionuclides of cesium, ruthenium, and tellurium greater than ________ _____ when subjected to a thermal environment in which the heat input to the system and the air velocity are no less than those that would result from exposure to a fire under conditions specified in Table E-1.

<u>Impact - Dissolution Criterion</u> - The HLSW system shall not release to water a fraction of the total radioactivity greater than ________ ________ during an immersion that follows a free drop. The free drop shall be made through a distance producing an impact velocity of ___ mph (see Table E-1) onto a flat, essentially unyielding horizontal surface, striking the surface in an orientation for which maximum damage is expected. The immersion shall be performed in water having initial concentrations of dissolved ions, pH, oxidation potential (E_h), flow rate, and temperature that are substantially equivalent to those of typical U.S. river water, under conditions specified in Table E-1.

<u>Puncture - Dissolution Criterion</u> - The HLSW system shall not release to water a fraction of the total radioactivity greater than ____________ __________ during an immersion that follows a free drop. The free drop shall be made through a distance producing an impact velocity of ___ mph (see Table E-1), striking in an orientation for which maximum damage is expected, the top end of a vertical, cylindrical, mild steel bar mounted on an essentially unyielding horizontal surface. The bar shall have a 6-inch diameter, with the top horizontal and its edge rounded to a radius of not more than 0.25 inch, and it shall be of such a length as to cause

maximum damage to the system, but not less than 8 inches. The long axis of the bar shall be perpendicular to the unyielding horizontal surface. The immersion shall be performed in water having initial concentrations of dissolved ions, pH, oxidation potential (E_h), flow rate, and temperature that are substantially equivalent to those of typical U.S. river water, under conditions specified in Table E-1.

Fire - Dissolution Criterion - The HLSW system shall not release to water a fraction of the total radioactivity greater than ________ ________ during an immersion that follows subjection to a thermal environment. The thermal environment shall be such that the heat input to the system is no less than that which would result from exposure to a fire under conditions specified in Table E-1. The immersion shall be performed in water having initial concentrations of dissolved ions, pH, oxidation potential (E_h), flow rate, and temperature that are substantially equivalent to those of typical U.S. river water, under conditions specified in Table E-1. The immersion shall take place before significant cooling of the HLSW system has occurred.

2.4 POST EMPLACEMENT SOLUTION CRITERIA

The analysis discussed in Sec. 6 indicates that under assumed post-emplacement conditions, the rate of waste dissolution in groundwater is not a significant factor in determining dose to man. Only if some other factor such as a requirement for retrievability were incorporated in the analysis, would dissolution in groundwater become a factor to be considered. For this reason, no criteria are recommended.

2.5 HLSW FORM PERFORMANCE CRITERIA

This group applies to the waste form alone. These criteria will be quantitatively defined at the conclusion of work now in progress, which is expected in mid-1977.

2.6 DEFINITION OF FORM

The HLSW form shall be a solid.

HLSW Post-Impact Respirability Criterion - A representative sample of the HLSW form must consist of no more than a specified weight fraction of dispersible particles smaller than 10 micrometers (aerodynamic diameter) after subjection to an impact induced by a falling weight that has a specified kinetic energy per unit mass of the sample. The weight fraction and kinetic energy shall be chosen such that the impact-, puncture-, and fire-particulate dispersibility criteria given above will be satisifed. The expected behavior of the canister and cask must also be taken into account.

HLSW Leachability Criterion - A representative sample of the HLSW form must not release to a flowing water solution by dissolution more than specified fractions of the radioactivity of the nuclides present under specified

734 285

conditions of sample size; thermal, irradiation, and impact pretreatment; flow rate; initial concentrations of dissolved ions; initial pH; initial oxidation potential (E_h); temperature; and leach time. These fractions and conditions shall be chosen such that the impact-, puncture-, and fire-dissolution criteria given above will be satisfied. The expected behavior of the canister and cask must also be taken into account.

HLSW Volatility Criterion - A representative sample of the HLSW form must not release to moving air as gases more than specified fractions of the radioactivity of the nuclides present under specified conditions of sample size; thermal, irradiation, and impact pretreatment; container material, shape, and size; relative humidity; temperature; pressure; flow rate; and volatilization time. These fractions and conditions shall be chosen such that the less of coolant-volatility and fire-volatility criteria given above will be satisfied. The expected behavior of the canister and cask must also be taken into account.

3 DETERMINATION OF OBJECTIVE FOR WASTE MANAGEMENT

3.1 INTRODUCTION

It is necessary to establish objectives for the overall waste-management system before determining the acceptability of a particular HLSW form. This section reports on the effort to provide such functions relevant to environmental concerns. Various postulated waste-management systems were examined to determine their environmental impacts and to rank the relative importance of those impacts. This examination found that radiation dose to man was the most significant and probably the overriding impact. Therefore, this section describes methods of determining objectives for waste management with respect to the resulting dose to man.

The search for an objective function began with a reading of federal regulations and the output of various standard setting councils and committees. Ample guidance was found on maximum allowable doses to individuals, but there was neither guidance on allowable doses to large populations nor criteria by which waste-management systems could be compared or optimized. Several ways, in addition to maximum allowable dose to individuals, were postulated by which the systems could be evaluated. One of these, the man•rem per megawatt electric•year provides a measure of system performance that balances the population dose commitment with the concurrent benefit. The combination of limiting doses to maximally affected individuals and assuring that the risks attributed to integrated population doses are compared to the benefits derived from that which led to the dose, appears to offer a viable method of comparing waste-management systems.

734 287

The following subsections outline the applicable federal laws, assess the need for further study, and describe some of the possible criteria to be used in determining objectives for a waste-management system.

3.2 EXISTING REGULATIONS AND GUIDELINES FOR WASTE MANAGEMENT

Federal regulations on the licensing and operation of waste-management programs are presented below. State regulations concerning waste management have not been examined because of the limited scope of waste-management operations that states may regulate. Laws of other countries have not been examined. The licensing of waste-management programs is governed by the following parts of title 10 of the *Code of Federal Regulations* (CFR):

- Part 51, "Licensing and Regulatory Policy and Procedures for Environmental Protection," sets forth the policy and procedures for the preparation and processing of environmental impact statements and related documents. Part 51 requires that environmental impact statements be filed for FRPs and waste-disposal operations (for certain quantities) prior to their licensing in accordance with the National Environmental Policy Act of 1969 (NEPA).

- Part 20, Secs. 301-5, "Standards for Protection Against Radiation — Waste Disposal," governs the disposal, by burial or by discharge to a sanitary sewer or water system, of small quantities of radioactive material. Section 302 requires that land burial be on property owned by either the state or federal government. It also requires that pertinent information on the nature of the environment be included in the environmental impact statement and that procedures be followed to minimize unexpected exposures.

- Part 30, "Rules of General Applicability to Licensing of Byproduct Material," defines the maximum quantities and concentrations of by-product

734 288

material that may be handled without a license. "By-product material" means any radioactive material (except special nuclear material) yielded in or made radioactive by exposure to radiation, incidental to the production or use of special nuclear material.

- Part 40, "Licensing of Source Material," provides that no person without a license shall own, use, or handle source material after mining. "Source material" means: (1) uranium, thorium, or any combination thereof in any physical or chemical form; or (2) ores that contain 0.05 wt% or more of uranium, thorium, or any combination thereof. Source material does not include special nuclear material.

- Part 50, Appendix F, "Licensing of Production and Utilization Facilities, Policy Related to the Siting of Fuel Reprocessing Plants and Related Waste Management Facilities," requires that high-level waste be solidified within five years and shipped to a FWR on federally owned and controlled land within ten years.

- Part 70, "Special Nuclear Material," establishes licensing requirements for special nuclear materials. "Special nuclear material" means: (1) plutonium, uranium-233, uranium enriched in the isotope 233 or in the isotope 235, and any other material that NRC determines to be special material, but not source material; or (2) any material artificially enriched in any of the foregoing, but not source material.

Operations of waste-management systems are governed by the following:

- Title 10, CFR, Part 20, "Standards for Protection Against Radiation," details maximum permissible radiation doses, maximum permissible radionuclide air concentrations, and maximum permissible radionuclide water concentrations that apply to both occupational and public exposure.

- Title 10, CFR, Part 71 "Packaging of Radioactive Material for Transport and Transportation of Radioactive Material Under Certain Conditions,"

734 289

describes regulations for the handling and shipping of licensed material. The requirements of this part are to be applied in addition to the requirements of Title 49, CFR.

- ERDA Manual, Ch. 0524, applies the radiation-dose and air- and water-concentration limits of Title 10, CFR, Part 20, to contractors who operate ERDA-owned or -controlled facilities.

- Title 49, CFR, Subtitle B, Ch. 1, Parts 171-79, "Hazardous Materials Regulations Board, Department of Transportation," covers preparation and shipping of hazardous materials, including radioactive materials.

- ERDA Manual, Ch. 0511, "Radioactive Waste Management," applies to contractors who operate ERDA-owned or -controlled facilities. Included in this chapter is the provision that solid waste containing significant uranium-233 or transuranic contamination greater than 10 nanocuries per gram be segregated and placed in retrievable storage.

- Title 40, CFR, Part 141, "Interim Primary Drinking Water Regulations," provides concentration and dose limits in drinking water for man-made and natural nuclides. Although the burden of compliance for this regulation is on the supplier of drinking water, we feel that cognizance must be taken of these limits in evaluating discharges to water.

- Title 40, CFR, Part 190, "Proposed Environmental Radiation Protection Standards for Nuclear Power Operations," defines 25 man•rem per year as the maximum individual dose equivalent allowed from the uranium fuel cycle and sets release limits in curies per gigawatt•year for some long-lived nuclides. Although not applicable to waste disposal sites, this proposed regulation is mentioned for its unique measurement units — dose due to the fuel cycle, not a specific facility, and release per unit energy, not per unit volume.

734 290

3.3 ASSESSMENT OF REGULATIONS

Title 10, CFR, Part 20, Sec. 302 requires several specific environmental considerations in the choice of a waste-disposal site. Part 51 requires an environmental statement as a licensing condition for waste-management operations. Attention has been focused on what is felt to be the most important and probably overriding environmental factor: the dose to man resulting from radioactive waste management. Therefore, the following discussions deal only with dose to man. Part 20 requires that operations at a facility be such that radioactive releases and radiation exposures be as low as reasonably achievable (ALARA) and limited to less than 0.5 rem per year to any individual in an unrestricted area. The regulation requires that ALARA evaluations balance technology for reducing releases against considerations of economy and public health and safety. If one accepts the linear, no-threshold hypothesis for the biological effects of radiation (National Academy of Sciences — National Research Council, 1972), then in evaluating radiological impacts on public health both the man•rem and the dose to a maximally exposed individual should be considered. Such an approach has been taken in Title 10, CFR, Part 50, Appendix I.

It is assumed at this time that population doses from waste management will be very small if the waste-management operations proceed according to design. It is likely that population exposures due to potential accidents, although small, will be of greater significance than potential exposures from normal operations. No regulations have been found that apply to waste management, giving guidance to acceptable individual or population exposures under accident conditions. Title 10, CFR, Part 100, "Reactor Site Criteria," gives some guidance for subject reactors by setting maximum calculated doses for various places outside the reactor. These doses can be used for evaluating site suitability under maximum-dose accident conditions.

3.4 POSSIBLE CRITERIA

It is assumed that all the presently considered options for waste management will result in radiation dose to man within the limits of the existing regulations. What follows is a discussion of some possible criteria by which one might compare alternative methods and strategies to evaluate relative merit or to decide what is ALARA. The reader is again cautioned that the discussion is limited to considerations of radiation dose to man.

3.4.1 Expected Value

The expected value of an event is the probability of its occurrence multiplied by the value of its outcome. For example, a hypothetical accident with an expected frequency of once every five years and an expected population dose of 10 man•rem has an expected value of 2 man•rem per year. A hypothetical waste-management system, whose accidents can be described by a probability density function p(x) (in yr^{-1}) and a consequence function q(x) (in man•rem), has an expected value for accidents of

$$D = \int^{x} p(x)q(x)dx \text{ (in man•rem/yr).}$$

Use of expected value allows direct comparison of two systems with different probability functions (e.g., system A, which might produce several small accidents, vs system B, which might produce a few large accidents). Expected value also allows accidental doses to be compared with and combined with doses from normal operation in the evaluation of a particular system's total dose-to-man impact.

734 292

3.4.2 Comparison with Natural Hazards

Comparison of the dose to man from waste-management activities with that from natural hazards could be a way of measuring acceptability of a system and a way of placing radiation risks in perspective with other risks. For example:

- Compare the toxicity of radioactive waste with the toxicity of naturally occurring ores (uranium, mercury, lead, chrome, etc.).
- Compare radiation doses with natural radioactive background.
- Compare the equivalent hazard from effluents of waste management with the hazards of permissible levels for nonradioactive materials (as set in drinking water standards and national ambient air quality standards).
- Make a study of waste-management risks similar to the Rasmussen report on reactor-related risks (U.S. Atomic Energy Commission, 1974) with graphs of accident frequency versus severity, thereby comparing the risks of waste-management activities with those of:
  - Man-caused accidents (fire, collision, etc.),
  - Natural accidents (earthquake, tornado, flood, meteorite, etc.).

3.4.3 Comparison with Benefit and Cost

Title 10, CFR, Part 20 requires doses to be ALARA, a standard determined by balancing doses against technology, economics of improvement, and utilization of energy. Compound units that have units of cost-per-unit dose or dose-per-unit benefit simplify the required comparisons.

The dollar cost per man•rem balances risk against cost. Appendix I of Title 10, CFR, Part 50 requires the use of dollars per man•rem to evaluate different dose-control strategies (i.e., the cost of additional controls is divided by the calculated dose reduction, and if the quotient is less than \$1000 per man•rem the additional dose reduction is "reasonably achievable."

Man•rem per megawatt electric•year could be used to compare dose with production. Such an approach is mentioned in the statements of consideration for an amendment to Part 50, Appendix I (*Federal Register*, 1971). Note that the expressions of cost per dose and dose per benefit have the same sense (that is, both measures become increasingly desirable as their magnitude decreases).

3.5 DISCUSSION

There are at least two objectives in a program for controlling radiation dose: to ensure that no individual receives a radiation dose leading to any identifiable effect; and to ensure that the effect of radiation on a population will be minimized, consistent with certain benefits. Guidance on the first objective is given for radioactive waste management by many standard setting bodies, such as the the National Council on Radiation Protection and Measurements, and the International Atomic Energy Agency, as well as by federal regulations (e.g., Title 10 of the CFR). These same regulations and standard setting bodies call attention to the second objective with words such as "as low as reasonably achievable." With regard to waste management, guidance for ALARA is still subjective (i.e., a specific criterion quantifying ALARA for radioactive waste-management systems has not yet been promulgated).

734 294

3.6 REFERENCES

Federal Register, Vol. 40, No. 87, p. 19441, May 4, 1975.

Federal Register, Vol. 36, No. 111, p. 11115, June 9, 1971.

National Academy of Sciences — National Research Council, *The Effects on Populations of Exposure to Low Levels of Ionizing Radiation* (NAS-NRC, Washington, D.C., 1972).

U.S. Atomic Energy Commission, *Reactor Safety Study*, WASH-1400 (U.S. AEC, Washington, D.C., 1974).

U.S. Environmental Protection Agency, *Environmental Analysis of the Uranium Fuel Cycle, Part II: Nuclear Power Reactors*, EPA-520/9-73-003-C (U.S. EPA, Washington, D.C., 1973).

734 295

4 HIGH-LEVEL SOLID WASTE AND RADIONUCLIDE RELEASE

Understanding what HLSW is, how it is contained, how it will likely be managed, what environments it is likely to experience, how radionuclides might be released, and what properties of the HLSW and canister are important in determining the amounts of radionuclides released will provide a basis for the model to be described in Sec. 5. These topics will be discussed in order. Following that, this section will describe the objectives of HLSW development up to the present and give values of properties of proposed HLSW forms where known.

It must be pointed out that this section describes primarily what presently exists, not necessarily what should be done. The findings from the overall study are expected to influence the HLSW system in the future, but the study must be based on present ideas of what will be done, even if these ideas are tentative.

In addition, it should be recognized that the present understanding of many of the topics described in this section is incomplete because of limited experimental data. It is anticipated that this study will help to delineate areas where more detailed knowledge should be obtained. These uncertainties are not expected to have a strong influence on the performance criteria that are derived, but they will affect one's ability to compare proposed waste forms against the criteria.

4.1 HLSW DEFINED

The Code of Federal Regulations in 10 CFR 50, Appendix F(1976) defines high-level liquid radioactive wastes as "those aqueous wastes resulting from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated wastes from subsequent extraction cycles, or equivalent, in a

734 296

facility for reprocessing irradiated reactor fuels." HLSW, for the purposes of this study, is defined as waste resulting from the conversion of these high-level liquid wastes to a solid form, in addition to the undissolved solids removed by centrifugation during the feed clarification step that follows the dissolving of the spent fuel elements.

4.2 GENERAL CHARACTERISTICS OF HLSW

The main features that all HLSW forms have in common are the presence of radioactive fission products and actinides and the concomitant emission of ionizing radiation, much of which is converted to heat energy in the body of the waste.

The amounts and species of the radionuclides present in HLSW can be calculated from a knowledge of the history of the spent reactor fuel to be processed and the partitioning that occurs in the reprocessing plant and the waste solidification facility. Calculations of this type have been performed by Blomeke *et al.* (1974) at Oak Ridge National Laboratory, using the ORIGEN computer code (ORNL, 1970). Because of radioactive decay and the wide spread in the values of the half-lives of the various nuclides, the amounts and species present greatly depend on the age of the wastes.

When spent fuel is first discharged from a reactor, its activity is very high and it is generally stored under water for a period of time to allow decay of the shorter-lived nuclides. This reduces the decay heat output and shielding requirement to levels where safe shipping to the reprocessing plant would be practicable. A time of about 90 days would be needed for this. (This should be regarded only as an estimate, since it depends on a number of changeable safety and economic factors.) Once the spent fuel is received at the reprocessing plant, it may be necessary to store it somewhat longer in order to insure a sufficiently low level of iodine-131 (half-life 8.04 da) and a sufficiently low external dose rate so that the plant shielding is adequate. As an estimate, the minimum age for the spent fuel at this step may be about 150 days. The maximum age may be several years. (This is particularly true for the earliest fuel to be processed, which will come from the accumulating backlog.)

After reprocessing, the high-level liquid waste could be solidified immediately, or it could be stored for as long as five years before solidification (10 CFR 50 App. F [1976]). In view of this, the activity levels, heat output, and external radiation dose rates for newly solidified wastes could vary over a wide range. As an example, consider the HLSW which results from the generation of one megawatt electric•year of energy. The volume of this waste, depending on the form selected, would lie in the range of about 1-3 x 10^{-3} cubic meters (ERDA, 1976), that is, a cube between 10 and 15 centimeters on a side. At a time of 150 days after discharge from the reactor, there would be about 50 different nuclides present that would still have significant activity (Blomeke, 1974). The total activity of this cube would be about 0.1 MCi (megacurie). The heat generation would be about 600 watts, and the external gamma dose rate without shielding would be over 10 000 rads per hour at a distance of one meter from the cube. At an age of 1 year, the radioactivity would decrease by about a factor of 2, and at 10 years, it would be smaller by a factor of more than 10. It is clear that high levels of radioactivity, dose rate, and heat output are general characteristics of all HLSW forms, and the actual values depend on the age.

4.3 CANISTERS AND SHIPPING CASKS

The protection offered by canisters and transport casks is an important factor in determining the release of radionuclides from HLSW forms, and the entire system must be considered together to arrive at accurate estimates of system performance.

The canister is generally taken to mean a relatively thin metal container that is used to confine the wastes during handling and to provide a means of lifting. In some concepts, e.g., potcal and in-can glass melting, the canister also serves as a crucible in the solidification process. In most schemes, it is not intended to provide gamma-ray shielding or protection in severe accidents.

Because of the thermochemical treatment that the canister receives when used as a crucible, some concepts involve later inserting it into an overpack canister. The overpack would not have undergone a history that might promote grain boundary embrittlement, sensitivity to stress-corrosion cracking, and baked-in contamination on its outside surface. Its residual stresses would also be small in contrast to those in the canister, since in some processes differential thermal contraction between the HLSW and canister can put the canister in tension.

For shipping, thick gamma-ray shielding and protection against accidents must be provided. It is probable that the shipping casks would be similar to those presently used for spent fuel shipping. These casks are designed to satisfy the so-called Type B standards spelled out in 10 CFR 71 (1976) and 49 CFR 173.398(c) (1974). They have shielding equivalent to about 0.3 meter of iron and weigh many megagrams.

4.4 OVERVIEW OF THE HLSW MANAGEMENT SYSTEM

The first step in determining the pertinent input to the model is to decide what operations will be performed on the solid waste. These are not completely defined at present, but the main outlines are evident from the recent ERDA document on waste management alternatives (ERDA, 1976) and the papers presented at the most recent IAEA-OECD conferences on waste management

(OECD, 1973, and IAEA, 1976). The sequence of operations described below was determined by use of these documents, discussions with researchers in waste management, and technical judgment.

In order to insure completeness, the following sequence of operations will begin at the point where the waste form is introduced into the canister, rather than after the canister is sealed. This is important because of the effects that processing may have on the canister. The model itself deals only with the risks incurred from the waste-management operations that take place after sealing. All operations involving an exposed canister will be done with shielding because of the intense radiation fields. Where operations depend on the waste form, three examples are used for illustrative purposes: spray calcine, zinc-borosilicate glass, and a hypothetical glass-ceramic-lead matrix product.

4.4.1 Canister Filling

Calcine - The spray calcine will fall directly into the canister from the calciner.

Glass - Frit and calcine will be loaded together into the canister. Melting occurs concurrently with filling.

GCLM - Glass ceramic beads will be made in a continuous melter and cooled, and then they will be pushed into a canister containing molten lead (van Geel, 1976).

4.4.2 In-Can Heat Treatment

Calcine - The calcine will be heated to drive off remaining water and nitrogen oxides.

Glass - The glass will be melted and held in a molten state to homogenize the composition of the product. Cooling will be provided to prevent overheating.

GCLM - The temperature will be maintained above the melting point of the metal matrix.

4.4.3 Cooling

Calcine - The canister will be cooled in air, inert gas, or water, depending on the heat output and the canister properties.

Glass - The hot canister will probably be cooled in air. Water cooling is a possibility.

GCLM - The canister will be cooled in air, inert gas, or water, depending on the canister properties.

4.4.4 In-Process Storage

Canisters will probably be stored for a short time between several operations. This is distinguished from interim storage, which is intended to mean longer times, of the order of months or years. In-process storage could be in water or air, depending on the heat output. In-process storage will probably occur at several points in the sequence, but it is discussed only here.

4.4.5 Inspection and Testing

It is likely that some inspection will take place before sealing. For example, samples of waste might be taken, or the void volume might be measured. The canister would be in water or air.

4.4.6 Closure Welding

This may involve some cleaning or machining of the canister lip to ensure a satisfactory welding surface. Electric arc welding will probably be used. The main body of the canister will most likely be in water during this step.

4.4.7 Weld Heat-Treating

This may be performed to reduce the possibility of later corrosion problems in the weld area. It would involve heating the weld area, with the body of the canister in the water.

4.4.8 Weld Inspection

This will be performed visually (by remote techniques) or ultrasonically. The latter would involve covering the weld area with water.

4.4.9 Leak Checking

This would probably be done by the helium detection technique. Helium would be put into the canister prior to welding, and its passage through possible leaks would be detected either by using a probe in air or by evacuating the region around the closure weld or around the entire canister with a helium detector attached to the vacuum system. The method used would depend on the required sensitivity. If evacuation around the entire can were used, some type of contacts would probably be needed to conduct away heat during leak checking.

4.4.10 Decontamination

Radioactive material on the surface of the canister would have to be removed before handling and shipping. This could involve only water and steam if loosely adhering material were the sole concern, or it could involve chemical solutions or wet sandblasting if material tied up in the oxide layer must be removed.

734 302

4.4.11 Contamination Testing

This could involve a light swipe test or something designed to remove some of the oxide layer, depending on requirements. This would probably be done in air.

4.4.12 Other Testing

Calorimetry, weighing, radiation scanning, warpage checks, ultrasonic wall thickness measurement, surface temperature measurement, or other tests might be performed, depending on requirements. Most would be conducted in water, but some would be done in air.

4.4.13 Labeling

The canister would have to be marked in some way for accounting purposes. Perhaps this would be done prior to filling.

4.4.14 Overpacking

It is conceivable that in order to meet canister requirements an overpack would be used that had not been subjected to the high temperatures and contaminated environment that the canister would experience. In this case, the canister would be loaded into an overpack, and some of steps 4.4.5 through 4.4.13 might be repeated. The overpack would not provide significant shielding.

4.4.15 Interim Storage

This term is meant to apply to storage for months or years. The interim storage site could be located at the FRP, the FWR, or another location. Storage in water is most likely at the FRP. If interim storage is used, some of steps 4.4.15 through 4.4.26 could be repeated.

4.4.16 Inserting into Transport Cask

The canister, with or without overpack, would be loaded into a Type B transport cask for shielding and accident protection. This would probably involve lifting the canister slightly higher than its length and lowering it into the cask. This may be done under water.

4.4.17 Cask Sealing

The cask would probably be sealed by bolting on a large flange with a metal gasket.

4.4.18 Cask Checking

Checks of the seal, external dose rate, and external contamination would be performed.

4.4.19 Cask Loading

The cask would be loaded and securely fastened onto a truck or railroad car.

4.4.20 Transport

The cask would be transported to the FWR or to interim storage, if such a policy were adopted.

4.4.21 Cask Checking

Repeat of step 4.4.18.

4.4.22 Cask Unloading

4.4.23 Cask Unsealing

4.4.24 Canister (Overpack) Removal

4.4.25 Canister (Overpack) Checking

This may involve several tests to ascertain that criteria are met and that damage has not occurred in transit.

4.4.26 Damaged Canister Repairing or Overpacking

Hopefully this will not be necessary, but there should probably be some provision for it. Some of steps 4.4.5 through 4.4.13 might be repeated.

4.4.27 Emplacement in FWR

4.4.28 Canister Backfilling

4.4.29 Monitoring of Temperature and Radiation

4.4.30 Location Recording

4.4.31 Repository Sealing

4.4.32 Site Marking

4.4.33 Long-Term Residence

This would be unattended and subject to geological processes. The HLSW would be of interest as long as the wastes could have a significant effect on people.

4.4.34 Retrieval

This provision is conceivable in order to remedy unforeseen geological events or to take advantage of new technological or scientific developments. The present study does not take this into account.

4.5 ENVIRONMENTS TO WHICH SOLID WASTES AND CANISTERS MIGHT BE EXPOSED

Using the sequence of steps in the HLSW management system that was postulated in Sec. 4.4, one can arrive at a likely set of environments that the waste and canister might experience. The following items include both normal and abnormal conditions, and the sequence begins with the waste in the canister.

4.5.1 In-Can Heat Treatment

The can will probably be surrounded with air or an inert gas blanket. The environment inside will include air and any residual volatiles being driven off (e.g., water and nitrogen oxides, as well as cesium, ruthenium, tellurium, and others). The temperature will range up to 900 °C for the calcine, about 1050 °C for the glass, and about 350-400 °C for the GCLM.

4.5.2 Normal Cooling

The cooling could be done with water, air, or inert gas.

734 306

4.5.3 Abnormal Cooling

In the event of a power outage or furnace failure, the filled canister could cool sooner than planned, subject to the heat-transfer characteristics of the system.

4.5.4 Overheating

It is conceivable that the furnace controller could malfunction and cause the canister to overheat. The temperature limit for a resistance furnace would be the failure temperature of the furnace winding, about 1350 °C for Kanthal and 1400 °C for Nichrome V (Baumeister, 1967). The limit for an induction furnace would be the maximum power output. It is conceivable, but not likely, that the canister melting point could be reached with a malfunctioning induction furnace. It is also conceivable that failure of the cooling system built into the canister furnace could cause overheating.

4.5.5 Accidental Fire Conditions

A bare, full canister might be subjected to a fire, although there should not be much flammable material present in the canister-handling area. Possible flammable material includes oil from shielding windows (if oil is used) and manipulator boots. It is unlikely that a fire produced by these materials would heat the canister above its initial processing temperature.

4.5.6 Storage in Water

The water temperature will probably be about 40-50 °C, and the water will be deionized. The normal Cl and F ion concentrations will be below one part per million, and the pH above 7. However, in the case of an undetected failure of the deionizing system, it is possible that the F and Cl ion concentrations would rise above 10 parts per million. Radiolysis or added

734 307

impurities may cause the pH to drop below 7. If the cooling system were to fail, the water could boil off over an extended period of time. Remedial measures might involve rapid addition of tap water. It should be assumed that oxygen is abundant in the water. Galvanic couples are conceivable if the canister were suspended from or resting on parts made from metals having compositions different from that of the canister material.

4.5.7 Storage in Air

The air temperature could range from about 0 to 300 °C for normal operation. If all convection were blocked in one concept (ARHCO, 1974), air temperature could rise to 800 °C in three to six hours.

4.5.8 Leak Checking

It may be necessary to put the canister inside a vacuum chamber to accomplish this. The temperature will depend on thermal radiation heat transfer and whatever conduction is possible through contacts provided.

4.5.9 Transportation

Inside the transport cask, the chemical environment would consist of air or water and the stainless steel cask lining. The temperature will depend on design. If water is present, it will limit the design temperature. If lead shielding is used in the transport cask, its melting point (327 °C) could not be exceeded. If depleted uranium were used, the design temperature would probably be limited by the properties of the canister and waste. The canister will be subject to some vibration in transit.

734 308

4.5.10 Impact Accidents

It is conceivable that a filled canister might be dropped during handling or suffer impact in a transportation accident. When handled by itself, it is not likely that a canister would be dropped farther than about five meters, since it would not normally be lifted any higher. In transportation accidents, greater impacts are conceivable, but the transport cask provides major protection.

4.5.11 Rapid Quenching

In this case, a filled canister of "young" waste is allowed to heat up in ambient air to its steady state temperature and is then plunged into a water bath and rapidly quenched. This could happen accidentally or intentionally, and could be repeated a few times between various steps in the process.

4.5.12 Geologic Storage

This can be broken into two types, salt and non-salt formations. In the salt case, the environments that could be considered are (1) dry NaCl, and (2) NaCl saturated with water. The temperatures of interest in salt are between 25 °C and about 250 °C. The hydrostatic pressures are from 8 to 13 megapascals, which will prevent boiling at these temperatures. In the non-salt case, the pressures are the same, but the temperatures may be higher, possibly above the boiling point. In the non-salt case, the environment is silicate minerals saturated with water up to the boiling point at the pressures of interest, or dry silicate minerals above that. The water is considered to contain the following dissolved ions expressed in weight parts per million:

Na^{+}-20

K^{+}-5

Ca^{++}-25

Mg^{++}-5

$SO_4^{=}$-15

NO_3^{-}-1

Cl^{-}-8

HCO_3^{-}-125

These concentrations are taken from Burkholder (1974) as being representative of Western U.S. desert ground water. In addition, the effects of F^{-} ion concentration up to 10 weight parts per million and pH below 7 should be considered.

4.5.13 Adversary Action

Terrorist activity has not been treated in this study.

4.6 POSTULATED RADIONUCLIDE RELEASE MECHANISMS

In order for radionuclides to be released from a solid waste form in a sealed canister, two conditions are necessary: the canister must be breached, and the radioisotopes must leave the body of the solid waste. Mechanisms for each of these processes will now be listed and described mathematically where possible. The choices of mechanisms are based upon a consideration of the environments described in Sec. 4.5 and individual judgment of experienced scientists.

4.6.1 List of Canister Breaching Mechanisms

Overheating leading to rapid corrosion by waste, external oxidation in air, creep to rupture, or melting,

Impact leading to rupture,

Undetected leak or construction defect,

Corrosion by water,

Rapid quenching leading to rupture by thermal shock,

Internal pressurization leading to bursting.

4.6.2 List of Radionuclide Release Mechanisms

Dissolution (leaching and corrosion),

Volatilization,

Airborne particulate dispersion,

Melting and liquid flow.

4.6.3 Canister Breaching Mechanisms - Discussion

Since canister breaching mechanisms depend on the construction material, three examples will be considered: 1020 carbon steel, 304L stainless steel, and 316L stainless steel. (High-nickel alloys are conceivable choices, but are not discussed here.) It should be noted that few data exist on several of these processes. Fortunately, the establishment of overall performance criteria does not depend on a detailed knowledge of them. Such a knowledge is important, however, in projecting whether or not a given HLSW-canister combination will meet the criteria.

Overheating Leading to Rapid Corrosion by Waste, External Oxidation in Air, Creep to Rupture, or Melting — Canisters are heated by energy internally generated as a result of the radioactive decay of fission products and actinides. Heat is lost from the surface by conduction, convection, and radiation, the dominant mode depending on the type of environment and its temperature. The environments of interest include vacuum, air, water, the transport cask, and geological storage. The most serious risk of overheating probably exists when the waste is "young" (less than five years out of the reactor). In particular, if a canister of young waste were involved in a fire fed by externally-supplied combustible material, the wall temperature would be increased substantially. The temperature reached under these circumstances can be calculated by standard heat transfer methods, e.g., the analysis of Hoskins (1976), modified to include thermal radiation and convection in the waste if it becomes molten. Once the temperature is known, the importance of the effects in this category can be estimated.

High Temperature Corrosion — Corrosion by the waste on the canister depends on the materials involved. For the case of calcine and steels, not much is known. Available data on cesium oxide corrosion of 304L stainless steel (Maiya and Busch, 1973) indicate that intergranular corrosion occurs with kinetics that are linear in time, with a penetration rate given by $k(\mathrm{cm/sec}) = (2.90 \pm 1.78) \times 10^{-3} \exp(-9500/T)$, where temperature is expressed in kelvin. Corrosion of 1020 carbon steel by glass will probably not be significant below about 800 °C. Corrosion of 304L stainless steel by glass can be expressed as a uniform corrosion rate:

$$K(\mathrm{mm/da}) = 4.43 \times 10^{35}\, e^{-111,000/T} \quad ,$$

where temperature is expressed in kelvin. This is a fit to data given by McElroy (1976). Corrosion of 316L stainless steel by glass is probably not significant below 1000 °C. Lead corrosion of 1020 carbon steel will be intergranular in nature and will be important above about 900 °C. Little is known about lead corrosion of stainless steels, but it could be important above about 600 °C.

External Oxidation in Air — The canister material is subject to oxidation if heated in air. Data are sparse. For carbon steel, this may occur to a depth of 0.04 millimeter after 1000 hours at a temperature of 620 °C. For 304L and 316L, it may reach a depth of 0.05 millimeter after a year at 800 °C and a depth of 0.4 millimeter after 300 hours at 1050 °C. Arrhenius behavior can be expected.

<u>Creep to Rupture</u> — Creep becomes an important mechanism above about 400 °C for carbon steel and about 500 °C for stainless steel. The linear creep rate can be expressed in Arrhenius form, but it is difficult to apply such data without a detailed stress analysis of a particular canister design.

<u>Melting</u> — If the canister were able to survive the above processes during a warmup, it would eventually melt when its melting temperature was reached. The other processes would probably dominate unless it was heated rapidly to the melting point, say, in less than one hour.

<u>Size of Breach</u> — All the processes in this category could lead to massive breaches.

<u>Impact Leading to Rupture</u> — If a full canister is impacted, the probability of breaching, the character of the breach, and the size of the breach will depend on the magnitude of impact, the design of the canister, the canister and waste material (including its prior thermal and mechanical history), the temperature of the canister, and the location of impact.

<u>Probability of Breaching</u> — Under impact loads at temperatures near room temperature, 1020 carbon steel may behave in a brittle fashion. Under these conditions, a drop through one meter or less may be sufficient to

fracture the canister, particularly if the canister has a welded-on flat bottom, is filled with calcine, and is dropped on its edge. On the other hand, if the canister is made of reverse-redrawn stainless steel with a rounded bottom, the temperature is somewhat higher, and the canister is filled with glass, it may survive a drop of 20 meters or more.

Character and Size of the Breach — For 304L and 316L, the fracture is expected to be ductile for the temperatures and strain rates of interest. In this case, the breach will normally amount to only a small crack (say, 10^{-3} times the canister area) (Smith, 1975) for impacts equivalent to up to about a 20-meter drop. For 1020 carbon steel, the fracture may be brittle, in which case the breach could be quite large, say 0.1 square meter.

Undetected Leak or Construction Defect — This type of breach would be small, say 10^{-4} times the canister area.

Corrosion by Water — Data is lacking for many of the conditions of interest. The following are estimates only. For carbon steel, the corrosion would occur uniformly over the canister surface at a rate of about 0.1-0.2 millimeter per year, and when the canister was breached, the breach would occur over its entire surface at the same time. If the canister is made of stainless steel, the general corrosion rate would be less than 0.05 millimeter per year. However, if the pH is 7 or lower and the Cl or F ion concentrations are 10 parts per million or more, the canister may crack in a short time by stress corrosion. If solutions containing Cl or F are used for decontamination, stress corrosion cracking may occur in later water storage.

Rapid Quenching and Thermal Shock Rupture — Rupture does not seem to be very likely. As a matter of fact, rapid quenching may be a part of the glass waste processing. If the canister survives quenching during processing, it will likely survive it later. Steels are generally more ductile as the temperature increases and tend not to fracture in a brittle manner under these conditions.

Internal Pressurization Leading to Bursting — This could be caused by helium buildup from alpha-decay of actinides and (n,alpha) reactions on boron (if present), radiolysis and thermal decomposition of waste, transmutations to gaseous species, or volatilization of any water accidentally added to the filled, baked-out canister prior to closure welding, as well as by increase of the pressure of confined gases as a result of heating. For the three examples of waste forms considered, all of which have been heated to at least 900 °C during processing, thermal decomposition will not be a problem if the material is later heated to temperatures less than this. If the processing temperature is exceeded, a volatile species such as Cs may present problems. For UO_2 waste, the helium buildup is not important (Mendel, 1976). Radiolysis is not significant for the examples considered. If organic, nitrated, or hydrated materials were proposed, radiolysis could be a serious problem. The only gaseous transmutation product appears to be iodine from tellurium decay, and the quantity is too small to be important.

It appears that pressure buildup will be insignificant for the waste forms considered up to 900 °C, unless water is accidentally added before sealing. In this case, the canister could rupture at a temperature above 100 °C, the actual value depending on material and design. A six-millimeter-thick carbon steel canister with flat, welded-on ends would be most vulnerable, and a thicker stainless steel canister with rounded ends would be stronger.

4.6.4 Radionuclide Release Mechanisms - Discussion

In order to focus the discussion, the same three HLSW forms will be considered as examples: spray calcine, zinc borosilicate glass, and glass-ceramic-lead-matrix (GCLM). The information that follows is based largely on the best judgment of the authors, who have considered the available data, information published in the literature, and discussions with workers in the field.

Dissolution — This process includes both leaching and corrosion of the solid waste form by a solvent, in this case, water. Leaching involves selective diffusion of radionuclides from a depth in the matrix of the material, while corrosion is related to removal of a portion of the entire matrix, including the radionuclides contained in it. The mechanisms involved are complex and not thoroughly understood. The important variables are the chemical properties of the solid matrix, the chemical properties of the solvent, and the temperature at the interface between them. The flow velocity of the solvent and the state of subdivision of the solid also influence the dissolution. For a system with fixed chemical composition, temperature, and flow velocity, one can usually fit the available data for a single radionuclide with an expression of the type

$$L = A\, t^{-1/2} e^{-H_1/RT} + Be^{-H_2/RT} + C \quad , \qquad (4\text{-}1)$$

where L is the dissolution rate in grams per square meter·second, t is the time after solvent contacts solid in seconds, A, B, and C are constants, H_1 is the activation enthalpy associated with leaching in joules per mole, H_2 is the activation enthalpy associated with corrosion in joules per mole, R is the gas constant, 8.31441 joules per mole•kelvin, and T is the temperature at the interface in kelvin.

The rate of release of a given radionuclide to the solvent is then given by the following equation, which assumes that the concentration of solutes in the solvent is constant:

$$S = L(t)\ A_L\ C_r \quad , \tag{4-2}$$

where S is the rate of release in becquerals per second•cubic meter-solid, L(t) is the dissolution rate at time t in grams per square meter•second, A_L is the effective area of solid exposed in square meters per cubic meter-solid, and C_r is the concentration of radioactivity in the solid for the particular nuclide, in becquerals per gram.

Unfortunately, there are insufficient data to define uniquely all the parameters in Eq. (4-1) for all the waste forms and all the nuclides of interest. In addition, the flow properties and the effective area A_L are not well defined in the situations of interest. Accordingly, some approximations must be made. The following appears to be the best procedure for now.

Dissolution Rate — For the case of calcine which has been processed at 900 °C, assume that 25% of the Cs and Sr dissolve immediately on contact with water. (This is possible, provided at least about 0.01 m^3 of water per canister is available.) After this initial rapid dissolution, assume that the remaining Cs and Sr and the other nuclides go into solution with a specific mass loss rate given by

$$L = (2.0 \times 10^9\ t^{-1/2} + 2.2 \times 10^8)e^{-5041/T} \quad , \tag{4-3}$$

734 317

where L is in milligrams-solid per square meter-day, t is in days, and T is in kelvin. This is based on the assumption that the remaining nuclides in calcine are a factor of 10 more leachable than those in glass.

For glass which has not been devitrified, use

$$L = (2.0 \times 10^{8}\ t^{-1/2} + 2.2 \times 10^{7})e^{-5041/T} \qquad (4\text{-}4)$$

This is an approximate fit to data given by Mendel (1977), with the temperature dependence adjusted to give a factor of 30 increase in L between 298 and 373 kelvin. For glass which has been devitrified (by heating to a temperature between 600 and 850 °C for one week or longer), use

$$L = (2.0 \times 10^{9}\ t^{-1/2} + 2.2 \times 10^{8})e^{-5041/T} \qquad (4\text{-}5)$$

This is based on the observation that devitrification raises the leach rate by about a factor of 10 (Mendel, 1977).

For the GCLM material, use

$$L = (1.0 \times 10^{9}\ t^{-1/2} + 1.1 \times 10^{8})e^{-5041/T} \qquad (4\text{-}6)$$

This is an estimate based upon data from van Geel (1976), De (1976), and Mendel (1977).

<u>Effective Area</u> — The effective area (A_L) for dissolution depends on the condition of the waste, the canister, and the leaching medium. If the canister is not breached, A is equal to zero. The most likely modes of breaching the canister are probably by corrosion in water storage, overheating

734 318

and corrosion by waste, impacting during an accident, or an original leak that was undetected. These will be discussed separately.

Corrosion in Water Storage — If a carbon steel canister is used, assume the corrosion occurs uniformly over the canister at a rate of 0.2 millimeter per year and that when the canister is breached, the breach occurs over the entire surface area at the same time. For calcine, A_L would then be equal to 10^6 square meter per cubic meter solid. For glass, A_L would be equal to $\frac{20(r+h)}{rh}$, where r and h are the radius and length, respectively, in meters. For GCLM, A_L would be $\frac{2(r+h)}{rh}$. The additional factor of 10 for glass is due to fracturing.

If the canister is made of stainless steel, the general corrosion rate is negligible. However, if the pH is 7 or lower and the Cl or F ion concentrations are 10 parts per million or more, assume that the canister cracks by stress corrosion. The resulting cracks will represent only a very small fraction of the canister area. The effective area (A_L) will depend on whether the water is flowing or still, and what the waste form is. If the water is still, decrease the above areas by a factor of 1000. If the water is flowing, decrease these areas by a factor of 100.

Overheating and Corrosion of Canister by Waste, Followed by Contact with Water — If the canister remained in contact with water, it is not likely that the waste would corrode through it because of the low temperature. This section will deal with the case of corrosion by overheating out of water (for example, by internal heat generation or fires) and subsequent contact with water (for example by attempts to cool an overheated canister or to put out a fire).

If the temperature is high enough for the canister to be breached in this way, the glass and GCLM waste forms would probably be molten and would flow out until the surface area increased to the point that sufficient cooling occurred to solidify the waste. The calcine would not melt until about 1400 °C, so it would more likely slump down into a pile.

For the calcine, use the value 10^6 square meters per cubic meter solid. For glass and GCLM, use $\frac{20(r+h)}{rh}$.

Impacting the Canister in an Accident, Followed by Contact with Water — For impacts resulting from glass-containing canister velocities less than 18 meters per second, use $A_L=0$, because the canister will probably not be breached (Smith, 1975). For impacts at velocities between 18 and 36 meters per second, breaching occurs in the form of small cracks near the point of impact. Higher velocities do not seem probable. In the case of a calcine canister, breaching is more probable. For calcine, use $A_L = 10^3$ square meters per cubic meter solid for still water and 10^4 square meters per cubic meter solid for flowing water.

For a glass canister breached by impact, the effective area results from a combination of the size of the crack and the increased surface area of the glass resulting from the fracture. The area of the crack is approximately 10^{-3} of the canister surface area, and the glass surface area increases approximately by a factor of 2 at an impact velocity of 8 meters per second and by a factor of 10 at 19 meters per second (Smith, 1975). For a glass-containing canister that is impact-breached, use $A_L = \frac{0.2(r+h)}{rh}$ for still water and $\frac{2(r+h)}{rh}$ for flowing water. For a GCLM canister, use $A_L = \frac{0.02(r+h)}{rh}$.

Original Leak Undetected - Contact with Water — An undetected leak would probably be quite small, amounting to no more than about 10^{-4} of the canister area. For calcine and still water, use $A_L = 10^2$ square meters per cubic meter solid. For glass, use $A_L = 2 \times 10^{-3} \frac{(r+h)}{rh}$, and for GCLM, use $A_L = 2 \times 10^{-4} \frac{(r+h)}{rh}$. For flowing water, multiply these by a factor of 10.

Volatilization — This process involves evaporation of chemical species from the solid waste at elevated temperatures. In order for this to occur, the nuclide of interest normally must first diffuse to the solid surface and then acquire sufficient energy to break its bonds and evaporate. A particular nuclide may evaporate as the element or as one of its oxides, depending on the partial pressures of oxygen and water in the surrounding gas phase, normally air in this context.

The rate of release will depend strongly on the temperature and the condition of the canister. If the canister is not breached, obviously no release will occur. The most likely modes of breaching the canister are the same as those described above. These modes can be divided into two groups: those which open up only a small crack in the canister, and those which expose a large fraction of the canister surface area. In the first group are impacts, water corrosion of stainless steel, and undetected leaks. In the second group are water corrosion of carbon steel and overheating due to corrosion by the waste.

The mechanism of volatilization release through a crack depends on whether the combined equilibrium vapor pressures of the species in the canister are in excess of atmospheric pressure or not. If the total vapor pressure is below atmospheric pressure, the radioisotopes can only escape by gaseous diffusion through the crack, and a state approaching thermodynamic equilibrium will exist inside the can. If the total is above atmospheric pressure, the gases will undergo viscous flow driven by the pressure difference. This will still be near-equilibrium, but at a higher pressure. Calculation of the release by these mechanisms and of the temperature at which the total vapor pressure exceeds atmospheric requires a knowledge of the equilibrium vapor pressures as a function of the temperature for the chemical species present.

734 321

The equilibrium vapor pressures will depend on the partial pressures of oxygen and water in the ambient gas phase and on the (chemical) activity coefficients of the species of interest in the solid. These activity coefficients in turn depend on the chemical composition of the solid. Unfortunately, very little information of this type is known for the solids of interest.

For the case of a large fraction of the canister area being exposed and rapid movement of the ambient air, the behavior is closer to free evaporation of material from the surface, without back condensation to establish thermodynamic equilibrium. In this case, the process is likely to be limited by diffusion of the species of interest within the solid. Unfortunately, there are very few data available on solid state diffusion in the solids of interest to waste management.

In view of the lack of complete data, the best current approach is to make an empirical fit to existing data and to apply this fit to both the above cases. The following is based on unpublished data (Gray, 1976) and technical judgment.

For calcine, the volatilization loss rate per projected area (not total surface area) is

$$L_v = 1000\ e^{-7650/T}\ (\mathrm{kCi/m^2 \cdot hr})\ \text{for}\ {}^{134}\mathrm{Cs}\ , \qquad (4\text{-}7)$$

$$L_v = 500\ e^{-7650/T}\ (\mathrm{kCi/m^2 \cdot hr})\ \text{for}\ {}^{137}\mathrm{Cs}\ , \qquad (4\text{-}8)$$

and

$$L_v = 5 \times 10^7\ e^{-21600/T}\ (\mathrm{kCi/m^2 \cdot hr})\ \text{for}\ {}^{106}\mathrm{Ru}\ , \qquad (4\text{-}9)$$

where T is in kelvin.

734 322

For glass,

$$L_v = 2 \times 10^9\ e^{-29200/T}\ (\mathrm{kCi/m^2 \cdot hr})\ \text{for}\ ^{134}\mathrm{Cs}\ , \qquad (410)$$

$$L_v = 1 \times 10^9\ e^{-29200/T}\ (\mathrm{kCi/m^2 \cdot hr})\ \text{for}\ ^{137}\mathrm{Cs}, \qquad (4\text{-}11)$$

and

$$L_v = 2 \times 10^9\ e^{-31600/T}\ (\mathrm{kCi/m^2 \cdot hr})\ \text{for}\ ^{106}\mathrm{Ru}\ . \qquad (4\text{-}12)$$

For GCLM, use the same values as those for glass.

The projected areas to be used depend on the canister breach. For the small crack cases, use 10^{-3} square meter. For the large breaches, use 1 square meter. The total volatilization release rate is then the product of L_v and this area (temperature is that of the waste surface in the large breaches and that of the canister wall for the small breaches).

In addition to the nuclides above, ^{103}Ru and the Te nuclides also have appreciable volatility, but the above species are the major ones of interest. Of course, at sufficiently high temperatures all material will enter the gas phase, but the species selected appear to be the only ones of interest for plausible temperatures.

Airborne Particulate Dispersion — This refers to spreading by air currents of particulate matter containing radionuclides. The respirable fraction of the particulates (less than 10-μm diam) is particularly important. As in the other processes, canister breaching is a prerequisite for radionuclide release.

For glass and GCLM, particulate dispersion will be quite small except in the case of an impact that fractures the glass. In general, it requires a high concentration of stress to break glass into fine particles, so it can be anticipated that the fines produced will be located near the point of impact. In order for release to occur, the canister must rupture at a location adjacent to the region containing the fines. Since stress concentration is responsible for both, this is likely. The driving force for dispersion would then have to come from the mechanical energy of the impact or the motion of ambient air. It is difficult to estimate these effects, in the absence of a detailed canister design and a more complete understanding of the phenomena involved. The best approach appears to be a judgment based on the data of Smith (1975). For glass, it appears that 5×10^{-5} weight percent of the glass could be released as particles less than 10 micrometers in diameter for a bare canister impact on a hard surface at 20 meters per second, and 5×10^{-3} weight percent at 40 meters per second.

For GCLM the fractions will be lower, because the fractured material will be largely confined inside the lead, which will deform plastically on impact.

For spray calcine, half the material is already below 10 micrometers in diameter. Thus, it is more vulnerable to this mechanism. In this case, as much as 25 weight percent of the calcine might be released for a bare canister impact at 40 meters per second. Overheating and corroding through the canister could subject about 25 weight percent of the calcine material to respirable dispersal if a sufficiently strong wind were present. However, this would be decreased by the fact that much of the calcine would have melted together into a sticky mass in the event of such temperatures.

If a spray calcine-containing canister corroded through in water storage, it is unlikely that airborne dispersion would occur, unless the water evaporated and a wind came up. In this case, 50 weight percent of the material could be dispersed as respirable particles.

Melting and Liquid Flow — The calcine melts at about 1400 °C. Such a calcine temperature is difficult to maintain in an accident configuration, and volatilization and dispersion would probably dominate in this case, anyway.

The glass softens at 550 °C and flows well at about 900 °C. If it melted in an accident, it could spread on the floor or ground until the larger surface area cooled it to a solid. The amount of spreading would be minimal. For GCLM, the metal could melt (327 °C for lead), but the glass beads would not spread far. Melting as a release mechanism can be ignored for now.

4.7 PERTINENT PROPERTIES OF HLSW AND CANISTERS

In Sec. 4.6, it was necessary to make several approximations and simplifications because of the lack of detailed property data. This section will present a list of the types of data that would be necessary for a more detailed analysis. Following this, data will be given for the three examples of HLSW forms and the three examples of canister materials where possible, in order to delineate the range of values encountered. Section 4.9 will compare some of the properties of the broad range of proposed HLSW forms.

4.7.1 List of Solid Waste Properties

Nuclear

Radionuclide inventory as a function of time.

Chemical

Composition and concentration,

Chemical form,

Gas generation rates,

Solubility and diffusivity of gases,

734 325

Vapor pressure of species present,

Solubility in water,

Corrosion and leaching properties by water.

Physical

Density,

Porosity,

Initial size distribution.

Thermal

Heat generation rate and its homogeneity,

Thermal conductivity,

Specific heat,

Thermal expansivity,

Melting or softening temperature.

Mechanical

Residual stresses,

Tensile strength,

Impact strength,

Size distribution after fracturing.

4.7.2. List of Canister Properties

Chemical

Corrosion of waste on canister,

Corrosion of water on canister,

Oxidation of canister in air.

734 326

Physical

- Density

Thermal

- Thermal conductivity,
- Specific heat,
- Emissivity,
- Heat transfer coefficient at waste-canister interface,
- Presence or absence of heat transfer fins inside canister.
- Thermal expansivity,
- Melting temperature.

Mechanical

- Shape of canister,
- Dimensions of canister,
- Creep-rupture strength,
- Residual stresses,
- Tensile strength,
- Yield strength,
- Ductility,
- Resistance to penetration,
- Weld characteristics,
- Void space inside.

4.7.3 Discussion of Solid Waste Properties

Nuclear — Radionuclide Inventory as a Function of Time — Numerous radionuclides are present in high level waste, but a relatively small number of them contribute the major hazard potential (see Appendix A). For purposes of the initial work, consideration has been limited to the nuclides shown in Table 4-1. These data are based on ORNL (1970), Blomeke (1974), and Gera, (1975), with a power level of 30 megawatts per megagram of uranium, a burnup of 33 000 megawatt days per megagram of uranium, a flux of 2.92×10^{13} neutrons per square centimeter·second, and a thermal-to-electrical conversion efficiency of 35.4 percent. The fuel is UO_2. Zero is taken as the time at which the fuel is removed from the reactor. The ^{129}I concentration depends on the time of reprocessing, which is taken to be 150 days.

If the fuel were mixed oxide (PuO_2 as well as UO_2), the numbers would be similar for the fission products (within a factor of 2) but larger for the actinides (about a factor of 10, depending on the species).

These nuclides were selected for their biological hazard potential. Te is included because of its high volatility. This same inventory should apply to all HLSW forms. (Minor differences in the volatile species could result from different processing temperatures, but these are ignored for now.)

Chemical

Composition and Concentration

Spray Calcine — The calcine is composed of oxides of the fission products, the actinides, and stable elements including Fe, Cr, Ni, Na, and Gd, B, or Cd. The volume of calcine is about 30-60 litres per megagram of uranium or about $1\text{-}2 \times 10^{-3}$ cubic meters per megawatt electric·year.

Zinc-borosilicate Glass — The chemical composition of the glass waste form is as follows (Mendel, 1976):

Table 4-1. Source terms for biologically significant nuclides (units are Ci/MWe•y).

	150 da	1 yr	2	5	10	10^2	10^3	10^4	10^5	10^6
^{89}Sr	3.0E3	1.7E2	1.4E0	8.[illegible]	0	0	0	0	0	0
^{90}Sr	2.4E3	2.4E3	2.3E3	2.1E3	1.9E3	2.0E2	4.7E-8	0	0	0
^{90}Y	2.4E3	2.4E3	2.3E3	2.1E3	1.9E3	2.0E2	4.7E-8	0	0	0
^{91}Y	5.0E3	3.9E2	5.4E0	1.4E-5	0	0	0	0	0	0
^{93}Zr	5.9E-2	5.9E-2	5.9E-2	5.9E-2	5.9E-2	5.9E-2	5.9E-2	5.9E-2	5.7E-2	3.9E-2
^{93m}Nb	1.2E-3	2.9E-3	5.7E-3	[illegible]	2.4E-2	5.9E-2	5.9E-2	5.9E-2	5.7E-2	3.9E-2
^{95}Zr	8.6E3	8.7E2	1.9E1	1.7E-4	0	0	0	0	0	0
^{95}Nb	1.6E4	1.9E3	4.1E1	3.6E-4	0	0	0	0	0	0
^{99}Tc	4.4E-1	4.4E-1	4.4E-1	4.4E-1	4.4E-1	4.4E-1	4.4E-1	4.4E-1	3.3E-1	1.7E-2
^{103}Ru	2.0E3	6.5E1	1.1E-1	0	0	0	0	0	0	0
^{106}Ru	1.3E4	8.5E3	4.2E3	5.3E2	1.7E1	0	0	0	0	0
^{106}Rh	1.3E4	8.5E3	4.2E3	5.3E2	1.7E1	0	0	0	0	0
^{125}Sb	2.5E2	2.2E2	1.7E2	7.9E1	2.2E1	0	0	0	0	0
^{126}Sn	1.7E-2	1.7E-2	1.7E-2	1.7E-2	1.7E-2	1.7E-2	1.7E-2	1.7E-2	9.1E-3	1.8E-5
^{127m}Te	1.9E2	4.9E1	4.8E0	4.5E-3	4.1E-8	0	0	0	0	0
^{127}Te	1.9E2	4.9E1	4.8E0	4.5E-3	4.1E-8	0	0	0	0	0
^{129m}Te	2.1E2	2.6E0	1.6E-3	0	0	0	0	0	0	0
^{129}Te	1.3E2	1.7E0	1.0E-3	0	0	0	0	0	0	0
^{129}I	2.4E-6	2.4E-6	2.4E-6	2.4E-6	2.4E-6	2.4E-6	2.4E-6	2.4E-6	2.4E-6	2.3E-6
^{134}Cs	6.7E3	5.5E3	3.9E3	1.4E3	2.6E2	0	0	0	0	0
^{137}Cs	3.3E3	3.3E3	3.2E3	3.0E3	2.7E3	3.3E2	3.1E-7	0	0	0
^{144}Ce	2.4E4	1.4E4	5.7E3	3.8E2	4.7E0	0	0	0	0	0
^{147}Pm	3.1E3	2.7E3	2.1E3	9.3E2	2.5E2	1.1E-8	0	0	0	0
^{154}Eu	2.1E2	2.1E2	2.0E2	1.8E2	1.4E2	2.9E0	0	0	0	0
^{210}Pb	0	0	0	0	0	2.3E-8	2.2E-6	8.8E-5	7.0E-4	1.7E-4

Table 4-1. (Continued)

	150 da	1 yr	2	5	10	10^2	10^3	10^4	10^5	10^6
^{210}Po	0	0	0	0	0	2.3E-8	2.2E-6	8.8E-5	7.0E-4	1.7E-4
^{226}Ra	0	0	0	1.7E-9	3.4E-9	3.5E-8	2.2E-6	8.8E-5	7.0E-4	1.7E-4
^{229}Th	0	0	0	0	1.3E-9	2.3E-8	2.2E-6	1.7E-4	3.7E-3	9.1E-3
^{230}Th	6.6E-7	6.6E-7	6.6E-7	6.6E-7	6.6E-7	1.0E-6	1.2E-5	1.1E-4	6.9E-4	1.7E-4
^{231}Pa	7.7E-7	7.7E-7	7.7E-7	7.7E-7	7.7E-7	7.7E-7	8.1E-7	1.2E-6	7.5E-6	1.0E-5
^{233}U	4.6E-7	4.6E-7	4.6E-7	4.6E-7	4.6E-7	4.6E-6	4.8E-5	4.9E-4	4.0E-3	9.0E-3
^{237}Np	1.1E-2	1.1E-2	1.1E-2	1.1E-2	1.1E-2	1.1E-2	1.2E-2	1.2E-2	1.1E-2	8.5E-3
^{238}Pu	1.1E0	2.5E0	5.2E0	1.5E1	3.2E0	1.6E0	3.8E-3	0	0	0
^{239}Pu	5.2E-2	5.2E-2	5.2E-2	5.2E-2	5.2E-2	5.2E-2	6.4E-2	1.3E-1	1.8E-2	7.6E-9
^{240}Pu	7.6E-2	8.1E-2	9.2E-2	1.2E-1	1.4E-1	2.7E-1	2.5E-1	9.9E-2	9.7E-6	0
^{241}Pu	1.8E1	1.7E1	1.6E1	1.4E1	1.0E1	1.5E-1	9.8E-3	4.6E-3	2.4E-6	0
^{241}Am	5.4E0	5.4E0	5.4E0	5.4E0	5.5E0	5.1E0	1.1E0	4.6E-3	0	0
^{243}Am	5.4E-1	5.4E-1	5.4E-1	5.4E-1	5.4E-1	5.4E-1	5.0E-1	2.3E-1	6.6E-5	5.2E-9
^{242}Cm	**4.7E2**	**1.9E2**	4.5E1	4.2E-1	2.2E-1	1.5E-1	2.4E-3	0	0	0
^{244}Cm	7.8E1	7.6E1	7.3E1	6.5E1	5.4E1	1.7E0	0	0	0	0

SiO_2	27.31 wt%	NiO	0.66	TeO_2	0.46
B_2O_3	11.15	P_2O_5	0.42	Cs_2O	1.82
Na_2O	4.06	Rb_2O	0.22	La_2O_3	0.93
K_2O	4.06	Y_2O_3	0.38	CeO_2	2.11
ZnO	21.34	ZrO_2	3.13	Pr_6O_{11}	0.94
CaO	1.47	MoO_3	4.03	Nd_2O_3	2.95
MgO	1.47	RuO_2	1.88	Sm_2O_3	0.58
SrO	2.15	Rh_2O_3	0.30	Eu_2O_3	0.13
BaO	2.47	PdO	0.94	Gd_2O_3	0.09
Fe_2O_3	0.95	Ag_2O	0.06	UO_2	1.26
Cr_2O_3	0.22	CdO	0.06		

The volume of the glass waste form is 40-100 liters/MgU or about 1.2 to 3.1×10^{-3} cubic meters per megawatt electric·year.

Glass-Ceramic-Lead-Matrix - For the glass-ceramic-in-lead waste form, the base chemical composition of the glass-ceramic before adding the waste oxides would be (Dé, 1976):

SiO_2	32-50 wt%
Al_2O_3	12-15
B_2O_3	5-10
CaO	12-14
Na_2O	5-8
Li_2O	0-3
TiO_2	10-14
ZnO	0-3
Cs_2O	0-2

The matrix is assumed to be lead of commercial purity (say 99 wt%). The volume of the composite material would be about 40-100 liters per megagram of uranium or about 1.2 to 3.1×10^{-3} cubic meters per megawatt electric•year.

Chemical Form

Calcine — The calcine is in powder form, probably consisting primarily of simple oxides of the waste cations.

Glass - The glass structure is a random network formed by bonding of the Si and B to oxygen atoms, as modified by the other constituents. Most of the species are dissolved in the glass. However, PdO, RuO_2, and Rh_2O_3 are not dissolved, and Mo may be present as a separate sodium molybdate phase, which can dissolve some Sr and Cs. The use of reducing agents is planned to control and hopefully eliminate this phase. Devitrification may cause some species to come out of solution.

GCLM — The glass ceramic consists of several different crystalline phases held together by a glassy phase. The dominant crystalline phase chosen here is perovskite, and others present are $CaMoO_4$, $(Ce,Zr)O_2$, and pollucite (Dé, 1976).

Gas Generation (see also Sec. 4.6.3 — Internal Pressurization Leading to Bursting) Gas generation can come from alpha-decay, (n,alpha) reactions, radiolysis, thermal decomposition, transmutation, or volatilization. Alpha decay will produce the following total helium for the times shown estimated from curves given by Roberts (1976):

734 332

Time (yr)	Total He (atoms/MWe·yr)
1	5.3×10^{20}
10	1.8×10^{21}
100	4×10^{21}
1000	8×10^{21}
10 000	1.8×10^{22}

For Pu fuel, these numbers are about a factor of 10 larger. The other mechanisms do not appear to be significant for the waste forms considered.

Solubility and Diffusivity of Gases — Helium will be the only gas considered for now, since it would be the major one present under normal conditions. Helium can be considered to have zero solubility in the example waste forms and therefore tends to diffuse out to pores, interparticulate spaces, and the canister void space (plenum). The rate at which this occurs depends on the waste form, the temperature, and the radiation damage state. For calcine, the particle size is so small that immediate migration can be assumed. For glass and GCLM, the helium diffusivity can be taken as $D=2.1 \times 10^{-3}\ e^{-7500/T}$ square centimeters per second, where temperature is in kelvin (Turcotte, 1976). Radiation damage gives rise to trapping and non-Arrhenius behavior, which yields lower effective diffusivity, so this is a conservative value.

Vapor Pressure of Species Present (see also Sec. 4.6.4 — Volatilization) - Vapor pressures depend on the concentration and chemical form of the species present and are not known for these waste forms. If water is accidentally present, it will be the most volatile species. The vapor pressure for free water is as follows (Keenan, 1969):

734 333

Temperature (°C)	Pressure (kPa)
0	0.611
100	101.3
200	1 560
300	8 590
374.1 (critical temperature)	22 100

Above the critical temperature, the pressure will depend on the void volume and the amount of water present. In calcine, the volatile species will be oxygen and Cs, Te, Ru, Mo, and Rb in equilibrium with their oxides. For glass and GCLM, in addition to these, K, B, and Na will be in equilibrium with their oxides. The vapor pressure of lead metal is small compared to these.

Solubility in Water (see also Sec. 4.6.4 — Dissolution) — Among oxides, the most soluble are those of the Groups IA and IIA elements. Most of the others are not very soluble. Sodium molybdate is fairly soluble. Calcium molybdate is not.

Corrosion and Leaching (see Sec. 4.6.4 — Dissolution)

Physical

Density

Calcine	1000-2400 kilograms per cubic meter
Glass	3000-3600
GCLM	2400-5500

734 334

Porosity

Calcine	30-75 percent,
Glass	less than 1 percent,
GCLM	less than 1 percent.

Initial Size Distribution

Calcine - Average size by weight, 10 micrometers,
Average size by number, 2-5 micrometers,
Respirable fraction, 50 percent by weight.
(Bonner, 1976)

Glass - Large pieces, typically a few centimeters in size (a fractured monolith).

GCLM - Spherical glass beads, 5-millimeter diameter, imbedded in monolithic lead block that is the size of the canister.

Thermal

Heat Generation Rate and Its Homogeneity — The following heat generation rates (in W/MWe•yr) are for fuel irradiated to 32 megawatts electric•year per megagram of uranium and reprocessed 90 days after removal from the reactor (based on ORNL [1970]).

					Time					
	90 da	150	210	270	1 yr	2	5	10	10^2	10^3
Fission products	8.18E2	6.03E2	4.77E2	3.95E2	3.12E2	1.72E2	6.95E1	3.22E1	3.25E0	5.53E-4
Actinides	2.52E1	2.02E1	1.52E1	1.24E1	9.84E0	5.94E0	3.28E0	2.21E0	2.16E-1	7.09E-2
Total	8.43E2	6.22E2	4.91E2	**4.08E2**	3.22E2	1.78E2	7.31E1	3.44E1	3.56E0	7.15E-2

To calculate heat at other times in the range 150 days to 10 years, use (with time in years):

$$\text{Fission product heat} = \log_{10}^{-1}\ 2.49415 - 0.809319\,[\log_{10}t] - 0.176980\,[\log_{10}t]^2$$

$$\text{Actinide heat} = \log_{10}^{-1}\ 0.992995 - 0.764164\,[\log_{10}t] + 0.115561\,[\log_{10}t]^2$$

$$\text{Total decay heat} = \log_{10}^{-1}\ 2.50786 - 0.804713\,[\log_{10}t] - 0.166584\,[\log_{10}t]^2$$

The calcine may tend to pack more densely at the bottom of the canister, particularly if vibrated. In transport, the canister would be horizontal, so this should not be a problem. The thermal conductivity of the calcine is approximately proportional to the density, so the center temperature should be relatively independent of packing for a given heat generation per unit mass. If heated to about 1400 °C, the calcine could melt and become four times as dense, generating four times as much heat per unit volume.

The glass should be fairly homogeneous from a heat production standpoint up to at least 800 °C. Above this temperature, some segregation will occur, with some species moving up and some down because of density differences. This may have some effect on heat generation homogeneity.

The GCLM will be very homogeneous, and the high thermal conductivity of lead will smooth out most temperature variations.

Thermal Conductivity

Calcine - This property varies with both the temperature and the density. One can use the equation of Ross (1975), which was actually determined for a fluidized-bed calcine: $k = [0.177 + 10^{-4}\ (w-2100)]$ $[1 + 0.00148T]$, where k is in watts per meter·kelvin, w is the density in kilograms per cubic meter, and T is in degrees centigrade.

Glass - For temperatures from 25 °C to 600 °C, the thermal conductivity is given by: $k = 0.85 + 9.75 \times 10^{-4}$ (T-25), where k is in watts per meter·kelvin and T is in degrees centigrade. This is a fit to data given by Ross (1975b).

At temperatures above 600 °C, the effective thermal conductivity rises rapidly because of convection in the molten glass (McElroy, 1972). Use k = 2.0 watts per meter·kelvin at 700 °C and k = 2.5 watts per meter·kelvin at 730 °C.

GCLM - Use k = 10 watts per meter·kelvin (van Geel, 1976).

Specific Heat - Assume constant.

Calcine - 650 joules per kilogram·kelvin (approximate fit to data of Ross, 1975b),

Glass - 750 joules per kilogram·kelvin (approximate fit to data of Ross, 1975b),

GCLM - 350 joules per kilogram·kelvin

Thermal Expansivity

Calcine - Assume zero, because the particles will compress together to compensate for it.

Glass - 8×10^{-6} per kelvin,

GCLM - 29×10^{-6} per kelvin.

Melting or Softening Temperature

Calcine - 1400 °C,

Glass - 550 °C (dilatometric softening)

800 °C (molten),

GCLM - 327 °C for lead,

734 337

$$\left.\begin{array}{l}700\ ^{\circ}\mathrm{C}\ \text{(dilatometric)}\\ 800\ ^{\circ}\mathrm{C}\ \text{(molten)}\end{array}\right\}\ \text{for glass-ceramic.}$$

Mechanical

Residual stresses

Calcine - none.

Glass - Stresses can be trapped during cooling from the melt or impressed on glass by canister because of differential thermal expansion. Fast quenching increases the former but relieves the latter. Glass cracks during cooling partly in response to these stresses.

GCLM - Stresses are negligible because of small bead diameter, easy plastic deformation of lead, and larger thermal expansivity of GCLM than that of canister.

Tensile strength

Calcine - zero.

Glass - Depends on surface condition, environment, and size. Assume 35 megapascals.

GCLM - 4 [illegible]apascals.

Impact Strength — This is difficult to quantify. The calcine has no mechanical stability. The glass will fracture under impact, requiring about 0.1 joules per square centimeter of new surface area (Wallace and Kelley, 1976). The GCLM is very resistant to impact because of the energy absorbed in plastic deformation of lead.

Size Distribution after Fracturing

Calcine - unchanged.

Glass - For impact fracture, the fraction of the glass fragments smaller than 10 micrometers in size is as follows (data from Smith [1975]):

Impact Velocity (m/sec)	wt% below 10 μm
0	10^{-6}
10	10^{-5}
15	10^{-4}
20	10^{-3}
40	10^{-2}

For thermal fracture, the respirable fraction is negligible.

GCLM - Not much information is available, but the respirable fraction is expected to be small because glass ceramics break into large shards and the lead will tend to distribute loads. Use 10^{-4} weight percent for impact and 10^{-7} weight percent for thermal fracture.

4.7.4 Discussion of Canister Properties

Chemical

Corrosion of waste on canister (see Sec. 4.6.3),

Corrosion of water on canister (see Sec. 4.6.3),

Oxidation of canister in air (see Sec. 4.6.3).

Physical

Density

1020 carbon steel - 7860 kilograms per cubic meter,

304L stainless steel - 7900 kilograms per cubic meter,

316L stainless steel - 7900 kilograms per cubic meter.

734 339

Thermal

Thermal Conductivity

1020 steel - 52 watts per meter·kelvin,

304L steel - 17 watts per meter·kelvin,

316L steel - 17 watts per meter·kelvin.

Specific Heat — 500 joules per kilogram·kelvin for all.

Emissivity — Use 0.6 to 0.9. If the canister is oxidized, the emissivity increases. If it is shiny (e.g., stainless steel that has been decontaminated with an acid etch), the emissivity is decreased, possibly to as low as 0.4.

Heat Transfer Coefficient at Waste-Canister Interface — Assume good contact exists at the interface in all cases.

Heat Transfer Fins Inside Canister — It has been found advantageous to put radial fins inside the canister, both to transfer in heat during in-can melting and calcine bake-out, and later to transfer out internally-generated heat. For calcine-filled canisters, this becomes advantageous for quite small diameters. For glass, it is desirable for diameters greater than 0.25 meter in order to keep the centerline temperature below 800 °C during air storage.

Thermal expansivity

1020 steel - 11.7×10^{-6} per kelvin,

304L steel - 17×10^{-6} per kelvin,

316L steel - 17×10^{-6} per kelvin.

Melting temperature

1020 steel - 1536 °C
304L steel - 1400 °C
316L steel - 1400 °C
melting begins.

Mechanical

Shape of Canister — The canister is nominally a right cylinder. The ends could be flat or rounded. The top must be designed for lifting, coupling to the process equipment, and closure by welding.

Dimensions of Canister

Diameter - 0.16-0.6 meter,
Length - 3-4.5 meters,
Wall Thickness - 6-20 millimeters.

Creep-Rupture Properties (see Sec. 4.6.3).

Impact-Rupture Properties (see Sec. 4.6.3).

Residual Stresses — These can result from differential thermal contraction of the waste and canister, and from welding. In some concepts, the canister has final tensile hoop and axial stresses equal to its work-hardened yield strength.

Tensile Strength — Available in materials data sheets for the particular thermo-mechanical history.

734 341

Yield Strength — See materials data sheets.

Ductility — See materials data sheets. Could be affected by chemical environment. Grain boundary embrittlement may occur.

Resistance to Penetration — This is very geometry-dependent.

Weld Characteristics — These depend on type of weld, material, and heat treating.

Void Space Inside — About 10 percent is a reasonable estimate. It is used to prevent overfill, to keep waste below closure weld, and to serve as a gas plenum. With glass, the contraction on solidification produces some void space.

4.8 OBJECTIVES OF HLSW FORM DEVELOPMENT UP TO THE PRESENT

During the past few years a great deal of effort has been expended in this country and elsewhere to develop HLSW forms. This work is described in the recent ERDA document on technical alternatives for waste management (ERDA, 1976) and in the proceedings of two conferences held in Europe (OECD, 1973, and IAEA, 1976). In addition, there have been numerous papers in the technical journals describing various treatment processes and waste forms.

Although the detailed performance requirements for HLSW have not been evaluated up to this time, the development of waste forms appears to have been directed toward these general objectives:

734 342

- To reduce the volume of the liquid waste.
- To convert the waste into a form having greater chemical, thermal, radiolytic, and mechanical stability, so that it can be more readily confined and isolated from the biosphere for long time periods.
- To develop a process that is economically feasible, taking account of simplicity, reliability, energy requirements, hardware costs, and raw materials added in the process.

These objectives are generally felt to be best satisfied by solidification of the wastes, and this has become the policy of the U.S. government, as delineated in 10 CFR 50, Appendix F (1976), as well as that of several European countries. Accordingly, workers in various laboratories have developed a range of solid-waste form options that meet the above objectives to varying degrees.

4.9 PROPOSED HLSW FORMS AND PROPERTY VALUES

Proposed HLSW forms range from salt cake through calcines, glasses, supercalcine, coated particles, sintered products, glass ceramics, nepheline syenite, metal matrix composites, ion exchange products, and rock melt. (This is not an exhaustive list.) In general, the more complex or higher technology HLSW forms offer greater stability against radionuclide release, but they also cost more to produce. A description of these waste forms will now be presented, and the values for their most significant release-related properties will be given. It should be noted that many of these materials have not been thoroughly characterized, and property values often can only be estimated. There is a clear need for careful measurement of the release-related properties if one is to refine the estimates of radionuclide release under various conditions of interest in the waste-management system.

4.9.1 Salt Cake

The simplest way of reducing high-level liquid waste to a solid is to allow the water to evaporate, forming a cake of nitrate salts of the fission products, actinides, and other constituents. This method has been used at Hanford for processing ERDA wastes. The chief limitations of this waste form appear to be the presence of residual water and nitrates, which are subject to radiolysis, and the high solubility in water.

4.9.2 Calcines

By heating to somewhat higher temperatures, in the range 670 K (400 °C) to 1170 K (900 °C), the nitrate salts are converted to oxides and elements, depending on the feed composition and the abundance of oxygen present. This product is termed a calcine. The nitrogen oxides and water are driven off in vapor and gaseous forms. There are several processes which have been used for calcination, and the calcines produced differ from each other in some respects. Four products are discussed here. They are pot calcine, spray calcine, fluidized bed calcine, and rotary kiln calcine.

Pot Calcine - The pot calcination process (potcal) was developed by Oak Ridge National Laboratory in the early 1960s. In this process, concentrated waste from an evaporator is fed into a pot (canister) heated by a furnace, where the waste is denitrated and reduced to a calcine. Additives required to produce a desirable solid may be added to the waste or separately to the pot. As the liquid becomes concentrated, it begins to form scale on the inside walls of the canister. As the can becomes full of scale, the incoming feed addition rate continuously drops to a very low rate and is turned off. Radioactive decay heat causes the calcine temperature to rise at a fast rate so that the furnace temperature has to be steadily decreased from 900 °C while the calcine in the center is increasing to 900 °C, the required temperature for complete calcination.

The product from the potcal process is a porous cake of friable calcine containing about 90% high-level waste and 10% additives to reduce volatilization. The calcine is readily soluble up to 20 to 50%. The rest of the material is mostly insoluble. Thermal characteristics are relatively poor.

Radioactive demonstration of the pot calcination process was carried out in the Waste Solidification Engineering Prototypes (WSEP) program at Hanford. Although the process was satisfactorily operated with fully radioactive waste, the potcal process has not been pursued further because of its apparent capacity limitations.

Spray Calcine - The spray calcine product has been developed as an intermediary to some higher order solidification product such as a glass or glass-ceramic. The concept has been extensively developed at the Pacific Northwest Laboratory (PNL) operated by the Battelle Northwest Laboratories (BNWL), where the spray calcine is primarily used as the waste form in zinc-borosilicate glass.

In this process, the high-level liquid waste is pumped to an internal mixing pneumatic atomizing nozzle in the top of a heated (700 °C wall temperature) spray calciner barrel. The atomized droplets (nominally 70-μm-diam) are flash dried and calcined as they fall through the hot barrel. The finely divided powdery product is separated from the off-gas by sintered stainless steel filters.

The product is a very fine powder. The average size by number is 2-5 micrometers. The average size by weight is 10 micrometers. The product has a relatively low density and thermal conductivity.

Thirteen fully radioactive engineering-scale runs with the spray calciner were made in the WSEP program, and over 1000 operating hours with simulated wastes have shown the process to be simple and reliable. Although developed primarily for use in combination with glass processes, the spray calciner could be used alone if the product is found to be adequate. A 900 °C heat treatment would probably be needed to drive off residual nitrogen oxides and water.

Fluidized Bed Calcine - Fluidized bed calcination was developed at the Idaho National Engineering Laboratory during the 1952-59 period. In fluidized bed calcination, wastes are atomized into a fluidized bed and heated by in-bed combustion. The bed temperature is in the 500-600 °C range. Evaporation occurs on the surface of particles and results in a product consisting of granular bed material and powdered calcine, both of which are removed from the calciner. The size of the grains may range up to about 1 millimeter. Heating to 600-700 °C in a second fluidized bed reduces the residual nitrates and water to less than 0.02 percent and 0.006 percent by weight, respectively (Rindfleisch, 1976).

A demonstration plant-scale facility for solidifying aluminum nitrate wastes was constructed, and it began "hot" operation in 1963. A larger facility is under construction.

Rotary Kiln Calcine - The development of the rotary kiln calciner has been carried out almost entirely in France, where the process is designed as an intermediary to the vitrification of high-level nuclear wastes. In addition, some developmental work has been carried out at various U. S. research laboratories.

The rotary kiln calcine product is a finely divided oxide powder. The calcination reaction is driven by external resistance furnaces, which provide a 770 K (500 °C) environment for calcination. The French have required a post-calciner heat treatment of 1170 K (900 °C) for the calcine product in order to assure complete dehydration.

Most untreated and treated high-level liquid wastes may be calcined in the rotary kiln. Because the process has been designed as an intermediary to some higher order solidification process, a void of characterizing information exists which must be filled if this product is to be proposed as a final waste form for disposal.

4.9.3 Borosilicate and Phosphate Glass

Both borosilicate and phosphate glass formulations have been emphasized and developed internationally for the near-term immobilization of high-level liquid radioactive wastes. Several waste vitrification processes have been carried through various degrees of development, including two major radioactive pilot plant operations. In the U.S. from the years 1966 to 1970, the WSEP program demonstrated waste fixation in glass. In France, the PIVER pilot plant began vitrifying high-level liquid wastes from the Marcoule Purex plant in May 1969.

Glasses are composed of network formers and network modifiers. Silicon, boron, aluminum, and phosphorous are network formers because they bond to oxygen with high bond strength to form an extended network. Boron lowers the melting temperatures and, along with aluminum, increases chemical stability as

734 347

measured by leach tests. Currently, borosilicate glasses are favored over phosphate glasses because of their greater stability and the much lower leach rates of the devitrified products. Also, borosilicate glass formulations are less corrosive to the stainless steel processing vessel, which also serves as the final storage vessel. For high-level liquid wastes containing sulfate, phosphate glass formulations are preferred because sulfate forms an immiscible, water-soluble phase in borosilicate glass.

Because of progress in waste vitrification, several nations have chosen to convert to glass their high-level liquid wastes arising from nuclear power generation. Currently, borosilicate glass processes are being investigated by England, France, India, Germany, and the U.S. Phosphate glass processes are being investigated by Germany and the USSR.

4.9.4 Supercalcine

"Supercalcine" is a term for a "tailor-made" crystalline ceramic waste form prepared by modifying the high-level waste stream with selected additives (35 to 45% by weight) so that when it is calcined and given additional heat treatment, an assemblage of thermodynamically stable crystalline phases will be formed that is more refractory and leach-resistant than ordinary oxide calcine formed without additives. Supercalcine is a potentially superior calcine product as it stands; it can also be used as the core for a composite multibarrier waste form.

Additives (usually including Ca, Sr, Al and Si), selected according to the waste composition, are solution mixed with the high-level liquid wastes. The additive-waste solution can be calcined by any of the high-level liquid waste calcination procedures described previously. Heat treatment at 900 to 1100 °C is required to develop fully the desired crystalline species.

Supercalcine was developed at Pennsylvania State University starting in the fall of 1973. One of the formulations has undergone engineering scale tests at PNL by spray calcining.

4.9.5 Coated Particles

The coated pellet process is under early laboratory development in the U.S. The coating material, its thickness, and the process for applying it are currently being selected and optimized.

High-quality coatings can be applied by Chemical Vapor Deposition (CVD) techniques in either a fluid bed or a drum coater, as has been demonstrated with HTGR fuel at ORNL. The CVD coatings are formed from reactions such as:

$$2AlCl_3 + 3H_2O \rightarrow Al_2O_3 + 6HCl$$

in the presence of dry atmosphere.

Carbon coatings are formed by the thermal decomposition of hydrocarbon gases at temperatures above 800 °C.

Stable pellets of solidified high-level waste oxides completely sealed within a nonradioactive coating of leach-resistant material could provide good protection of the waste from leaching or vaporization for long periods. The pellets could then be imbedded in a metal matrix providing increased strength, impact resistance, and high thermal conductivity.

4.9.6 Sintering Process

The processes for sintered products involve a common high-temperature heat treatment where the products are reacted and densified. Sintered calcine-flux products have been developed at the Idaho National Engineering Laboratory (INEL) and at PNL. At INEL and PNL, the processes are similar (i.e., the calcine wastes and a flux or frit are mixed, formed into the desired shape, and sintered to react the calcine with the additives and to increase product density, strength, and leach-resistance). The products have both a glass and a crystalline phase, and all calcinable high-level liquid wastes and intermediate-level wastes are applicable.

4.9.7 Glass Ceramic

The term glass-ceramic, as used here, applies specifically to a homogeneous glass that has been subjected to a controlled temperature cycle in which crystals are nucleated throughout the body. Then the temperature is increased and the crystals allowed to grow until the body is no longer a glass, but mainly a very fine grained crystalline material.

A glass-ceramic is stronger than glass, is not susceptible to further divitrification at high storage temperatures, and has leach resistance similar to glass. Application of the glass-ceramic concept to high-level liquid waste solidification is known to improve the thermal and mechanical stability of the waste product and is currently being developed in Germany.

4.9.8 Nepheline Syenite

Nepheline syenite is a naturally occurring alumino-silicate mineral. In the 1950s, workers in Canada developed a glass material based upon this mineral and lime, incorporating a nitric acid solution of fission product waste. Some of these blocks have been buried and monitored for leachability for over 15 years, without any canister or containment. The leaching is reported to be quite low.

4.9.9 Metal Matrices

High-level wastes less than five years old have high heat-generation rates. Metal matrices are being developed to provide a high effective conductivity to lower centerline temperatures. The use of highly ductile metals, such as lead, also increases impact resistance. Molten metal cast matrices have been developed by EUROCHEMIC and Gelsenburg AG, and sintered metal matrices are being investigated at INEL and PNL. The waste forms have been either granular calcines or vitrified waste beads.

Powdered metal sintering processes are being investigated at INEL and PNL. In the INEL sintering process, calcine, as produced or stabilized, is mixed with the metal powder in order to coat the calcine. The mix is fed to a press, compacted, and rapidly brought to the sintering temperature of 520-600 K.

In the PNL gravity sintering process, the waste pellets or beads are loaded directly into the canister. The metal powder is loaded into the remaining void volume, and the canister is sealed and heated to the desired sintering temperature.

Wastes may be incorporated in the form of granules, beads, or pellets. Products have been made and tested on a laboratory scale.

4.9.10 Ion Exchange Products

The ion exchange process (exchange of ions across a boundary between two phases) has been used to remove cesium selectively from a high-level liquid waste on a zeolite bed at Savannah River Laboratory. It has also been used at Sandia Laboratories for total solidification by fixation of all ionic species, radionuclides and others, in the high-level liquid waste on artificial inorganic ion exchange media (e.g., sodium titanate, niobate, or zirconate).

734 351

These materials do not effectively trap cesium, and a zeolite bed is used to remove cesium. This material is added to the inorganic ion exchangers prior to formation of the final product, which is formed by compaction and sintering and is a tough ceramic with low leachability and high thermal stability.

4.9.11 Rock Melt

The rock melt has been suggested as a type of deep geological disposal method. The work has not advanced past the theoretical stage at Lawrence Livermore Laboratory (LLL) but has been further developed at Sandia Laboratories.

A promising rock melt scheme is to transport the solidified high-level wastes to an *in situ* melting site for disposal where the waste canisters are placed in a regular geometric arrangement in a crushed rock or glass backfill about 2000 to 3000 meters below ground surface. An approximate 6000 cubic meter volume of molten mass consisting of the radioactive waste, the canisters, and the rock would be formed because of the heat generation of the waste. The molten mixture would resolidify in 5 to 20 years as the radioactive wastes decay. Depending on the concept, the shaft leading to the cavity would be filled with a low-melting-temperature glass seal and cement after resolidification of the molten mass or after the waste containers are placed in the backfill.

The *in situ* plan would permit the creation of nationally central d waste repositories and would achieve a rapid isolation of high level radioactive wastes in an insoluble rock/waste matrix upon resolidification.

4.9.12 HLSW Form Property Values

The values for some of the properties most important to the radionuclide release processes are given in Table 4-2.

Table 4.2. Characterizing properties of waste.

		Waste Solids						
Property	Units	Salt cake	Spray calcine	Pot calcine	Rotary kiln calcine	Fluidized bed calcine	Super calcine	Coated particles
Solution rate	$\frac{mg}{m^2 sec}$	10^5 to 10^6	10 to 100	10 to 100	10 to 100	10 to 100	10^{-5} to .016	10^{-6} to 0.01
Corrosion to clad material	nm/sec	0 to 10^4	0 to 10	0 to 10	0 to 10	0 to 10	0 to 10	0 to 10
Residual nitrate and water	%	0.20 to 30	≤4.0	≤0.05	≤4.0	≤.03	0.005 to 0.05	0.005 to 0.05
Maximum processing temperature	K	310 to 720	970	695	870 to 1070	670 to 1070	1170 to 1370	1670
Ruthenium volatilized at processing	%	≤1.0	≤1.0	5 to 30	≤1.0	Insignificant	≤7.0	≤10.0
Volatility	n.a.	1000 K all Ru and Cs	1200 K all Ru and Cs	1200 K all Ru and Cs	1200 K all Ru and Cs	1200 K all Ru and Cs	1500 K all Ru and Cs	1670 K all Ru and Cs
Specific volume	$\frac{m^3}{MgU}$	0.05 to 1.0	.03 to .06	0.044 to 0.058	0.045	0.032 to 0.040	0.070	0.130
Wt% fission product oxides	Max.	≤80%	≤100%	≤90%	≤100%	≤100%	50%	≤80%
	Typical	≤80%	≤100%	≤90%	≤100%	≤100%	20%	45%
Specific area	$\frac{m^2}{Kg}$	100 to 5000	10 000 to 20 000	100 to 5000	100 to 5000	100 to 5000	10 000 to 20 000	100 to 5000
Form	N/A	Monolithic	Powder	Powder or scale	Granular	Granular	Powder	Beads
Structural quality	N/A	Crumbly	Soft and crumbly	Soft and crumbly	Soft and crumbly	Soft and crumbly	Soft and crumbly	Hard
Porosity	%	0	30 to 75	40 to 85	70 to 80	45 to 80	40 to 80	≤20.0
Density	$\frac{Kg}{m^3}$	1700	1000 to 2430	1100 to 1400	1000 to 1300	2000 to 2400	4000	1700
Coefficient of linear expansion	(K-1) x10^6	0	8.3	8.3	8.3	8.3	8.3	4.5
Thermal conductivity	$\frac{W}{mK}$	0.1 to 0.5	0.2	0.35 to 1.0	0.2	0.2 to 0.3	0.6	17
Heat capacity	$\frac{J}{KgK}$	500 to 800	650	650	650	650	670	680
Liquidus temperature	K	520	1670	1670	1670	1670	1670	2570
Transition temperature	K	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

Table 4.2. (Continued)

Waste Solids							
Phosphate glass	Borosilicate glass	Sintered process	Glass ceramic	Nepheline syenite	Metal matrix	Ion exchange	Rock melt
10^{-5} to 0.68	10^{-5} to 0.01	10^{-5} to 0.001	10^{-5} to 10^{-3}	1×10^{-5}	10^{-5} to 10^{-2}	0.001 to 0.01	10^{-7} to 0.001
0 to 10	0 to 10	0 to 10	0 to 10	0 to 10	0 to 10	0 to 10	n.a.
0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	≤10.0
1170	1270 to 1670	1235 to 1370	1270	1620	1470	1670	1000
3 to 15	≤2.0	≤10.0	≤10.0	≤10.0	≤10.0	≤10.0	≤5.0
<1500 K all Ru and Cs	<1500 K all Ru and Cs	<1370 all Ru and Cs	<1325 K all Ru and Cs	<1620 K all Ru and Cs	<1525 all Ru and Cs	<1670 K all Ru and Cs	n.a.
0.036 to 0.078	0.04 to 0.1	0.03 to 0.055	0.077	0.08	0.08	0.050	4 to 40
≤25%	≤50%	≤50%	30%	50%	≤65%	≤80%	10%
20%	20-35%	45%	25%	20%	33%	25%	5%
0.005 to 0.05	0.005 to 0.05	0.1 to 1.0	0.005 to 0.05	0.005 to 0.05	0.005 to 0.05	0.05 to 0.05	n.a.
Fractured Monolith	Fractured Monolith	Pellets	Monolithic	Monolithic	Monolithic	Mono lithic	*In situ* melt
Very hard and brittle	Very hard and brittle	Very hard	Hard and tough	Very hard and brittle	Hard and ductile	Hard and brittle	Hard
≤5.0	≤1.0	8 to 25	≤10.0	≤5.0	≤10.0	≤3.0	≤5.0
2700 to 3000	3000 to 3600	2400 to 3300	2850	3000	2400 to 5500	≤4500	2700
8 to 10	8 to 10	8.0	9.0 to 1.3	10	29	4.2	9.0 to 4.0×10^{-6}
0.8 to 1.3	0.9 to 1.3	0.7	2.2	1.2	5 to 35	1.2	1.0 to 1.5
1100 to 1200	750	1100 to 1200	1100 to 1200	1100 to 1200	350	550	1000 to 1600
820 to 1020	800 to 1600	970 to 1500	1070	850	600	2290	1320
770	870 to 970	n.a.	n.a.	900	n.a.	n.a.	n.a.

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734 356

5 MODEL DEVELOPMENT

The determination of the suitability of HLSW forms depends upon the consideration of the entire sequence of operations in handling, shipment, storage, and final isolation. Thus, potential environmental impacts — particularly the radiation dose to man — can be evaluated. Proposed methods for HLSW management under normal operating conditions result in human exposure that is zero or negligibly small. Therefore, it is necessary to conduct a systematic investigation that takes into account the possible occurrence of unexpected events, such as accidents and natural disasters. To do this, a systems model was constructed that can determine the parametric sensitivities of those factors relevant to HLSW properties and operations.

The systems model is based upon an event tree that delineates the different waste-management operations and failure mechanisms that lead to a release of radioactive material to the biosphere. The systems model can be used to compute probabilities and consequences associated with all branches of the tree and can be used to convert the results into utility functions commonly related to dose to humans. The model may also prove a valuable aid in assessing alternative policies, criteria, and standards.

5.1 FUNCTIONAL EVENT TREE AND GENERAL ASSUMPTIONS

5.1.1 Functional Event Tree

Figure 5-1 illustrates the major functional phases of the waste-management system. Not all branches of the tree are shown in order to avoid considerable replication. At the top level is the decision on the form and composition of the waste material prior to handling, storage, transportation,

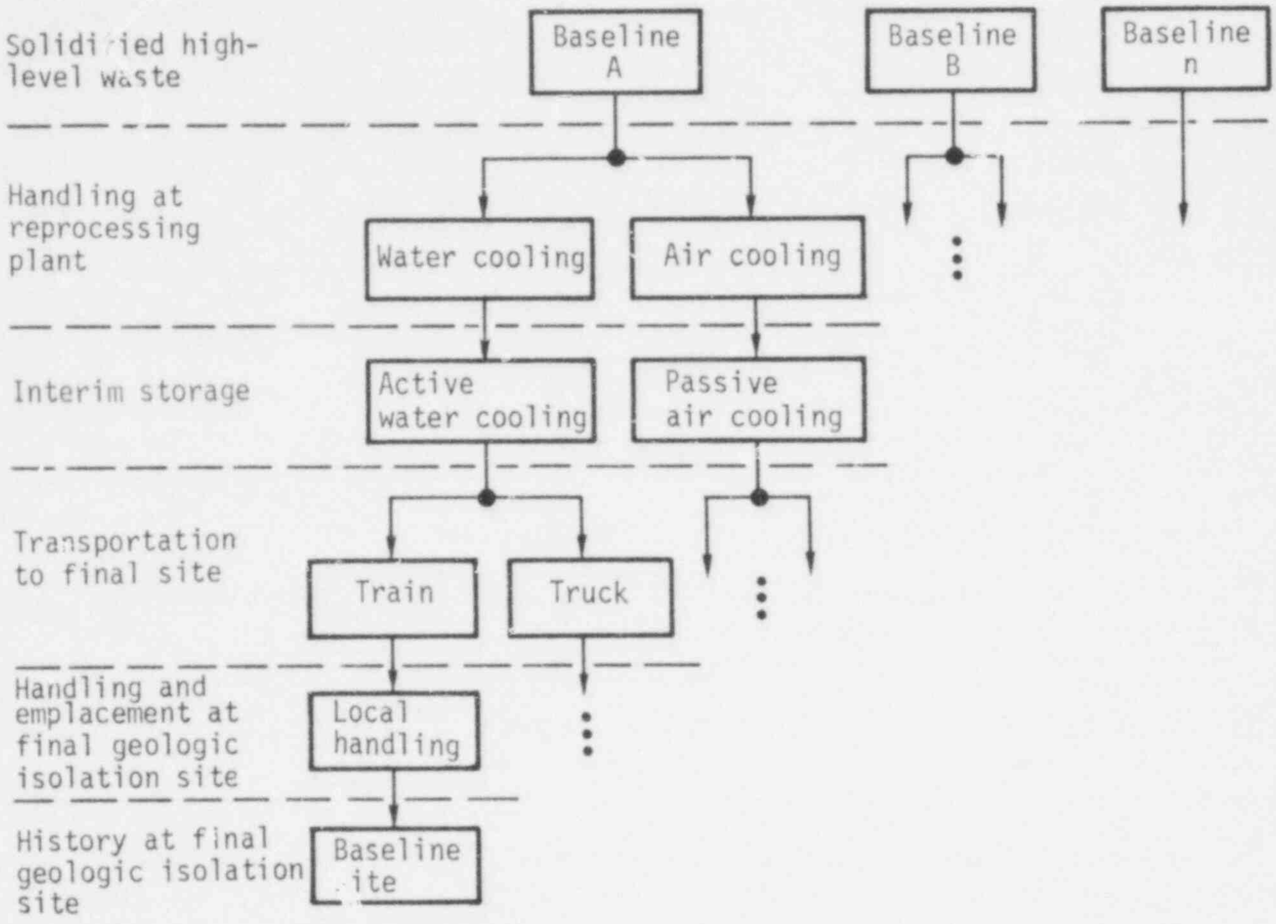


Fig. 5-1. Functional event tree.

and final disposal. Alternative forms are indicated in the figure as Baseline A, Baseline B, etc. The next level represents handling procedures to be used to move waste canisters from the reprocessing facility to the interim storage area at the fuel reprocessing plant.

This study assumes that interim storage will be accomplished in water-filled pools. Other possible storage media (e.g., air-cooling, steam-cooling), if used, would not have a significant effect on the model in this evaluation insofar as it is the loss of cooling capability that is important. Design criteria established for the storage facility will determine the relative reliability of the storage systems. Transport to the final site can involve one or multiple modes of transportation including over-water stages by barge; however, detailed consideration is restricted to train and truck transportation. Handling at the final site is included.

The behavior of the waste material after the final isolation site is sealed is examined in terms of a generic baseline site. The final site is assumed, in this report, to be in a deep continental geological formation separated from a shallow, ground-water-carrying layer (aquifer) by some form of impermeable barrier layer.

The following sample calculations illustrate the relative importance and sensitivities of each of the functional phases of the event tree shown in Fig. 5-1, in terms of expected material released and expected radiation dose to man per megawatt electric•year.

5.1.2 General Assumptions

The following assumptions pertain to the analyses in this section:

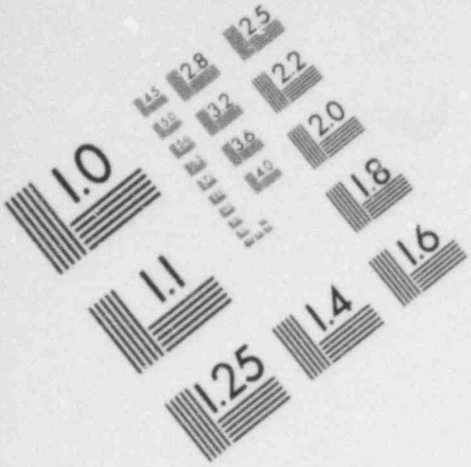
- Only HLSW from co-partitioning raffinate is considered.
- Waste from mixed-oxide fuel is not considered.

734 359

- Both uranium and plutonium are recovered (99.5%) at the fuel reprocessing plant (leaving 0.5% in the waste).
- The age of the waste handled up to the time of burial ranges from 150 days to 10 years. Specifically, 1 and 10 years are used to delineate the effects of age prior to sealing at the final geologic isolation site.
- Interim storage of HLSW is performed at the reprocessing plant site.
- "Secondary" waste streams (contaminated equipment, etc.) are not considered.
- Events up to 10^6 years after removal from the reactor are considered.
- No monitoring takes place at the final geological isolation site after emplacement.
- Two generic solidified waste forms are postulated: Baseline A and Baseline B. The former is considered to have low thermal conductivity and high solubility, particulate dispersibility, and volatility. The latter is considered to have relatively high conductivity and low solubility, particulate dispersibility, and volatility. These baseline waste forms are defined by their assumed release properties under specified environments at each node in the event tree.

5.2 HANDLING AT THE FUEL REPROCESSING PLANT

This section describes the calculations that determine the effect of accidental releases of high-level nuclear wastes during handling at the fuel reprocessing plant (FRP). The accidents postulated are representative of those that can occur when canisters of solidified waste are stored in a pool of water. Operations such as canister sealing, decontamination, and testing



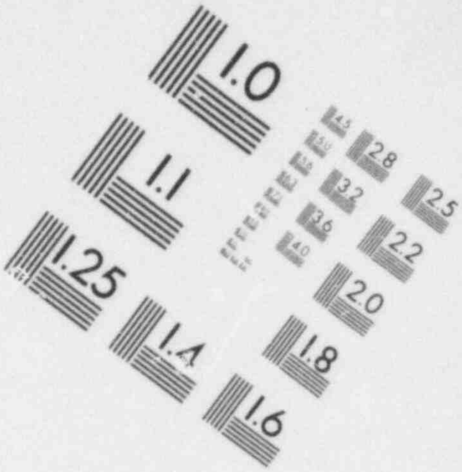
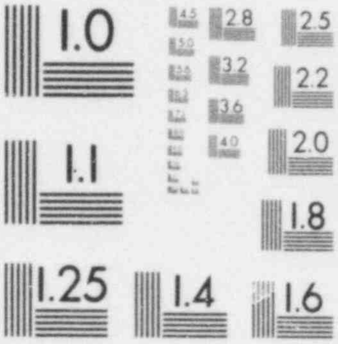
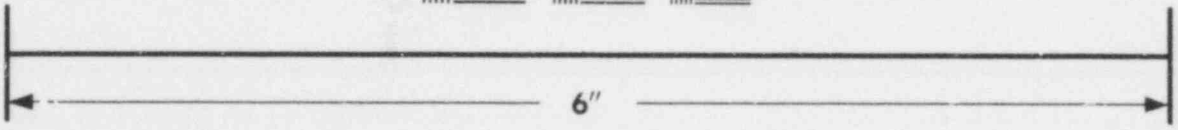
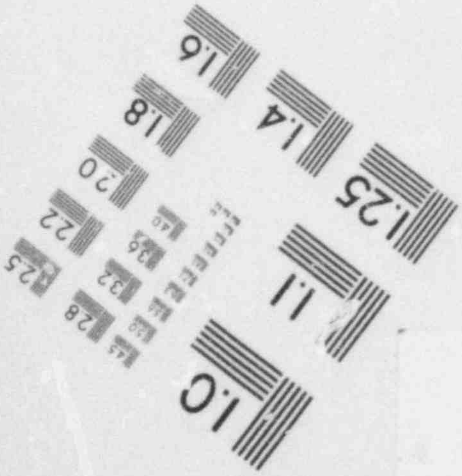
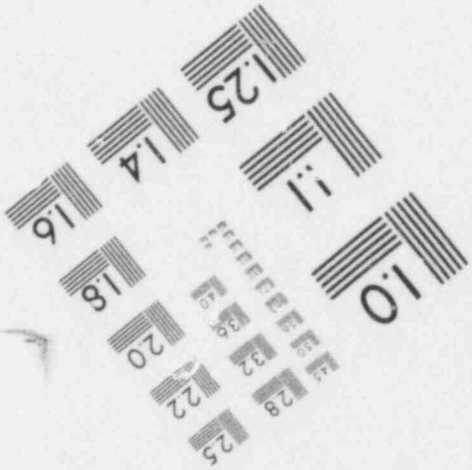


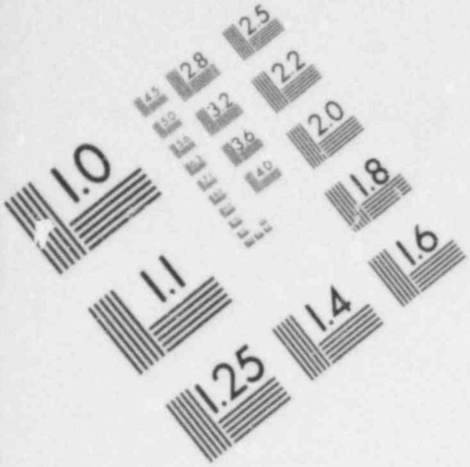
IMAGE EVALUATION TEST TARGET (MT-3)

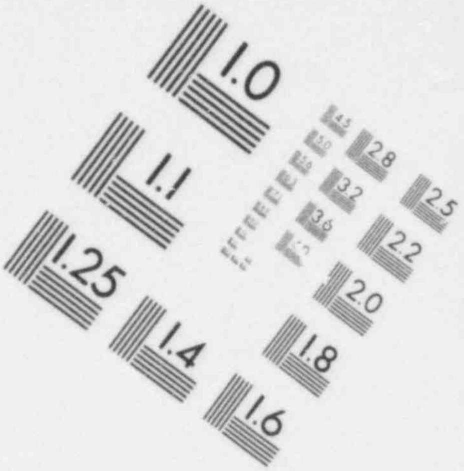












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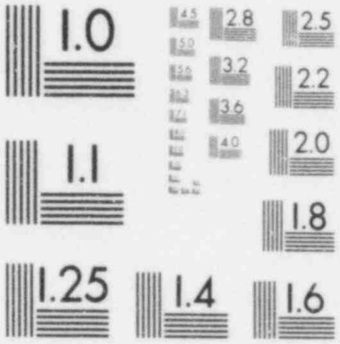

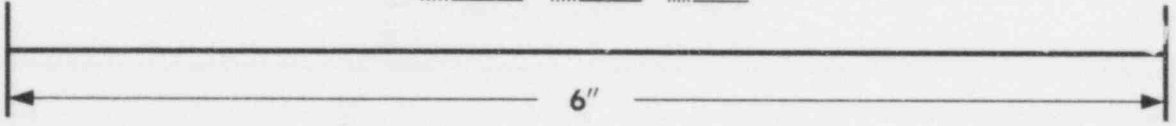

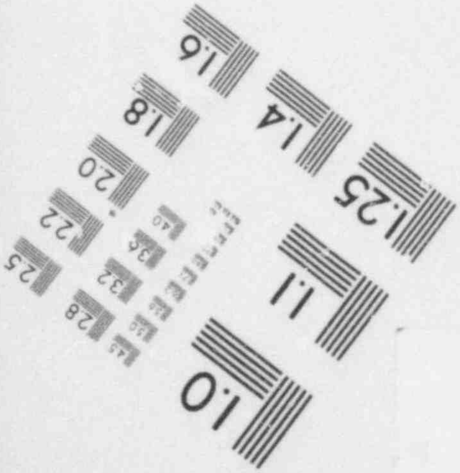

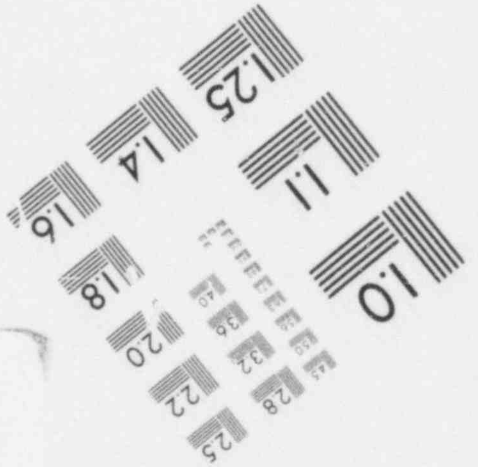

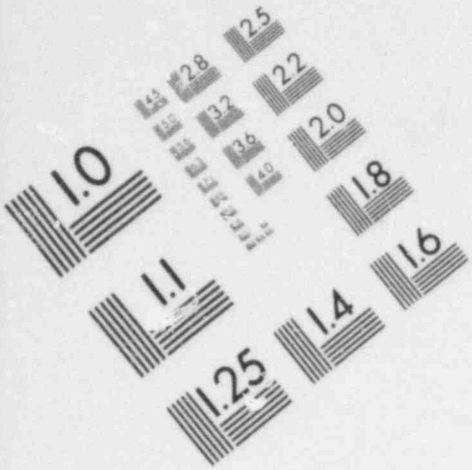

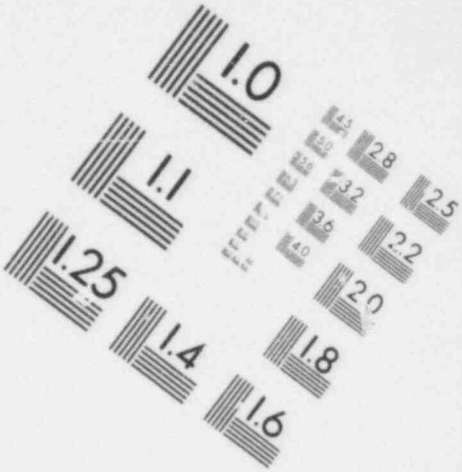

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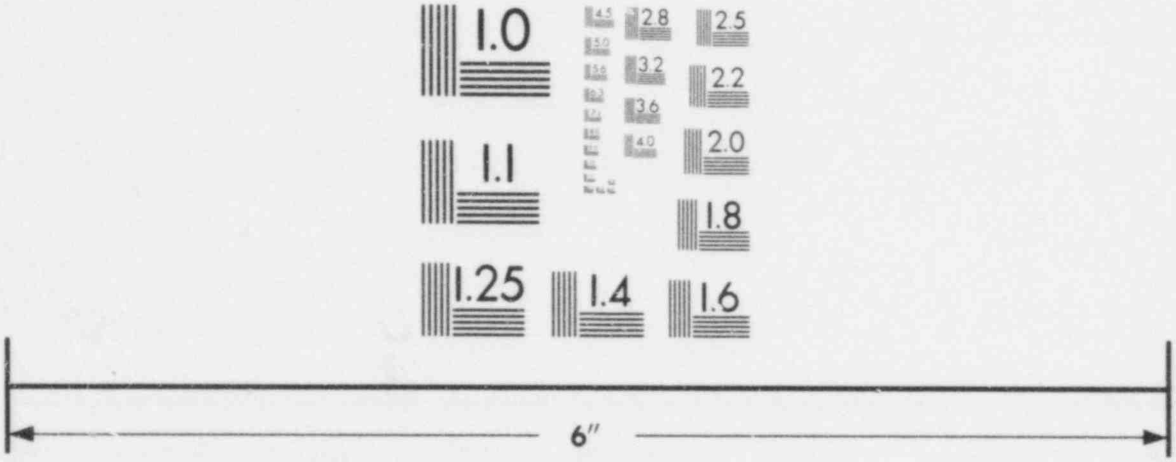

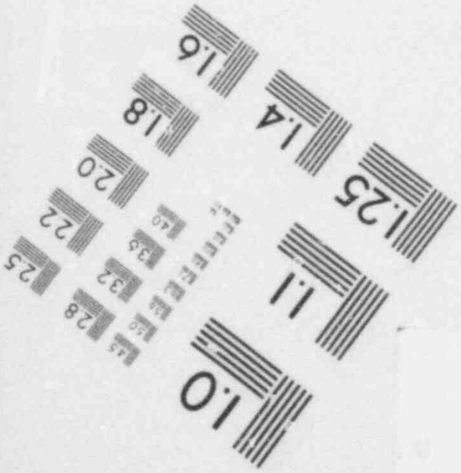

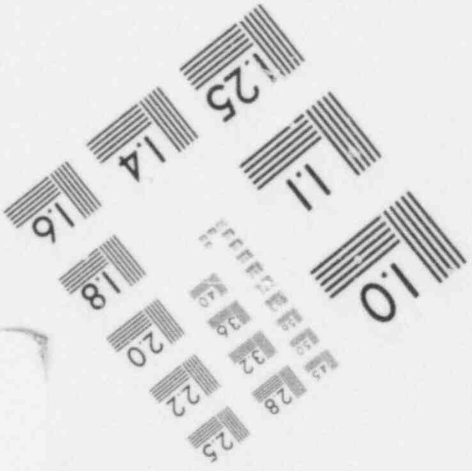

are not considered because they are conducted in-cell. The section concludes with a set of calculations demonstrating the relative unimportance of handling accidents at the FRP.

5.2.1 Assumptions and Sources of Data

The following assumptions apply to the analytic event tree for handling at the interim storage site. *Many of these assumptions are also key to the event tree for accidents occurring while the waste is in storage* (see Sec. 5.3). They are not repeated in Sec. 5.3.

- Nominal canister size is 30 centimeters in diameter by 3 meters long. This will hold 0.2 cubic meter of 10-year-old waste from 3.33 megagrams of uranium in LWR fuel. Exposure is assumed to be 33 000 megawatt•days per megagram of uranium at 35.4 percent efficiency. One canister is assumed to hold the waste for 100 megawatt electric•years of electrical energy generation.
- The composition of the HLSW, in terms of biologically significant nuclides, is presented and discussed in Appendix A.
- In considering the impact of thermal conductivity, time since removal from the reactor, and volume of waste generated, a basic first-order linearity is used in the model. Specifically, if twice as many canisters, rail cars, etc., are needed to handle the waste from 1 megawatt electric•year because of a driving parameter, then each canister will have half as much radioactive material (for a given time since removal from the reactor). Therefore, while the probability of an accident doubles, the maximum radioactive material *available for release in an accident* halves. The most significant factors are those characteristics that determine the *percent of available radioactivity released* in a given accident mode.

- All operations in interim water storage of HLSW take place within a building that is sealed under normal circumstances.
- A transfer cask will be used to protect the canister during most movement out of water. The cask is capable of protecting the canister from overheating and from damage due to drops. The only time the canister is in danger is when it is outside the cask.
- Equipment will be arranged so that the transfer cask cannot be moved over the water storage basin, which precludes dropping the cask into the basin.
- Release does not occur if a canister dropped from the crane lands in the pool.
- The most significant mechanism for release into the sealed building is that of a canister failure caused by overheating.
- The functioning air filtration system passes 10^{-11} of all volatiles released from canister accidents within the plant.
- The probability of filtration-system failure is 10^{-6} per year. This is identical to the HEPA failure rate given in the U.S. NRC study of reactor safety (1975). The probability that filter failure will allow release of volatile materials from an accident within the sealed building is based on the probability that the filter will fail within one week after the accident ($1/52 \times 10^{-6}$). One week is required to clean up failed canisters.
- If the filter fails within one week of an accident, 1 percent of the volatiles released within the building will escape into the atmosphere. The rest will condense inside the building.
- Nominal probability of crane drop is 3×10^{-6} per hour of operation (from U.S. NRC reactor safety study [1975]).

735 002

- Nominal probability of crane stall is 1.5×10^{-6} per hour of operation.*
- Nominal time spent handling each canister in air outside the cask is 20 minutes. This includes time for insertion into the pool, retrieval, and insertion into a transportation cask.
- Given a crane stall or drop outside the pool, the probability of overheat to canister failure is 0.5 for 1-year-old Baseline A waste, zero for 1-year-old Baseline B waste, and zero for all 10-year-old waste.

5.2.2 Event Tree

Figure 5-2 shows the event tree used to represent major accidental release modes during HLSW handling at the water storage site. Consistent with the assumptions stated in Sec. 5.2.1, crane stall and crane drop when the canister is not over water are the two major accident modes.

5.2.3 Calculation of Release per Megawatt Electric•Year

The assumptions and probabilities stated in Sec. 5.2.1 make possible the computation of expected values of waste released per megawatt electric•year. *Note that no 10-year-old waste is released because the probability of canister breach is zero for that condition.* In these calculations, it is assumed that 100 percent of the volatiles available (specifically Cs, Ru, and Te) are released into the building that houses the pool. This very conservative assumption is made because the expected release from handling accidents at the interim storage site is insignificant relative to expected releases from other portions of the waste-disposal tree. Consequently, the development of release

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*Personal communication, Mr. Norman Smith, Materials Handling Institute.

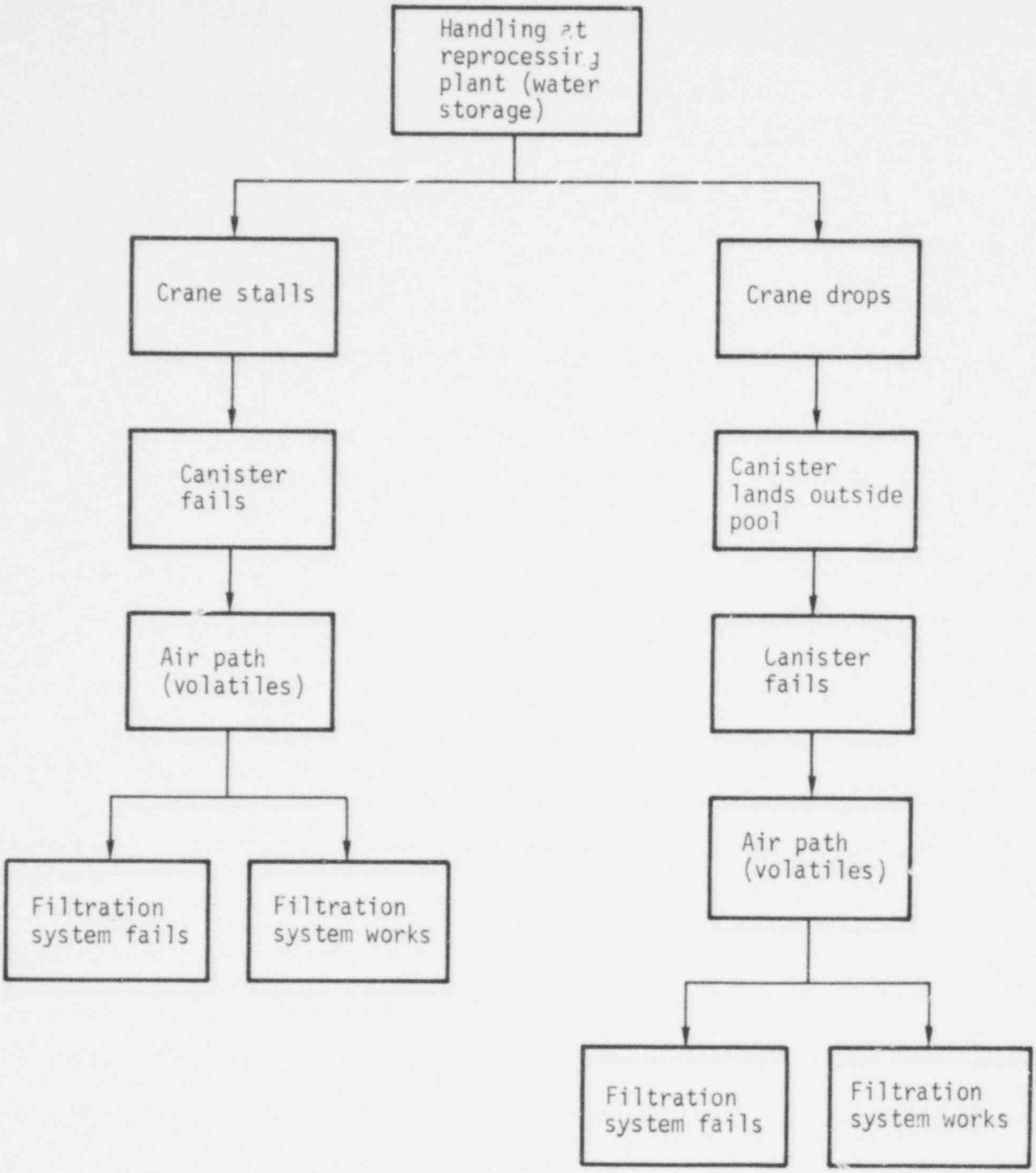

Fig. 5-2. Event tree for accidental release during handling at water storage site.

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fractions (those fractions of available material actually released in a given accident) is unnecessary, as demonstrated in the following paragraphs.

Crane Stall — Progressing down the left side of the event tree in Fig. 5-2, the probability of a crane-stall accident per canister of solidified waste is

$$\left(1.5 \times 10^{-6} \; \frac{\text{stalls}}{\text{hr of operation}}\right)\left(\frac{\text{1 hr operation}}{\text{3 can}}\right) = 5 \times 10^{-7} \; \frac{\text{stalls}}{\text{can}} .$$

Each canister contains 100 megawatt electric•years of solid waste. Therefore, there are 5×10^{-9} stalls per megawatt electric•year. Two possibilities ensue: If the filtration system works for one week after the accident, 10^{-11} of the amount released will enter the atmosphere. Assuming 100 percent of the volatiles available are released, the expected amount released per megawatt electric•year is 5×10^{-20} times the activity in one canister (due to 100 MWe•yr of waste).* If the filtration system fails in the week following the accident, 10^{-2} of the available material will be released. The probability of filter failure is

$$\left(10^{-6} \; \frac{1}{\text{yr}}\right) \left(\frac{\text{1 yr}}{\text{52 wk}}\right) = 1.92 \times 10^{-8} \; \frac{1}{\text{wk}} .$$

The expected amount released due to crane-stall accidents followed by filter failure (1-yr-old waste only), then, is

$$\left(5 \times 10^{-9}\right) \left(10^{-2}\right) \left(1.92 \times 10^{-8}\right) = 9.6 \times 10^{-19} \; \frac{1}{\text{MWe•yr}}$$

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*Strictly speaking, this must be multiplied by one minus the probability of filter failure. To a very good approximation, the number is unity.

times the activity in one canister. Note that this is approximately 20 times the expected release from crane-stall accidents where the filter does not fail.

The net expected release for a crane-stall accident is the sum of the above two numbers (i.e., 10^{-18} times the activity of the contents of one nominal canister). *This is small relative to releases expected for storage and transportation accidents.* Differences in population distribution used in dose calculations do not alter this conclusion.

Crane Drop — A similar sequence of calculations yields the expected release for a crane-drop accident. The probability of a crane drop is twice that of a stall, while the probability of a canister failure given a drop is half that of a stall; all other numbers in the calculation are the same. Consequently, the expected release due to a crane-drop accident is the same as that for a crane-stall accident: 10^{-18} times the activity of the contents of one nominal canister.

Handling accidents at the interim storage site do not significantly contribute to the expected dose from high-level nuclear waste disposal.

5.3 INTERIM HLSW STORAGE AT THE FUEL REPROCESSING PLANT

This section describes the calculations that determine the results of accidental releases of high-level waste during storage in a pool of water. The wastes are assumed to be cooled by water conduction and convection. The water, in turn, is assumed to be cooled by an external mechanism, such as a cooling tower. Active cooling of the water is required for normal operation. All probabilities computed in this section are stated *per year of storage* and must be multiplied by the assumed period of storage.

5.3.1 Assumptions and Sources of Data

The following assumptions apply to the analytic event tree for interim storage. *These assumptions are in addition to those already stated in Sec. 5.2.*

- Multiple canisters are assumed to be stored in each pool. The exact number per pool is not specified but is assumed to be sufficiently large to expect a fraction of the canisters to fail when cooling water is absent for a long time. The mechanism for failure is assumed to involve co-heating by thermal radiation in the empty pool.
- When absence of water is caused by loss of circulation, the canisters fail with probability 1 for 1-year-old Baseline A waste, 0.5 for 1-year-old Baseline B waste, and zero for 10-year-old waste. When absence of water is caused by pool drainage due to a failure of the pool structure, the corresponding probabilities are 1, 0.6, and 0.1, respectively. These probabilities are higher because the event causing the pool structure to fail can rearrange the canisters and aggravate the self-heating process.
- For the drainage-of-storage-pool accident followed by canister failure, it is assumed that 0.1 of the activity of Cs, Ru, and Te is released as volatiles. For Baseline A, 0.01 of the total activity is released as dispersable particulates. For Baseline B, 10^{-7} of the total activity is released as dispersable particulates.
- Tornados and hurricanes are assumed to be no danger to the pool, which is below grade and strong enough to withstand impacts of missiles from these causes. Meteors are taken to be extraordinary occurrences of too small a probability to be significant.

- Earthquakes and aircraft crashes are the most serious probable causes of cooling-pool drainage. In both cases, the event causing drainage will also rupture the air seal that prevents release into the atmosphere.
- For an earthquake, it is assumed that there is no restoration of water cooling (in any form) before the canisters melt.
- For an aircraft accident, the probability of restoration of water cooling (e.g., hoses played on the waste) before the canisters melt is assumed to be 0.9. This reflects the fact that the aircraft accident is a local phenomenon, leaving water supplies intact.
- The water pathway for release into the environment is ignored since the consequences are minimal as long as there is a reasonable distance from the pool to the nearest surface water body. Also, the possibility for cleanup exists.
- When the sealed building enclosing the pool remains intact, the filter system characteristics and probabilities stated in Sec. 5.2.1 are in effect.
- The nominal probability of a loss of active cooling of the pool water is 10^{-1} per year (U.S. NRC reactor safety study [1975]). The probability that loss of cooling cannot be corrected before the contents of the pool boil off is 10^{-6} (U.S. NRC, [1975]).
- The density of aircraft accidents is computed from U.S. NRC (1975), which states that 10^{-6} to 10^{-7} crashes are expected per year within a 5-mile radius of airports. Allowing for the fact that the interim storage site will be far from busy airports, a frequency of 10^{-8} per year in a 5-mile radius is used to calculate a density of 1.27×10^{-10} crashes per year•square mile.

- A nominal storage pool is assumed to present an area of 2500 square yards or 8.07×10^{-4} square miles to the possibility of an aircraft crash.
- The probability of having a severe earthquake (see Sec. 5.6 for definition away from tectonic plate boundaries is 10^{-5} per year (Schneider and Platt, 1974).
- There is a 0.1 probability of pool drainage resulting from a severe earthquake since the storage pool is designed to withstand these.
- An aircraft crash may breach some of the canisters in the pool. However, the major mechanism for release is pool drainage. The fire from the aircraft's fuel extinguishes itself before the pool empties enough to allow release of dispersible waste fractions.
- Meteorological and demographic assumptions are given in Appendix D. Dose-conversion factors are presented in Appendix B.

5.3.2 Event Tree

Figure 5-3 shows the event tree for major modes of accidental release of nuclear waste from interim storage in water.

5.3.3 Calculation of Release per Megawatt Electric•Year

Loss of Cooling Circulation — A calculation following the left side of the event tree is useful to show that the loss-of-cooling-circulation accident is not a major contributor to the total expected dose from waste-disposal operations.

Using the probabilities stated in the assumptions in Sec. 5.3.1, the expected frequency of occurrence of loss of cooling circulation and boil-off of the cooling water is $(10^{-1})(10^{-6})$ liters per year = 10^{-7} liters per year. Given a boil-off, 1-year-old Baseline A waste will overheat to the point of

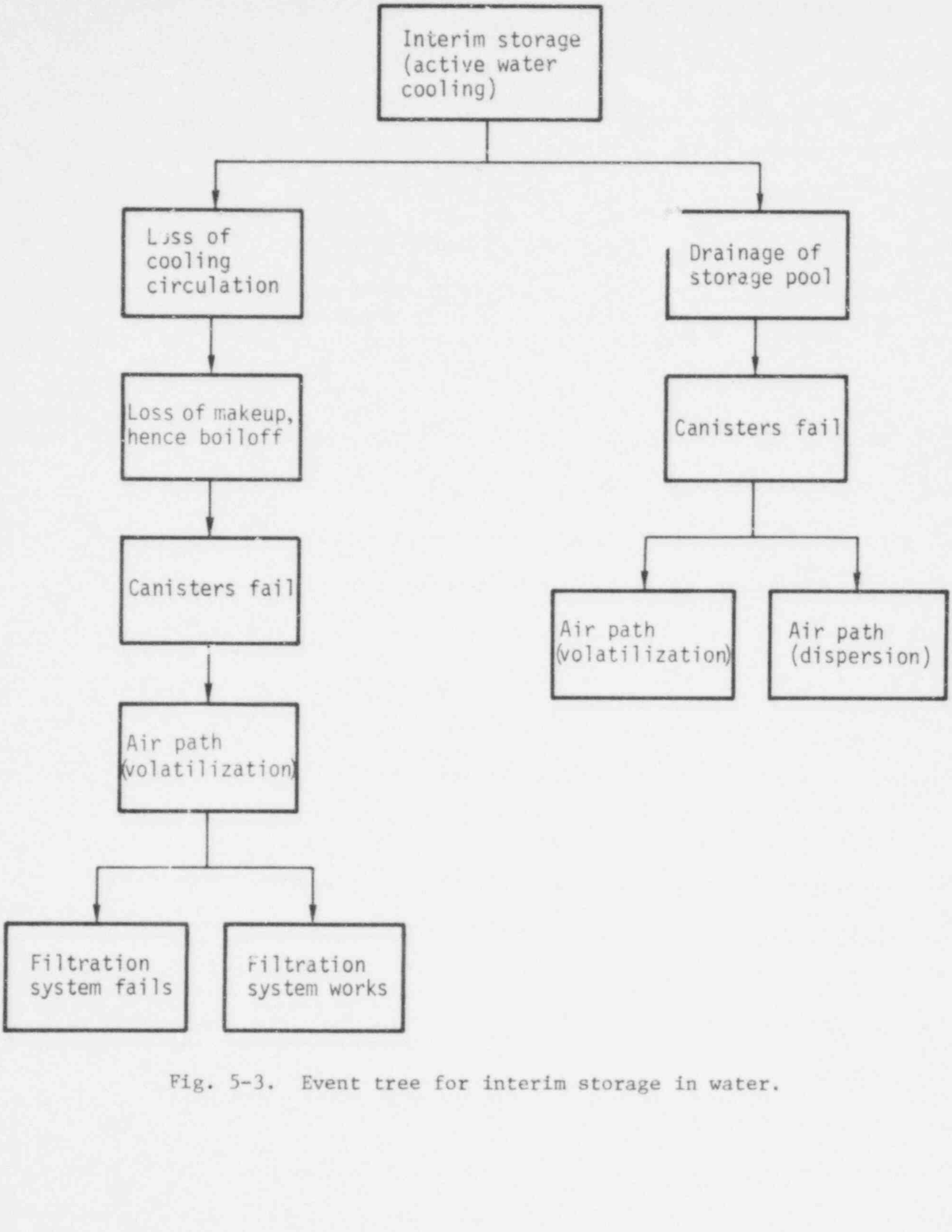

Fig. 5-3. Event tree for interim storage in water.

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canister failure and volatile release of Cs, Ru, and Te. The expected activity within the building enclosing the pool is 10^{-7} of the activity available in the volatiles generated per megawatt electric•year equivalent of waste stored, per year of storage. If the filtration system works, the above activity is reduced by 11 orders of magnitude before entering the atmosphere. The probability of the filtration system failing in the week following boil-off is 1.92×10^{-8} and, if the system fails, 10^{-2} of the available volatiles will be released into the atmosphere. The expected release from the accident sequence — circulation failure, boil-off, filter failure in one week — is 1.92×10^{-17} of the volatiles generated per megawatt electric•year of waste stored, per year of storage. This adds with 10^{-18} for the case when the filter does not fail. The maximum anticipated storage period (10 yr) will give an expected release from this mode of 2×10^{-16} of the volatiles available from 1 megawatt electric•year, per megawatt electric•year. (Recall that all these numbers assume a 100% release fraction.)

Drainage of the Storage Pool — Accidents involving drainage of the interim storage pool are a major contributor to the expected dose from solidified-waste-disposal activities. Also, because such an accident typically can involve a large amount of waste, it can therefore provide high individual doses to people in the vicinity. As stated previously, two types of accidents are considered possible contributors to this release mode: earthquakes and aircraft crashes. The aircraft-crash density and pool area described in the above assumptions are used to compute an expected aircraft-accident frequency as follows:

$$\left(1.27 \times 10^{-10}\ \frac{\text{crashes}}{\text{yr}\cdot\text{mi}^2}\right)\left(8.07 \times 10^{-4}\ \text{mi}^2\right) = 1.02 \times 10^{-13}\ \frac{\text{crashes}}{\text{yr}} .$$

Consequently, aircraft crash is of negligible significance compared to seismic events as a cause of pool drainage.

The maximum expected fraction of waste that will be released per megawatt electric•year, per year, is 10 . This number must be multiplied by the release fractions given in Sec. 5.3.1; the duration of storage; and the appropriate activity levels (depending on the age or ages of the waste assumed to be in the cooling pool) to obtain the expected amount of activity released per megawatt electric•year.

5.4 TRANSPORTATION

Two modes of transportation were given primary consideration in computing probable releases of radioactive materials and the resultant doses: train and truck. Other means of transportation (e.g., barges) can be considered at a later time.

The current investigation relied on the work of Clarke *et al.* (1975), with consideration of the fact that large transportation casks rather than small packages are of primary interest in this study. Conversations with personnel at Sandia and Battelle Pacific Northwest Laboratories, analysis of the NRC-NUREG-0034 draft environmental statement on transporting radioactive materials, and evaluation of the work of Brobst (1972) and Langhaar (1976) were used as supplementary sources of data.

5.4.1 Assumptions and Sources of Data

General — The following assumptions pertain to both train and truck accidents:

- No special transportation procedures are followed. Accident statistics for normal freight operations apply.
- Crush is not a factor for the massive transportation casks involved. Impact, fire, and puncture are considered (Clarke *et al.*, 1975).
- Immersion is considered an extraordinary event (U.S. NRC reactor safety study, 1975) and therefore of low probability.
- Transportation casks are designed to meet "Type B" standards (NRC Regulations, 10 CFR, Part 71; and IAEA Safety Series No. 6, 1976). Specifically: impact, 30-foot drop onto unyielding surface; fire, 44 250 °F•min; puncture, V/R of 58 $\sec^{-1}$.
- Canisters offer inconsequential protection in transportation accidents relative to transportation casks. Therefore, if the cask is breached, the canisters are assumed breached.
- The transportation casks are assumed to rely on passive convection air cooling to maintain satisfactory temperatures in the waste canisters.
- The nominal transportation distance from interim storage to the final disposal site is 2000 miles.
- Transportation accidents are assumed to take place with equal probability in urban and rural areas.
- Given an accident, a water pathway (rain, ditch, stream, or river) is present 1 percent of the time. For fire accidents, water played on the fire will prevent the time-temperature product from reaching the value at which release occurs.
- Given an impact or puncture accident, an air-dispersion energy source is present 10 percent of the time.
- An air-dispersion energy source (firestorm) is always present for a fire of size sufficient to cause release from the cask.

735 013

Train — The following assumptions and sources of data pertain specifically to train accidents:

- The rail transportation cask will hold nine canisters.
- The frequency of rail accidents is given by Clarke *et al.* (1975) as 1.37×10^{-6} car accidents per car•mile.
- Because of speed limits, no urban train accidents occur at speeds above 40 mph. Half of all accidents below 40 mph occur in urban areas.
- Only impacts with "extremely rigid" fixed objects will impose sufficient energy to breach the cask (Clarke *et al.*, 1975). The nominal probability of these accidents is computed from Clarke *et al.* (1975) as 0.051 per car•accident.
- In an impact accident, the cask has the same velocity as the train and all of the cask's resultant kinetic energy is dissipated on the cask.
- The velocity distributions for fixed-object collisions are taken from Table III, Vol. IV of Clarke *et al.* (1975).
- The probability of a fire for a rail accident is 0.059 (Clarke *et al.*, 1975). This is modified downward by a factor of 0.625 since no cargo is transported in a car carrying a cask.*
- The probability of puncture encounter for a train accident is 0.546 (Clarke *et al.*, 1975), and the probability distribution for puncture accident severity is given in Fig. 28 (Assumption B) of Vol. IV (Clarke *et al.*, 1975).
- A rail puncture accident involves two of the nine canisters in the cask.

Truck — The following assumptions and sources of data pertain specifically to truck accidents:

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*Personel communication with A. Dennis, Sandia Laboratories, Albuquerque, New Mexico (1976).

- The truck-transportation cask will hold two canisters.
- Only impact with "extremely rigid" fixed objects and accidents with trains at crossings will impose sufficient energy to breach the cask. Probabilities for these impact events are 0.051 and 0.0062 (Clarke *et al.*, 1975).
- The probability of a crossing accident is reduced to 0.0031 since major energy is dissipated on the cask only when the locomotive frame hits it.*
- In impact accidents, all of the energy provided by the truck (train) velocity is dissipated on the cask.
- The velocity distributions for fixed-object collisions and crossing accidents are given by Clarke *et al.* (1975) in Fig. 4, Vol. III, and Table II, Vol. IV, respectively.
- The probability of a fire in a truck accident is 0.016 (Clarke *et al.*, 1975). This probability is reduced by a factor of 0.625 since no other cargo is carried.*
- The probability of a puncture encounter in a truck accident is 0.2, as computed from information given on page 80 of Vol. III (Clarke *et al.*, 1975). The probability distribution for severity is given in Fig. 45, Vol. III, of the same reference.

A truck puncture accident involves one of the two canisters in the cask. Use of the above assumptions, probabilities, and probability distributions will be explained further in the following sections.

5.4.2 Event Tree

Train — Figure 5-4 shows the event tree used to represent release of nuclear waste materials from train accidents. The three major transportation

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*Telephone conversation with A. Dennis, Sandia Laboratories, Albuquerque, New Mexico (1976).

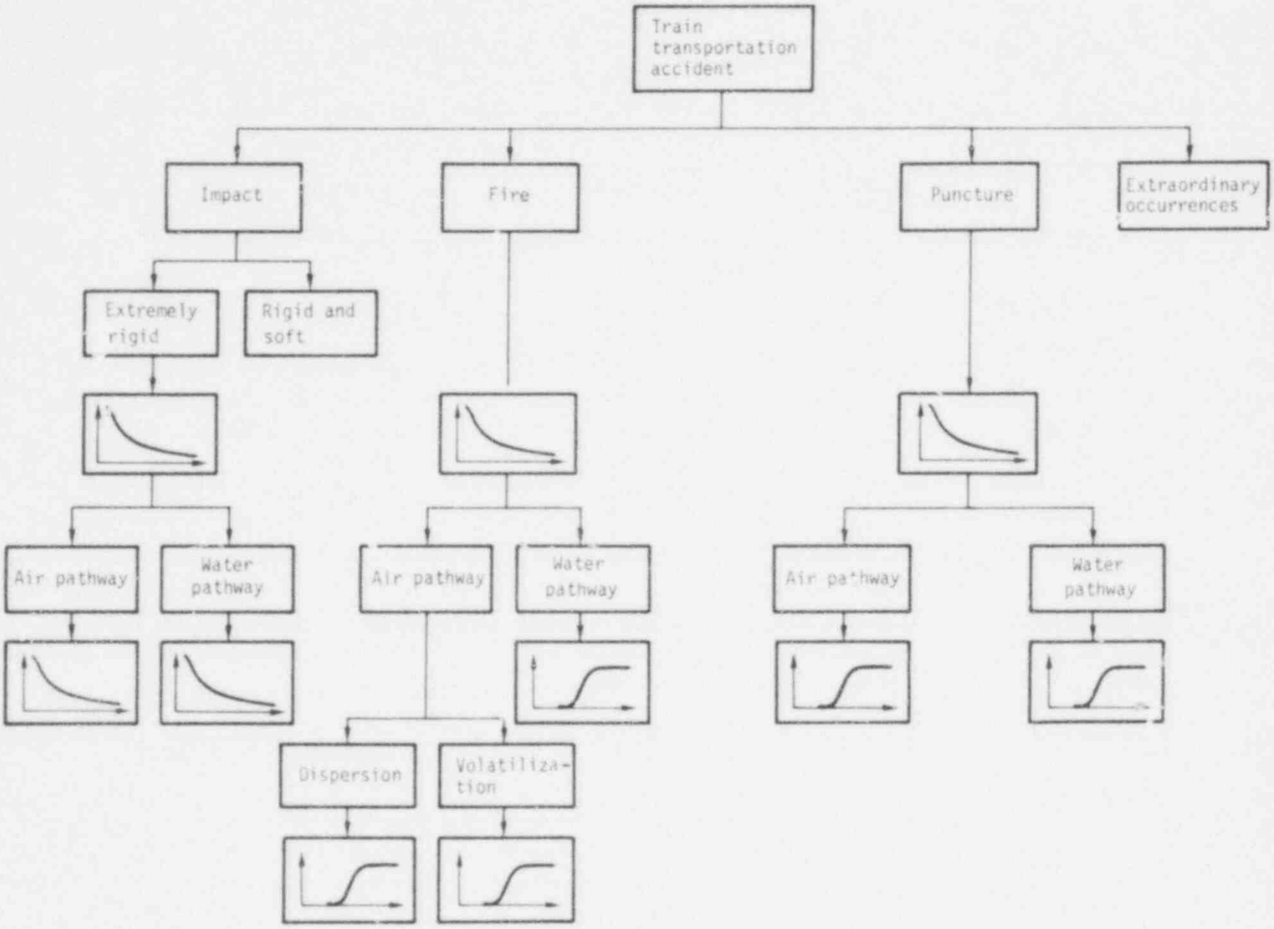

Fig. 5-4. Event tree for train-transportation accident.

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accident modes — impact, fire, and puncture — are shown, along with a fourth category, extraordinary occurrences. The probability of the latter mode is defined as negligible.

Only collision with extremely rigid objects is considered, as stated in the preceding set of assumptions. A probability density function (pdf) for accidents was derived from Clarke *et al.*, 1975. Figure 5-4 represents the pdf as a continuous function of velocity; the representation in the figure is conceptual in that the available data only permit calculation of constant pdfs over 10-mph intervals.

Below the pdf for velocity of impact with extremely rigid objects is a representation of the weighting of the amount of material released as a function of accident severity. The Type B cask is designed to withstand a 30-foot drop onto an unyielding surface. This is equivalent to a 30-mph collision with an unyielding surface. The *release functions* used in this study do not represent any release for collisions at velocities below the cask design value. A typical release function (shown in Fig. 5-4) allows the fraction of material to grow to a maximum value at 90 mph. The function is represented analytically by

Fraction of maximum release	Velocity (mph)
0	$V \le 30$
$\frac{1}{2}\left(1-\cos\frac{V-30}{60}\pi\right)$	$30 < V < 90$
1	$90 \le V$

The maximum release fraction is a function of the characteristics of the solid-waste form, the canister, and the cask. It can be specified in terms of a constant, which multiplies the values computed from the event tree.

The *shape* of all the release functions used in analyzing transportation accidents is the same as that discussed above. Only the *end points* of the curve change with accident mode and transportation mode (truck or train). Table 5-1 gives the end points for the S-curves of release functions. The values are estimates; Type B standards are used as the minima.

Figure 5-4 distinguishes the major pathways for the released radioactive material. The primary distinction is between water and air pathways. For the fire accident, the latter is further separated into dispersion and volatization modes of release because the release function used can be different for these two pathways.

Truck — Figure 5-5 shows the event tree used to analyze release of nuclear waste materials from truck accidents. In accordance with the assumptions stated above, the structure of this tree differs from that for the rail

Table 5-1. Release-function end points.

Accident mode	Train		Truck	
	Minimum	Maximum	Minimum	Maximum
Impact (mph)				
Air path (dispersion)	30	90	30	90
Water path	30	90	30	90
Fire (°F•min)				
Air path (dispersion)	44 250	200 000	44 250	200 000
Air path (volatiles)	44 250	800 000	44 250	200 000
Water	44 250	640 000	44 250	320 000
Puncture (sec^{-1})				
Air path (dispersion)	58	116	58	116
Water path	58	116	58	116

Release-function equation:

$$\text{Release function} = \begin{cases} 0 & ; x \le x_{min} \\ \frac{1}{2}\left[1 - \cos\left(\frac{x-x_{min}}{x_{max}-x_{min}}\right)\pi\right] & ; x_{min} < x < x_{max} \\ 1 & ; x_{max} \le x \end{cases}$$

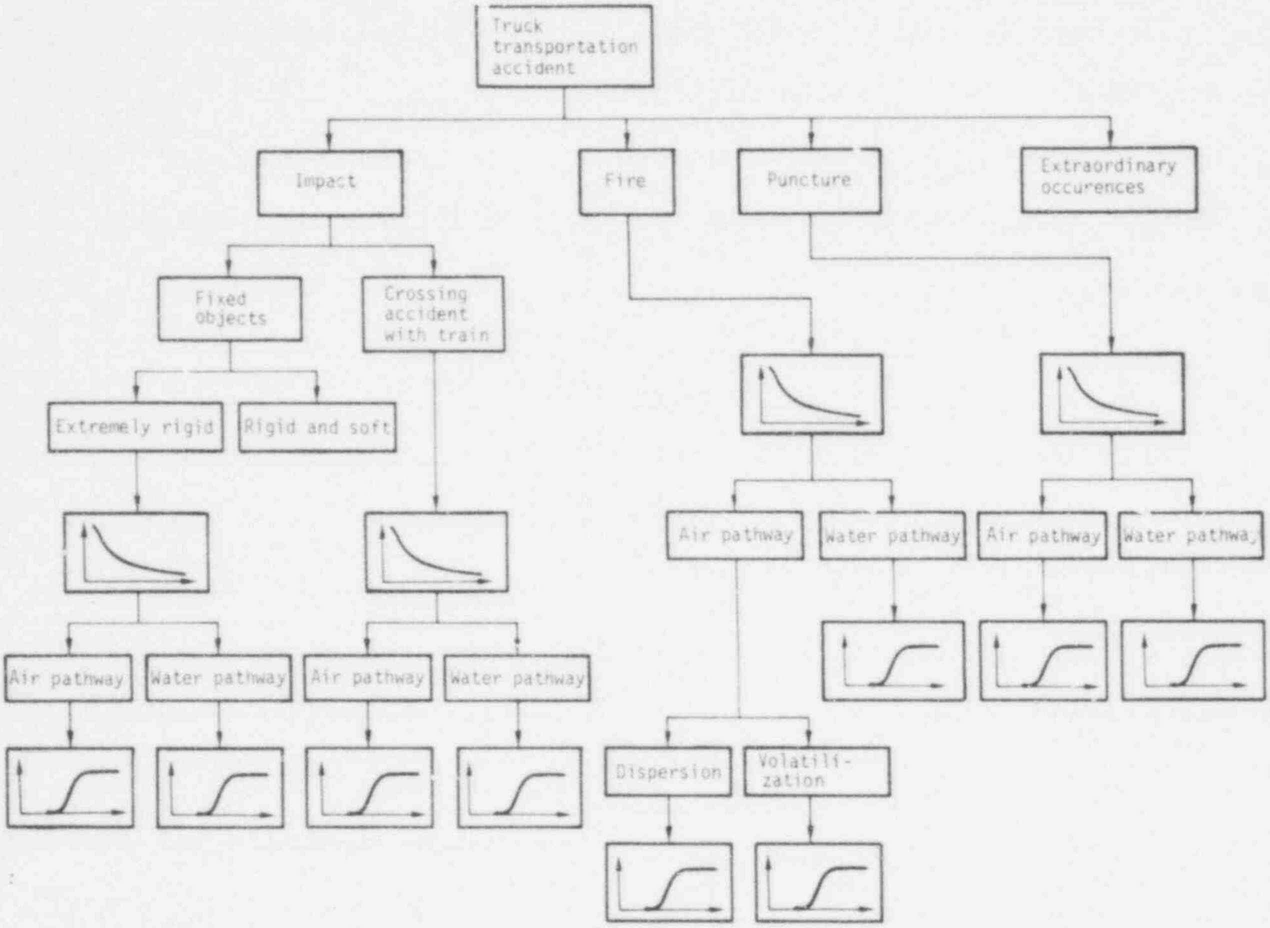

Fig. 5-5. Event e for truck-transportation accident.

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accident primarily in describing the impact mode. Specifically, the crossing accident, in which the truck is hit by a train, is added to this portion of the tree. Release functions are given in Table 5-1. In several cases, the upper ends of the S-curve are at values of the temperature-time product lower than those for the train accident. This reflects the fact that the rail-transportation cask is considerably more massive than that used with the truck, and, therefore, the former can withstand a considerably larger heat input before being affected enough to permit release.

The transportation-accident event trees, when combined with numbers for the maximum release fraction, permit calculation of expected release per megawatt electric-year.

5.4.3 Nominal Maximum Release Fractions

The choice of maximum release fractions defines Baselines A and B. They are selected to be in the likely range for candidate waste forms.

Table 5-2 gives the nominal maximum release fractions used in studying the effect of transportation accidents. The numbers reflect the characteristics of the Baseline A and B solids in terms of particle size, volatility, and solubility in water. They also represent the assumed probabilities of the existence of air and water pathways, which were stated in the preceding section, and an assumption that the solid waste is available for leaching for one full day for the water pathway.

The release of radioactive materials will be analyzed in more detail in the subsequent studies to determine the effect of accidents on casks and canisters.

5.4.4 Sample Calculations

To illustrate the calculations yielding the expected value of the number of curies of a particular nuclide released per megawatt electric·year, we will

Table 5-2. Nominal maximum release fractions: transportation accidents.

Accident		Maximum fraction available	
		Baseline A	Baseline B
Impact	Air (dispersion)	10^{-2}	10^{-6}
	Water	3×10^{-2}	3×10^{-7}
Fire	Air (dispersion)	10^{-2}	10^{-8}
	Air (volatiles)	10^{-2}	3×10^{-3}
	Water	3×10^{-2}	10^{-7}
Puncture	Air (dispersion)	10^{-3}	10^{-7}
	Water	3×10^{-3}	3×10^{-8}

concentrate on the release of Baseline A waste into the air in an urban rail-accident impact mode. The air-dispersion pathway for particulates from 10-year-old waste will be assumed.

If a canister holding 100 megawatt electric·years of waste and a rail-transportation cask holding nine canisters are used, each rail car will hold 900 megawatt electric·years of waste. If the transportation distance is 2000 miles, the number of car·miles per megawatt electric·year is computed as follows:

$$\left(\frac{1\ \text{car}}{900\ \text{MWe}\cdot\text{yr}}\right)\ (2000\ \text{mi}) = 2.22\ \frac{\text{car}\cdot\text{mi}}{\text{MWe}\cdot\text{yr}}\ .$$

Accident frequency is 1.37×10^{-6} car accidents per car·mile. Therefore, the expected number of rail car accidents per megawatt electric·year is computed:

$$\left(1.37 \times 10^{-6}\ \frac{\text{car accidents}}{\text{car·mi}}\right)\left(2.22\ \frac{\text{car·mi}}{\text{MWe·yr}}\right) = 3 \times 10^{-6}\ \frac{\text{car accidents}}{\text{MWe·yr}}$$

The integral of the product of the accident-severity probability-density function and the release function (see Fig. 5-4) for the impact accident is 9.23×10^{-4}. The probability of an impact with an extremely rigid object, given the occurrence of a rail accident, is 5.1×10^{-2}. Multiplying the expected number of car accidents per megawatt electric•year by these two numbers yields 1.41×10^{-10}, the expected number of car accidents per megawatt electric•year that involve urban impact on an extremely rigid object, weighted by the accident severity and the release function. To compute the expected amount of material released, one must multiply this number by the appropriate maximum release fraction (10^{-2}, in this example), the curies of individual nuclide present per megawatt electric•year (1.9×10^{3} for ^{90}Sr), and the amount of material carried on a single rail car, using the canister and cask designs. Thus, the expected release per megawatt electric•year is

$$\left(1.41 \times 10^{-10}\ \frac{\text{car accidents}}{\text{MWe·yr}}\right)\left(900\ \frac{\text{MWe·yr waste}}{\text{car}}\right)(10^{-2})\left(1.9 \times 10^{3}\ \frac{\text{Ci}}{\text{MWe·yr}}\right)$$

$$= 2.4 \times 10^{-6}\ \frac{\text{Ci}}{\text{MWe·yr}} .$$

Similarly, other release quantities can be computed on the basis of the nuclide of interest, accident mode, age of waste, solid characteristics, and method of migration into the biosphere.

735 022

5.5 HANDLING, STORAGE, AND EMPLACEMENT AT THE FINAL GEOLOGIC ISOLATION SITE

This section describes the calculations establishing the effect of accidental releases of high-level nuclear wastes between the time the waste material arrives at the final geologic isolation site and the time the repository is sealed. It is shown that the expected release from this portion of the waste-disposal operation is relatively unimportant compared to that from interim storage and transportation.

5.5.1 Assumptions and Sources of Data

The following assumptions were used in analyzing this portion of the waste-disposal sequence:

- The head of the repository shaft is contained within a sealed building. Under normal conditions, any contamination escaping into the air in this building is prevented by a filtration system from entering the atmosphere. The filtration-system parameters (probability of failure, efficiency) are identical to those used in analyzing the interim-storage portion of the waste-disposal operation (see Sec. 5.2).
- Transportation vehicles enter the sealed storage area for unloading. The canisters are not removed from their transportation casks until they are inside the building and the air seal is reestablished.
- The only time the canisters are "bare" is during transfer between casks and while they are being lowered into the repository.
- At this point in the disposal sequence, bare canisters can dissipate all heat generated into air. Therefore, the danger to canister integrity during handling is from impact when accidentally dropped.

735 023

- The nominal time the canister is outside the casks is 10 minutes per canister, which includes movement between transportation cask and transfer cask and between transfer cask and final location.
- The probability of a drop due to crane failure is the same as in the analysis of handling at interim storage: 3×10^{-6} per hour of operation.
- The principal danger to the canisters while waiting to be lowered into the repository is impact from an aircraft. The casks will protect against all other dangers, such as earthquake, tornado, etc. Meteors are an extraordinary occurrence of insignificant probability. The same probabilities of aircraft-crash density used in Sec. 5.3 apply.
- The transportation cask remains on the surface at the final site for a nominal period of one week before it is unloaded. This is the period of exposure to an aircraft impact.
- The water path for released wastes was not analyzed since precautions can be taken to avoid a direct path into surface-water bodies serving large populations. The opportunity for cleanup exists.

5.5.2 Event Tree

Figure 5-6 shows the event tree analyzed to determine the consequences of accidental release during handling at the final geologic isolation site. Two accident modes are present: crane drop due to failure and impact by an aircraft.

5.5.3 Calculation of Release per Megawatt Electric•Year

The expected release per megawatt electric•year for each path in the event tree shown in Fig. 5-6 can be calculated when it is assumed that the

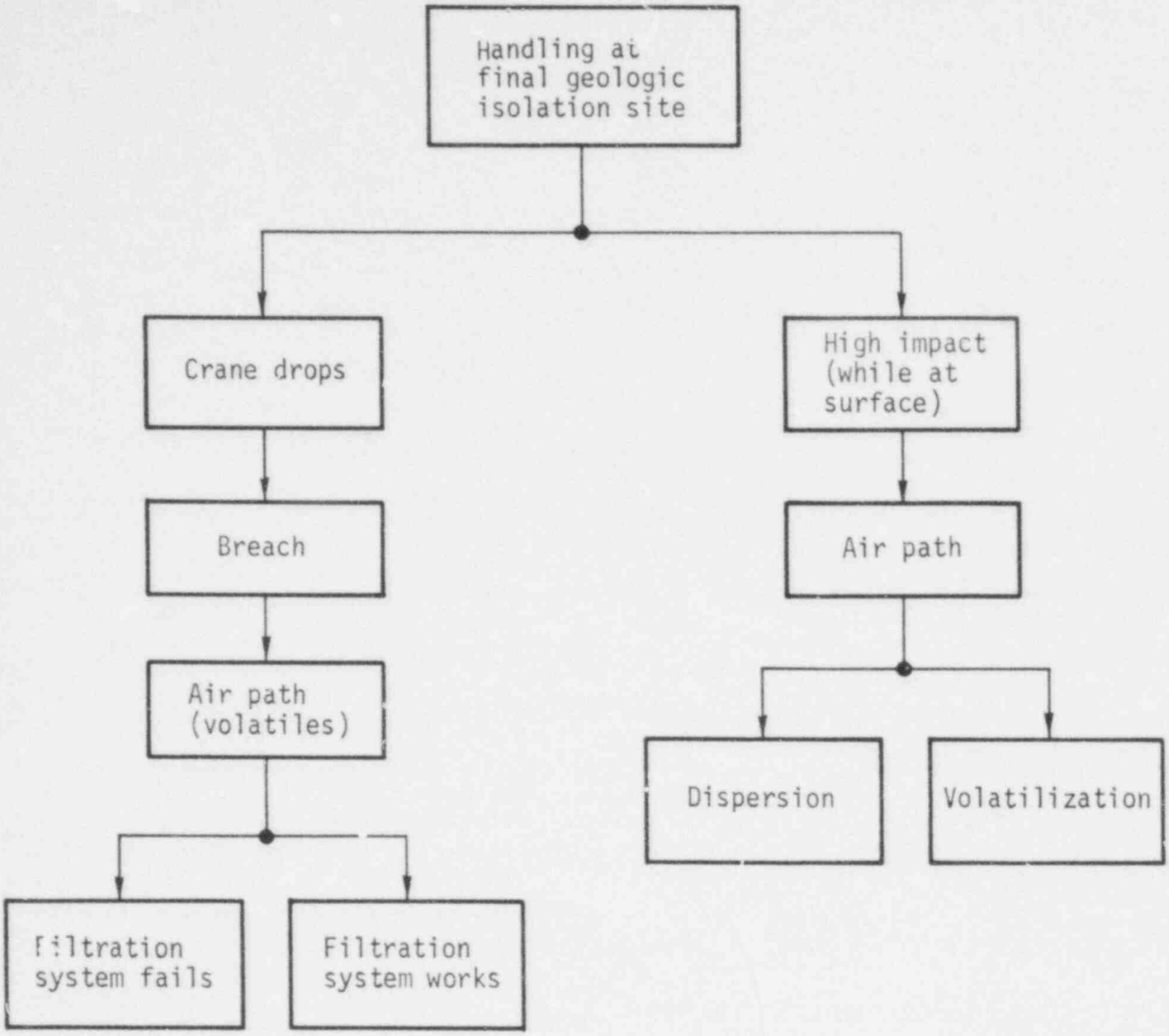

Fig. 5-6. Event tree for handling at the final geologic isolation site.

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release fractions are unity and that the canister will always breach when dropped or impacted; the expected release is insignificant compared to other parts of the waste-management process.

Crane Drop — Using the crane-drop probability (3×10^{-6}/hr) and the bare-canister handling time (10 min), the probability of a canister drop per megawatt electric•year is

$$\left(3 \times 10^{-6} \ \frac{\text{accidents}}{\text{hr}}\right)\left(\frac{1 \text{ hr}}{6 \text{ can}}\right)\left(\frac{1 \text{ can}}{100 \text{ MWe}\cdot\text{yr}}\right) = \frac{5 \times 10^{-9} \text{ accidents}}{\text{MWe}\cdot\text{yr}} .$$

This is the same accident probability as that computed for crane stall in Sec. 5.2. The effect of filter failure or correct operation also is the same, so, the results from that section apply. The expected release due to a drop caused by crane failure accident is 10^{-18} liter per megawatt electric•year times the activity contents (100 MWe•yr) of one canister, or 10^{-16} megawatt electric•year waste per megawatt electric•year.

Impact By an Aircraft — The expected release from an aircraft accident is computed using the accident density (1.27×10^{-10} crashes/yr•mi^2) given in Sec. 5.3 and a train-transportation cask cross section to aircraft crashes of 100 square yards per cask = 3.22×10^{-5} square miles per cask:

$$\left(1.27 \times 10^{-10} \ \frac{\text{crashes}}{\text{yr}\cdot\text{mi}^2}\right)\left(3.22 \times 10^{-5} \ \frac{\text{mi}^2}{\text{cask}}\right) = 4.09 \times 10^{-5} \ \frac{\text{crashes}}{\text{cask}\cdot\text{yr}} .$$

Since the cask is on site for one week, the expected release for aircraft crash on a train cask is (for release fractions of unity)

$$\frac{4.09 \times 10^{-15}}{52} = 7.09 \times 10^{-17} \ \frac{\text{MWe}\cdot\text{yr waste}}{\text{MWe}\cdot\text{yr}} .$$

(The calculation for truck transportation reduces this value since less waste is placed in trucks. The possibility of hitting several casks exists, but this could only raise the expected release by one order of magnitude, at most.)

5.6 RELEASE AFTER SEALING THE FINAL GEOLOGIC ISOLATION SITE

Analyzing the release of radionuclides from final geologic isolation sites requires a modeling approach considerably different from those discussed thus far because of the long time-spans of interest. The major pathways considered involve water reaching the burial cavity, leaching out the material, and slowly carrying it to surface water. Seismic events are important in initiating the conditions under which such processes would occur. Other pathways involve loss of administrative control and "extraordinary occurrences."

Construction of an event tree for this analysis relied on the discussion in chapter 3 of Schneider (1974). The tree was used as the basis for making a "probability flow diagram," which in turn was used to calculate the relevant "state probabilities." The computer program used to generate numerical results can readily be expanded as the event tree grows in complexity.

5.6.1 Assumptions and Sources of Data

The assumed final isolation geometry is shown in Fig. 5-7. The burial vault or repository is in a thick layer of material (shale, granite, etc.), which is dry at the time of emplacement. A vertical shaft from the surface to the repository passes through:

- an *aquifer* layer, containing slowly moving groundwater,
- a *barrier* layer, assumed to be impermeable to water.

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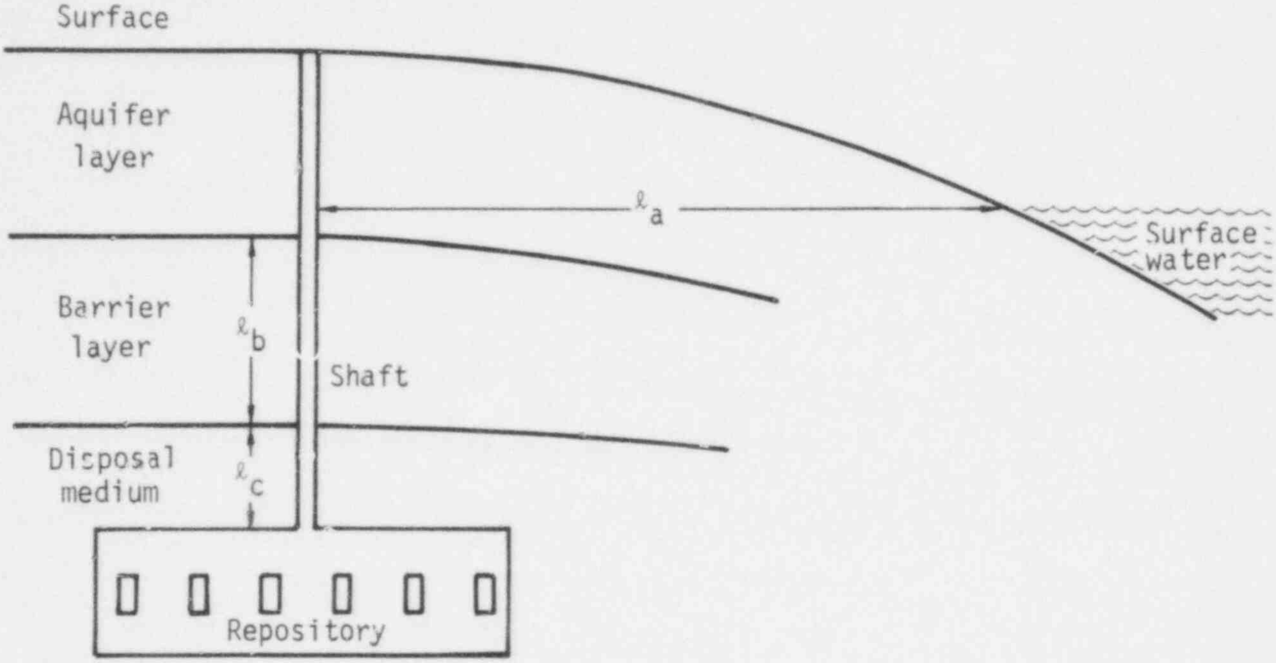

Fig. 5-7. Final geologic isolation geometry.

At some time after the waste canisters have been placed in the repository, the shaft is sealed so that water cannot enter. Reentry by humans would also be difficult.

In chapter 3 of Schneider (1974), a "fault-tree" approach was used to define possible sequences of events leading to release from geologic isolation. The accompanying discussion of the events and identified scenarios, including judgments expressed about the relative importance of these scenarios, guided the current investigation in two ways: first, in developing the event tree described in Sec. 5.6.2, and second, in initially choosing of some of the controlling probabilities used in generating numerical results.

It is assumed that the region selected is *not* one of high seismic activity (not near a tectonic plate boundary). However, seismic activity is important in a number of the pathways considered.

Seismic events and their effects have been classified as small, medium, and large. In actuality, a continuum of possibilities exists, but lumping is necessary to achieve a tractable, finite-state model. Table 5-3 summarizes probabilities and "transition rates" corresponding to these three types of seismic events and other events discussed in Sec. 5.6.2. *Transition rate*, λ_i, is defined in terms of the conditional probability of a transition to a particular state (due to an event "i") during an interval Δt, given that an appropriate predecessor state exists at the start of the interval. Namely,

$$\lambda_i = \lim_{\Delta t \to 0} \frac{\text{Prob[event i during } \Delta t\text{, appropriate predecessor]}}{\Delta t}$$

Transition rates have units of inverse time. Use of these parameters is discussed further in Sec. 5.6.3. One column in Table 5-3 lists expected values. The right two columns provide low and high or "endpoint" values, which are used in sensitivity calculations designed to illustrate the effect of changed assumptions.

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Table 5-3. Assumed probabilities and transition rates for final geologic isolation tree.

Probability (p) or transition rate (λ)	Event	Values			
		Nominal value	Units	End points	
				Low	High
p_A	Original flaw in seal or barrier	1×10^{-4}	-	1×10^{-2}	1×10^{-6}
λ_B	Small seismic activity	1×10^{-3}	yr^{-1}	1×10^{-2}	1×10^{-4}
λ_C	Medium seismic activity	1×10^{-4}	yr^{-1}	1×10^{-3}	1×10^{-5}
λ_D	Large seismic activity	1×10^{-5}	yr^{-1}	1×10^{-4}	1×10^{-6}
λ_E	Change in surface water proximity	1×10^{-6}	yr^{-1}	1×10^{-5}	1×10^{-7}
λ_F	Loss of administrative control	1×10^{-4}	yr^{-1}	1×10^{-3}	4×10^{-5}
λ_G	Open pathway due to drilling	4×10^{-4}	yr^{-1}	4×10^{-3}	4×10^{-5}
λ_H	Extraordinary occurrences	1×10^{-9}	yr^{-1}	1×10^{-7}	1×10^{-11}

Specific information pertaining to the events, transition rates, and data sources is summarized below:

- *Seismic Activity.* This type of activity is grouped into three categories or levels:

  Small: VI on the Modified Mercalli scale (Richter, 1958),
  Medium: VII to VIII on the Modified Mercalli scale,
  Large: IX or above on the Modified Mercalli scale.

  The different levels result in different effective column lengths along which nuclides migrate to surface water, and different groundwater velocities.
- *Transition Rates.* The estimated transition rates for the three seismic levels (λ_B, λ_C, λ_D) are based on data from Table 19-1 of Press and Sieves (1974) (data are plotted in Fig. 5-8) and the discussion in Schneider (1974). The latter cites Bollinger (1972) and Algermissen (1969) as original sources. A good discussion of the relationship between the Modified Mercalli scale (intensity at point of interest) and the Richter scale (magnitude at epicenter) is given by Richter (1958).
- *Original Flaw in Seal or Barrier.* The probability estimate follows the discussion on pages 3.23 and 3.27 of Schneider (1974); original sources cited are Bureau of Mines Circular No. 57, Katz and Coats (1968), and *Statistical Abstract of the United States* (1971).
- *Change in Surface-Water Proximity.* The transition-rate estimate follows the discussion on page 3.27 of Schneider (1974); original source cited is *The National Atlas of the United States* (1970).
- *Loss of Administrative Control.* The transition-rate estimate follows the discussion on the probability of war (Schneider, 1974, page 3.36) adjusted for likelihood of reinstatement of control.

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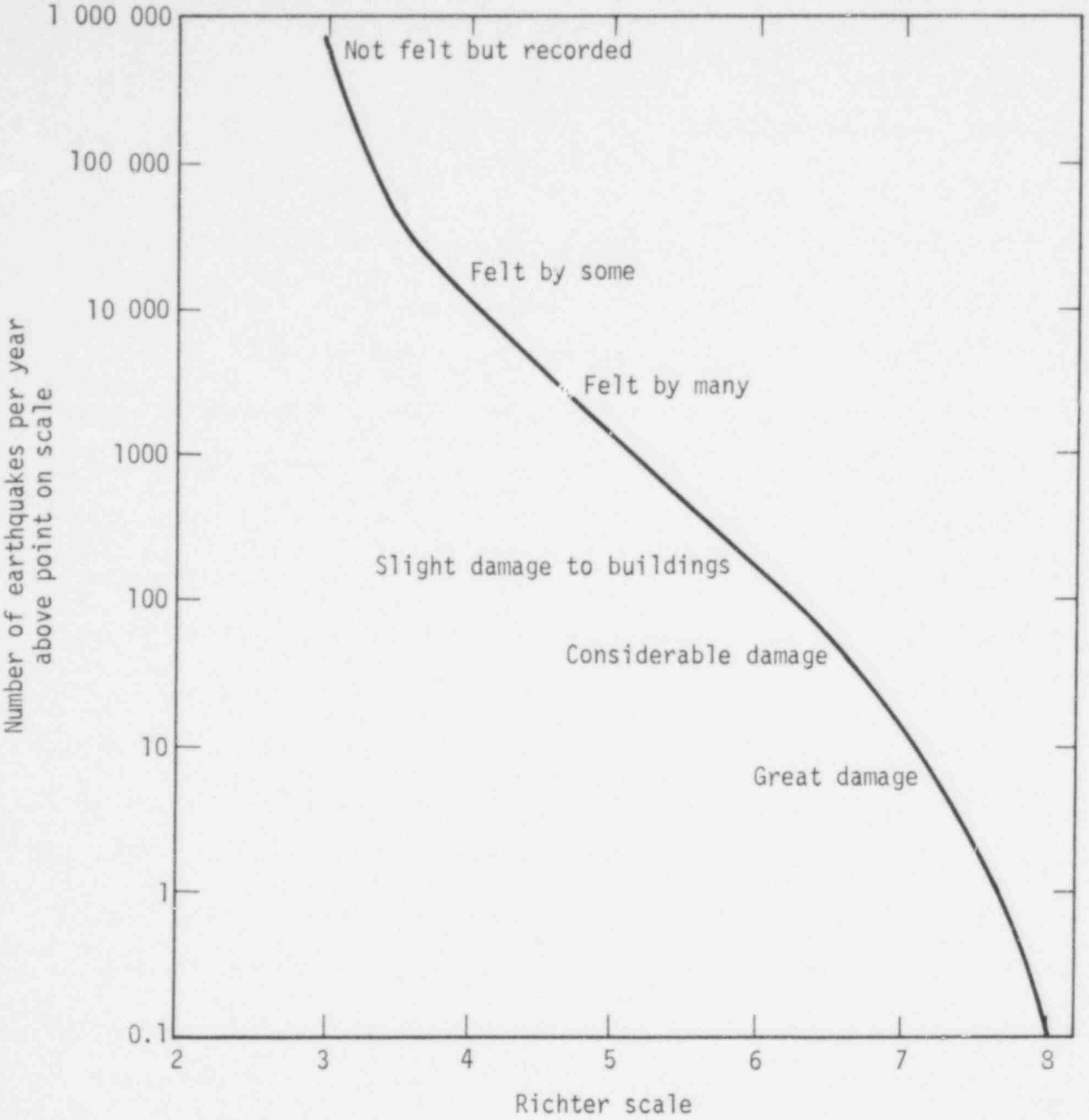

Fig. 5-8. Earthquake frequency vs magnitude. Plotted from Table 19-1 in F. Press and R. Siever, *Earth* (Freeman, San Francisco, Calif., 1974), p. 639.

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- *Drilling Opens Pathway.* The transition-rate estimate follows the discussion on pages 3.33-3.94 of Schneider (1974); original source cited is "Forecast/Review," *Oil and Gas Journal* (1973).
- *Extraordinary Occurrences:* Page 3.31 of Schneider (1974) gives the probability of a volcano as 10^{-4} times the probability of large seismic activity; original source cited is *The National Atlas of the United States*. Page 3.31 of Schneider (1974) gives the probability of a large meteor removing the barrier as 10^{-14} per site•year. Blake (1968) gives related information.

The estimate of frequency of occurrence (transition rate) of large seismic activity was taken from a discussion in Schreider (1974) related to a "generic Eastern site." This estimate could be modified as follows. First, Algermissen (1969) divides the U.S. into four broad zones. The "strain release index" for the Central Plains zone is approximately one-fifth that of the Eastern zone; the index for the Rocky Mountain zone is approximately twice that of the Eastern zone. The Pacific West index is considerably higher than the other three. Second, careful selection of a repository site within any of the quieter three zones and away from any known fault lines would justify a reduction in the assumed transition rates.

The following events and processes are *not* included in the initial analysis reported here: adversary action, glacial action, erosion of barrier, failures caused by monitoring or testing, migration of cavity, cavity collapse during emplacement, and volcanic or meteor effects (except minimal considerations under "extraordinary occurrences.") All of the above are discussed in chapter 3 of Schneider (1974) and are judged to be far less important than the factors included in the event tree discussed in the next section.

5.6.2 Event Tree

The event tree representing pathways from final geologic isolation is shown in Fig. 5-9. The block at the top of the diagram represents the initial condition or "state of the system," corresponding to zero failures or events of consequence. Various event chains or pathways lead to the bottom of the diagram, corresponding to conditions under which nuclides have entered ground water and are migrating toward the biosphere. The diagram is constructed so that, in general, reading from left to right, the pathways become less likely but have more severe consequences.

Some of the steps (lines connecting blocks) in Fig. 5-9 are labeled with probabilities or transition rates (P_A, λ_B, λ_C, etc.). These are the parameters that are tabulated in Sec. 5.6.1 and that control the calculations presented in Sec. 5.6.3. All of the other events shown are assumed to occur with a probability of 1, as a direct consequence of the controlling events. For example, given the fact that an original flaw exists in the seal or barrier, the subsequent events in the left-most path (failure to detect or correct flaw, water flow into cavity begins, canister disintegrates, and leaching and migration begins) are assumed to occur immediately.

5.6.3 Methodology and Sample Calculations

The event tree for final geologic isolation pathways (Fig. 5-9) does not lend itself directly to the generation of appropriate equations for computing probabilities. For example, an event C, initiating release of nuclides into ground water under one set of conditions, can be followed by an event D, worsening the conditions and speeding up the transport of nuclides to the biosphere. Thus, a more complex diagram is needed — one which takes such transitions into account. The new diagram can also be simplified in certain

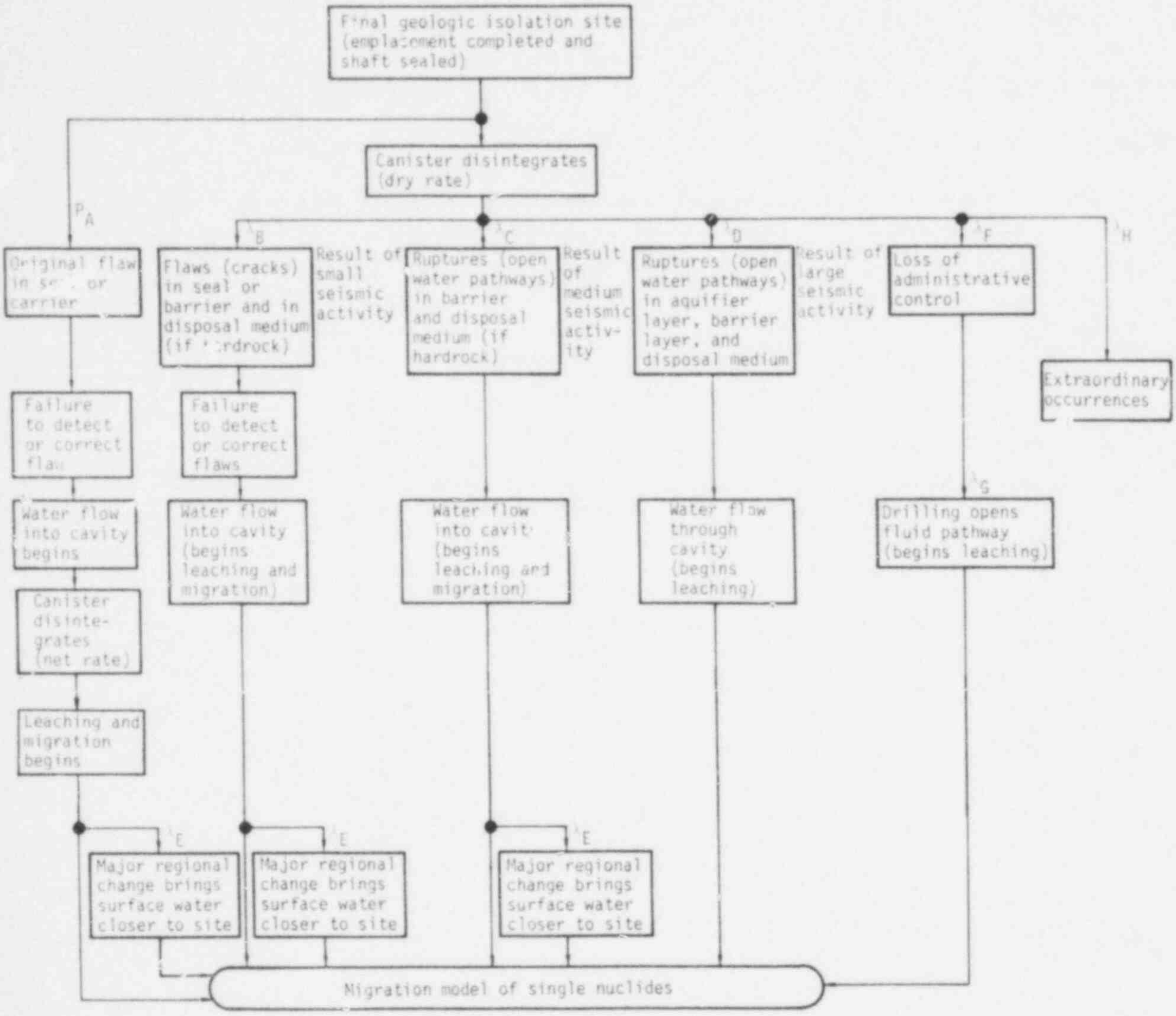

Fig. 5-9. Event tree: pathways from the final geologic isolation site.

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respects. For example, if two pathways lead to equivalent release conditions, they can be joined in a composite or merged node. Note also, that *simplifications based on small-number assumptions (U.S. NRC,* 1975*) cannot be used here because of the very long times involved.* That is, after sufficient time, some of the node probabilities will become large. Therefore, computer solutions are necessary that take proper account of all couplings through transitions between nodes.

Figure 5-10 is a probability flow diagram illustrating the transitions between nodes discussed above. Nodes can be categorized as follows:

- *Single-Event Nodes*, such as:

    C - Event C has occurred[*],

    E - Event E has occurred,

    Z - (Special case) Zero events have occurred.

- *Joint-Event Nodes*, such as:

    CE - Events C and E have occurred,

    EF - Events E and F have occurred.

- *Merged Nodes*, such as:

    A/B - Either A or B or both have occurred.

    D/G - Either D or G or both have occurred.

- *Compound Nodes*, such as:

    (A/B)E - Either A or B or both, and E have occurred,

    (A/B)EF - Either A or B or both, and E and F have occurred.

A set of simultaneous ordinary linear differential equations, describing how probability flows through the model, can be written by inspecting the diagram

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[*] A more precise definition is: Event C has occurred, but no other events leading to worse (or potentially worse) release conditions have occurred. Thus, A or B may have occurred, but D, E, F, G, or H have not.

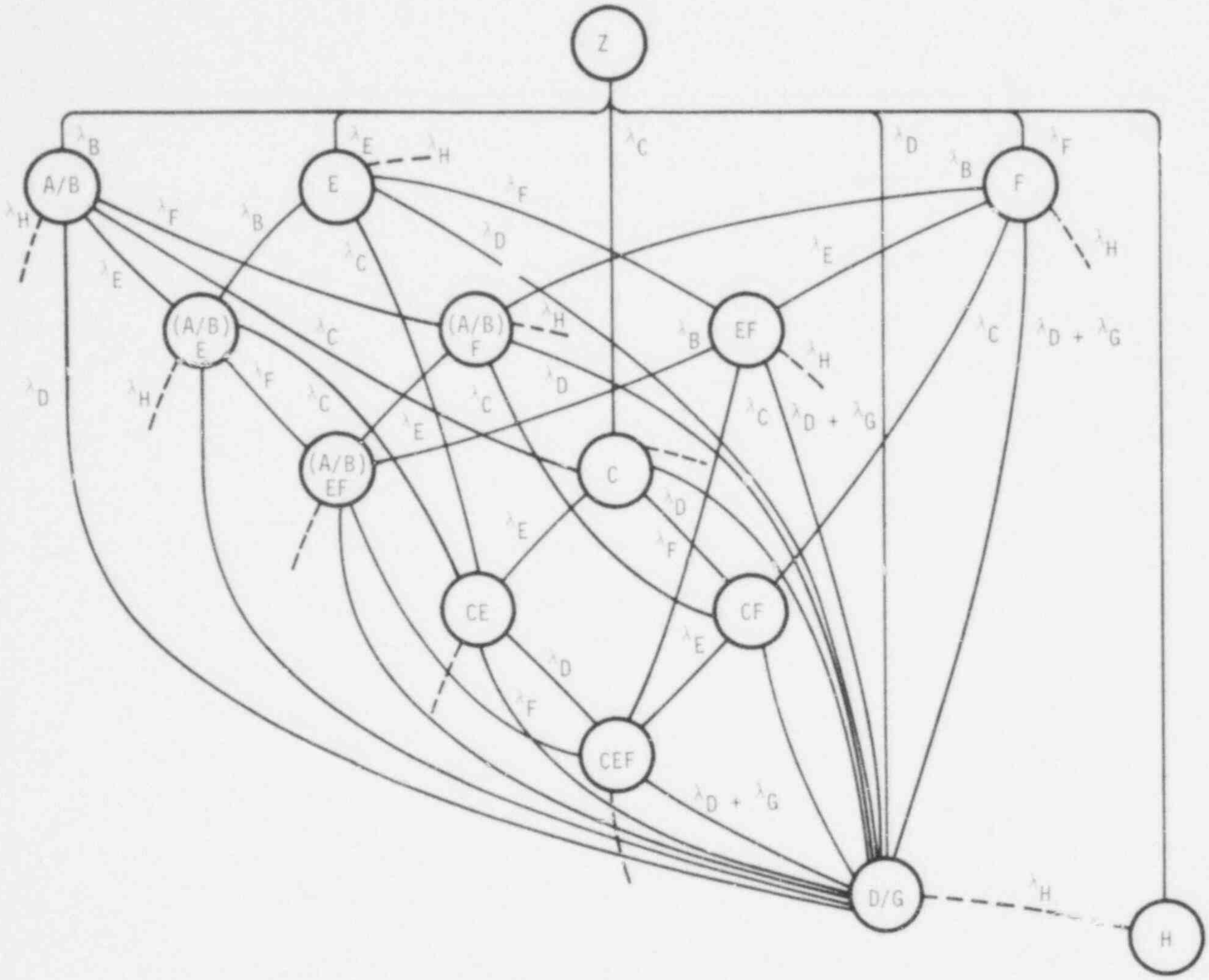

Fig. 5-10. Probability flow diagram corresponding to final geologic isolation event tree.

in Fig. 5-10. These equations are readily solved by using an appropriate computer program.

The 14 nodes or states of the diagram in Fig. 5-10 can be grouped according to subsets having equivalent release conditions. These subsets are:

I	Z	Zero-release conditions
	E	
	F	
	EF	
II	A/B	Increasing release rate (i.e., decreasing time of transport into surface water)
	(A/B)F	
III	C	
	CF	
IV	(A/B)E	
	(A/B)EF	
V	CE	
	CEF	
VI	D/G	
VII	H	Impulsive release

The state variables to be calculated are the elements of a 14-state probability vector, $p(t)$. Each element, $p_i(t)$, represents the probability of being in a particular state i at time t. The vector state-dynamic system equation to be solved is

$$\dot{p}(t) = M\, p(t),$$

where M is a 14 × 14-dimensional matrix of transition rates. For example, defining state 1 as the Z-state (zero events) and state 2 as the A/B-state, then

$$M_{1,1} = -\lambda_B - \lambda_C - \lambda_D - \lambda_E - \lambda_F - \lambda_H ,$$
$$M_{2,1} = +\lambda_B ,$$
$$M_{2,2} = -\lambda_C - \lambda_D - \lambda_E - \lambda_F - \lambda_H .$$

The computer solutions are found using the transition matrix $\phi(t)$ defined in terms of the matrix exponential function

$$\phi(t) = e^{Mt} .$$

The solution at any time t is determined by the inital condition, $p(0)$, and is given by

$$p(t) = \phi(t)\, p(0).$$

In this case, the initial condition is

$$p_1(0) = 1 - p_A ,$$
$$p_2(0) = p_A \text{ (probability of original flaw in seal or barrier)},$$
$$p_3(0) \rightarrow p_{14}(0) = 0.$$

Baseline Results — The 14-state system of equations outlined above was solved out to a million years (the baseline-case parameters shown in Table 5-3 were used). The 14 probabilities were grouped as listed above, and the group sums plotted versus time in Fig. 5-11. *It is important to note that the results plotted are very dependent on the assumptions used in these sample calculations.* Groups V, VI, and VII are not shown because their probabilities

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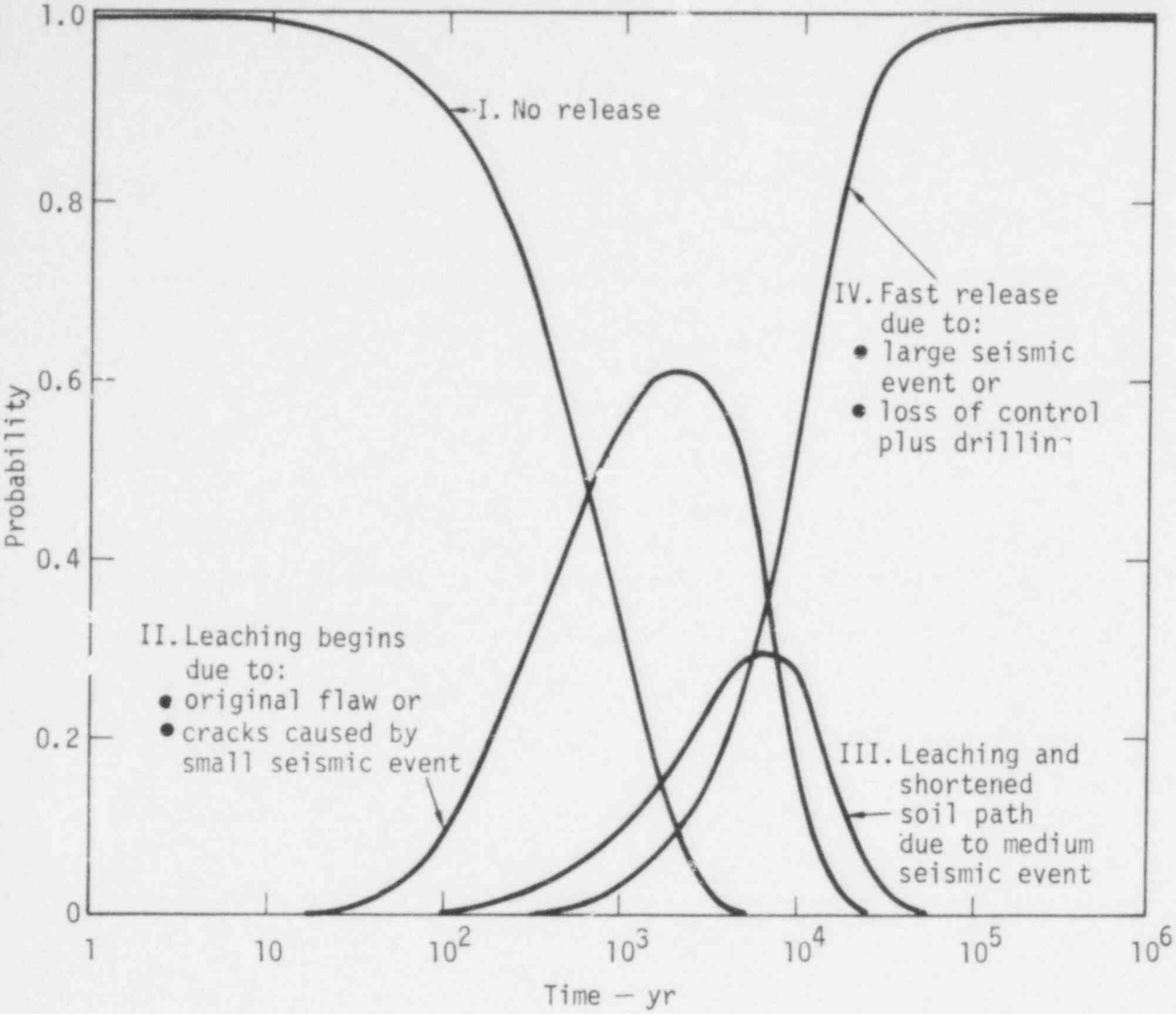

Fig. 5-11. Group probabilities vs time: baseline case.

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are negligible. (As time is extended toward a billion years, Group VII will ultimately show up.) Those plotted correspond to the following release scenarios:

Group I	No release.
Group II	Leaching begins due either to an original flaw in the seal or barrier or to cracks caused by a small seismic event. (The latter dominate.)
Group III	Leaching begins (or continues, if a Group II event predated the Group III event) and the effective soil column is shortened because of a medium-sized seismic event.
Group IV	Leaching begins (or continues, if a Group II or III event predated the Group VI event) directly into the surface water because of a large seismic event or due to the combination of loss of administrative control plus drilling by man. (The latter is more important in this case.)

Figure 5-11 shows the progression of probabilities of the above groups as a function of time after emplacement. Notice that the probability of Group I (no release) decreases monotonically with time, as expected — the longer the material has been in the ground, the higher the probability of some release. As time progresses in Fig. 5-11, situations implying intermediate release rates (Groups II and III) increase and then decrease in probability as each is supplanted by the rising probability of a more serious set of circumstances. After a certain time, the probability that Group IV (faster release) has been experienced, dominates.

Examination of the baseline case curves in Fig. 5-11 leads to the following observations:

- Release of nuclides is dominated by events leading to the states in Groups II and IV.

- After 2000 years, the probability is 0.6 that the state of the system lies in Group II and that leaching will have begun.
- After 30 000 years, the probability is near unity that the state of the system lies in Group IV. That is, either a large seismic event or the combination of loss of control plus drilling will have opened up a direct pathway to surface water.

Thus, a typical case might be: entry into Group II at 500 years, followed by entry into Group IV at 10 000 years. At the latter time, the amounts of nuclides remaining in the soil column, if any, depend on leach rate, groundwater velocity, column length, etc. Meaningful statistical calculations based on a model for leaching and migration require a joint probability-density function for the two variables:

τ_{II} - time of entry into Group II,

τ_{IV} - time of entry into Group IV.

Those cases in which the Group II event follows the Group IV event have no importance. To provide a relatively simple-to-work-with initial set of numerical data, a discrete joint density function was generated (approximate graphical techniques based on the computer data from this baseline case were used). The function is tabulated in Table 5-4; the 12 entries in the table serve as weighting functions for 12 migration model scenarios.

Table 5-4. Discrete joint density function for initiating event times: baseline case.

τ_{II} \ τ_{IV}	5000 yr	10 000 yr	20 000 yr	No event
200 yr	0.23	0.13	0.06	0.05
600 yr	0.16	0.09	0.04	0.03
1000 yr	0.10	0.06	0.03	0.02

Example Variations — A set of ten additional computer calculations were generated to show the effects of variations in the input parameters from their baseline values. The resulting probabilities for Groups I, II, III, and IV were plotted for three cases: Case 4 — high seismic activity, Case 5 — low seismic activity, and Case 9 — better administrative control (see Figs. 5-12, 5-13, and 5-14, respectively).

Comparison of Case 4 (Fig. 5-12) with the baseline case shows a similar pattern of group probability histories, but with events occurring (statistically) earlier in time. Also, Group III plays a more significant role — it is not so closely overriden by Group IV because Group IV events are no longer dominated by the F-G path (loss of control plus drilling) as in the baseline case. Rather, it is dominated by the D path (large seismic events), which is less likely than the C path (medium seismic events) of Group III.

Case 5 (Fig. 5-13) releases are dominated by Group IV alone. Here, the seismic activity paths (B, C, and D) have been so reduced in likelihood that the nonseismic part of Group IV — the F-G path (loss of control plus drilling) — is practically all that matters.

Case 9 (Fig. 5-14) shows a similar pattern to that of Case 4 (Fig. 5-12) but with everything happening later in time. As in Case 4, the three types of seismic events dominate, but here it is done by making the F-G path less probable rather than by making the B, C, and D paths more probable. The Groups I and II probability histories are very similar to those of the baseline case.

As stated earlier, the results illustrated by these sample calculations are very dependent on the assumed probabilities and event-tree structure. The methodology illustrated can be readily extended to a more complex event tree (or trees) corresponding to a more elaborate description of final geologic isolation pathways.

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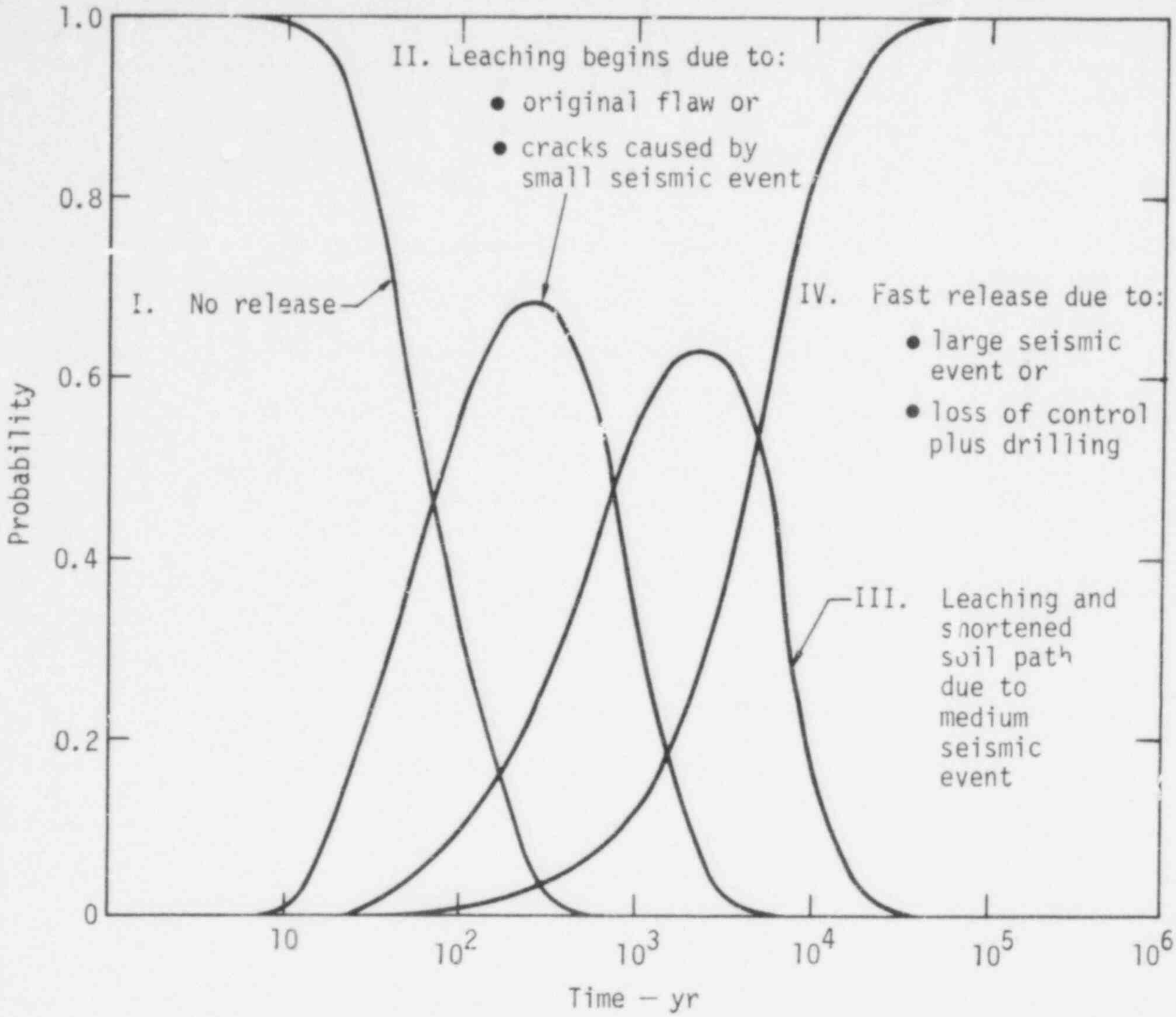

Fig. 5-12. Group probabilities vs time: high seismic activity.

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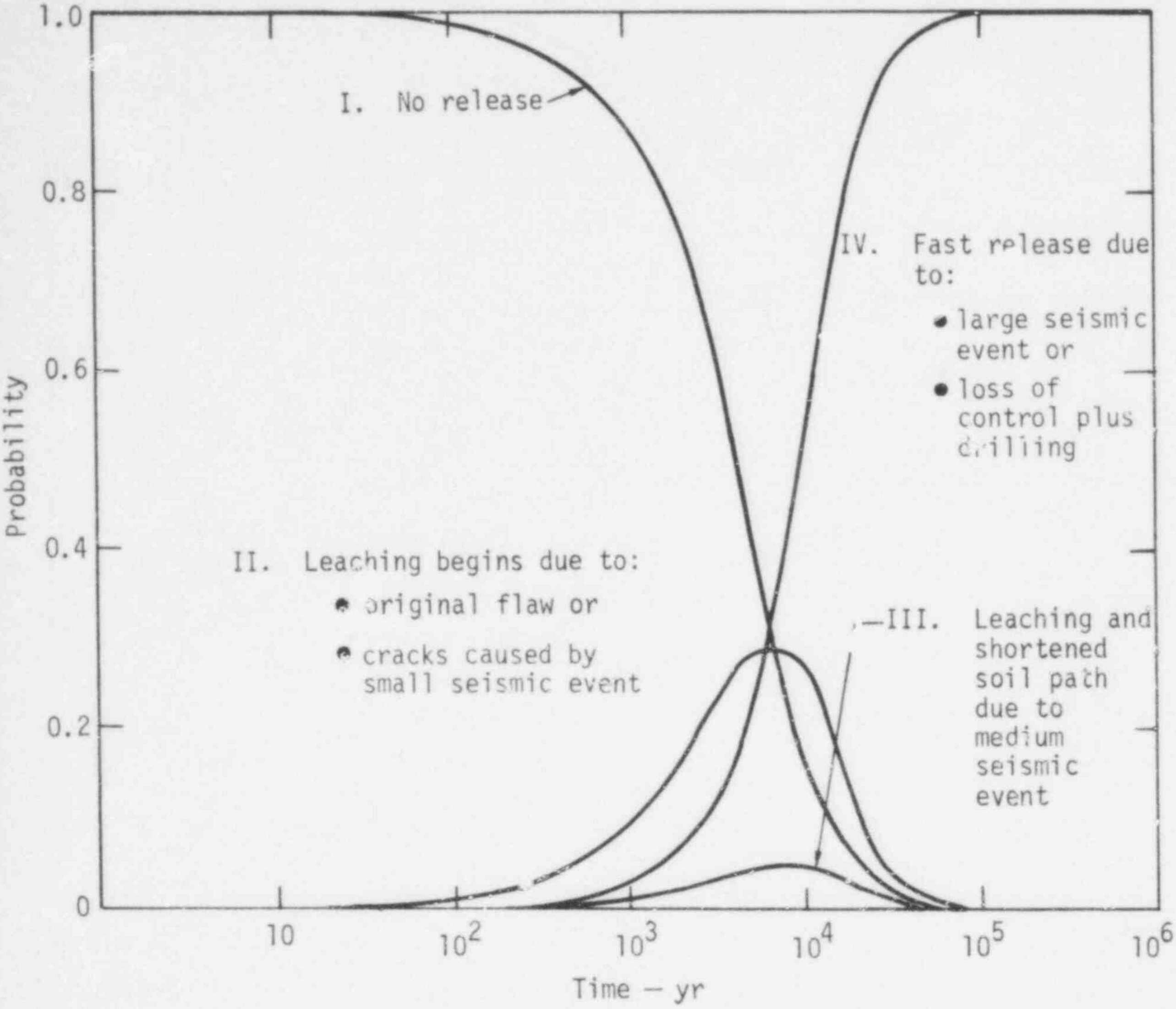

Fig. 5-13. Group probabilities vs time: low seismic activity.

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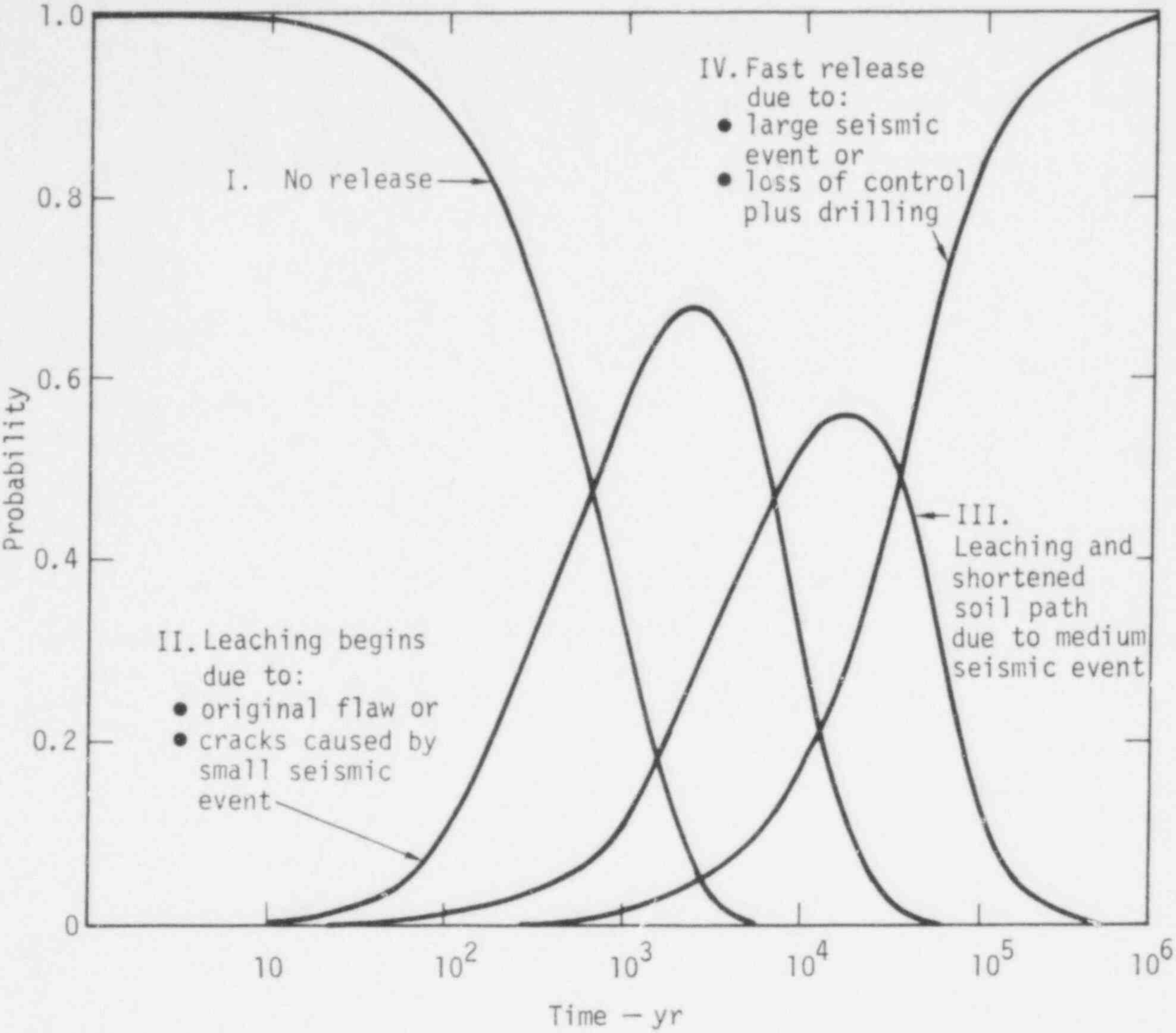

Fig. 5-14. Group probabilities vs time: better administrative control.

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6 RESULTS

6.1 SENSITIVITY OF POPULATION DOSE TO CHARACTERISTICS OF HIGH-LEVEL SOLIDIFIED WASTE

The mathematical models of release mechanisms and of pathways for propagation into man, which are described in Sec. 5 and Appendices B and C, were exercised to determine the sensitivity of population dose to key characteristics of the solid-waste form. Specifically, solubility, dispersibility, and volatility were considered. The effect of release is computed as the expected value of the 50-year dose to the general population per megawatt electric·year of generation. The dose to maximum individuals (those members of the general population whose diet and living habits maximize their exposure) is also determined. Where air pathways are concerned, the appropriate assumed population distribution (from Appendix D) is incorporated into the calculation. Where water pathways are concerned, the value computed is the expected value of the 50-year individual dose per megawatt electric·year to person living near the affected body of water; to determine population dose, a specific population must be assumed. The dose to maximum individuals is based upon critical path analysis.

The analysis divides naturally into two major categories — release prior to sealing the waste repository ("pre-emplacement") and release after sealing the repository ("post-emplacement"). The former involves a large number of possible release modes and pathways, and waste that has lost little of its radioactivity since removal from the reactor; the latter involves the dissolving of the solidified waste in the repository and its entry into ground water and, eventually, into surface water and man. The two categories are discussed separately in the following sections.

6.2 SENSITIVITY ANALYSIS FOR THE PRE-EMPLACEMENT PERIOD

6.2.1 Identification of Critical Accidents

The number of potential accident modes that can result in the release of radioactive nuclides from HLSW prior to emplacement is large. It will be recalled that for the storage, handling, and transportation operations considered in this study, two major accident types — drainage of the interim storage pool at the FRP and transportation to the repository — were determined to dominate (in terms of expected population dose per megawatt electric·year) all pre-emplacement accident modes. For that reason, sensitivity analyses were completed for these modes only.

6.2.2 Drainage of the Interim Storage Pool

The scenario analyzed was that described in Sec. 5.3, in which a massive earthquake ruptures the storage pool and the building enclosing it, and so disrupts services that the canisters of HLSW cannot be cooled. Under these circumstances, the waste will heat up and a certain number of the canisters will breach, releasing volatiles and particulates into the air.

Figure 6-1 shows the expected value of population dose per year of storage as a function of the fraction of material released in the accident described above. The release fraction may be related to the solid-waste form and can be used to evaluate solid-waste characteristics. The lines for 1-year-old waste assume a probability of 1 that the canisters will breach. Those representing 10-year-old waste are adjusted to account for a probability of 0.1 that the canisters will breach.

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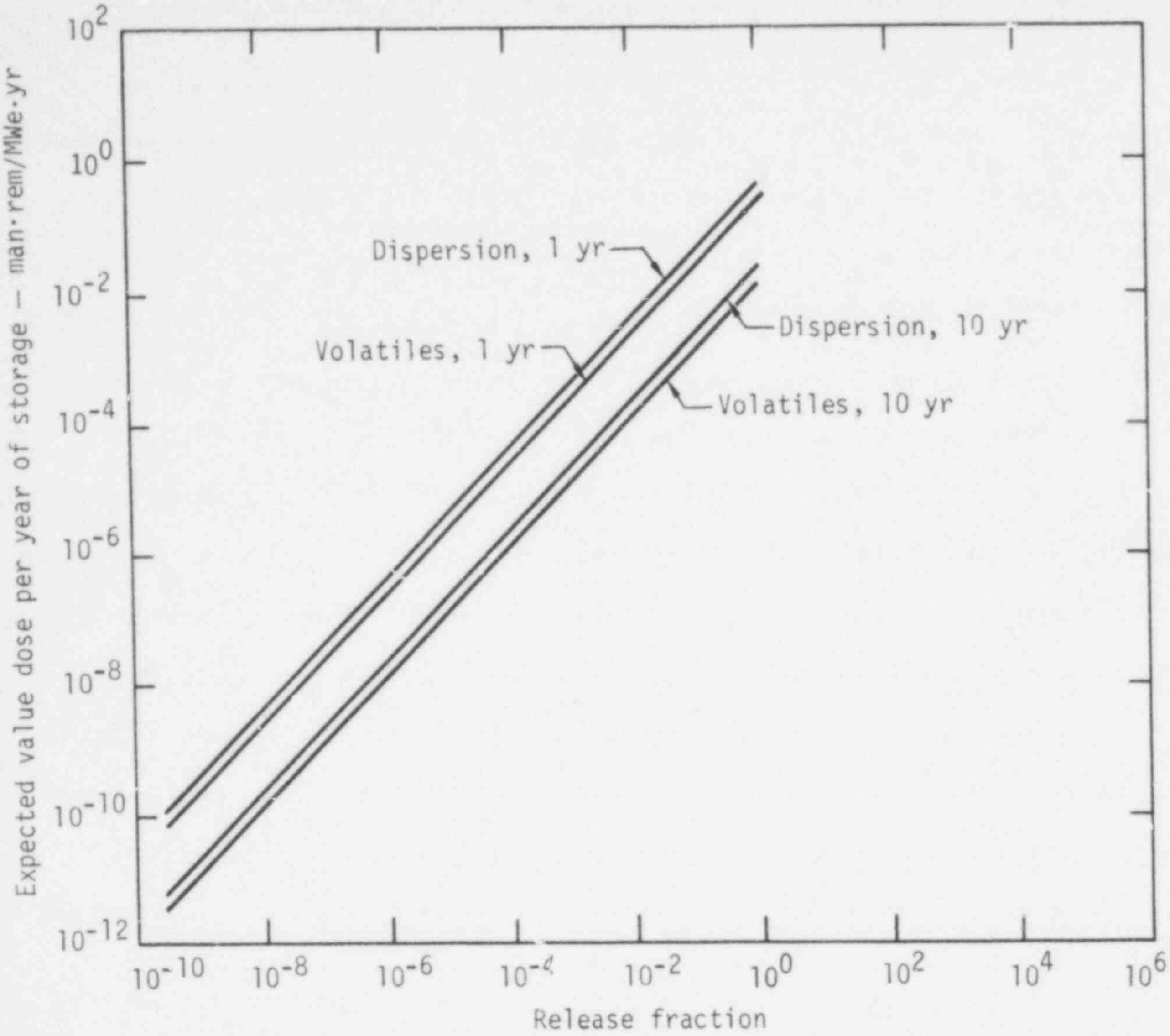

Fig. 6-1. Expected dose as a function of release fraction for drainage of the interim storage pool.

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6.2.3 Modal Transportation Accidents

In order to relate the expected population dose in transportation accidents to a single parameter — a characteristic of the HLSW — a modal accident was defined for each form of transportation accident. This was done by combining the probability density function (pdf) and the release function for each accident mode. (Both are described in Sec. 5.4.) Figure 6-2 illustrates how the modal accident is defined, using conceptual pdf and release-function curves. The product of the pdf and the release function is formed. In every case, it is zero at low accident severities (since no release occurs) and at very high accident severities (since the probability of such severities is zero). At some intermediate point, the product reaches a peak value. The modal accident is defined as one with a severity (mph for impact, °F•min for fire, and $\sec^{-1}$ for puncture) equal to that at which the peak value of the product occurs. The expected value of population dose is computed as a function of the release fraction — the fraction of available radioactivity that escapes from the protective system under conditions determined by the modal accident. In general, there are different modal accidents for each accident mode (impact, fire, and puncture), for each solid characteristic (dissolution, dispersion, and volatilization), and for each transportation mode (truck and train) studied. Table 6-1 provides a statement of the modal accident conditions (the modal fire severity [in °F•min] is broken down into separate components of time and temperature in Table 6-1).

6.2.4 Dependence of Dose on Release Fraction for Transportation Accidents

Figures 6-3 through 6-6 illustrate the dependence of the expected value of population dose on the release fraction for those transportation accidents involving air pathways to the biosphere. The plots are easily compared with

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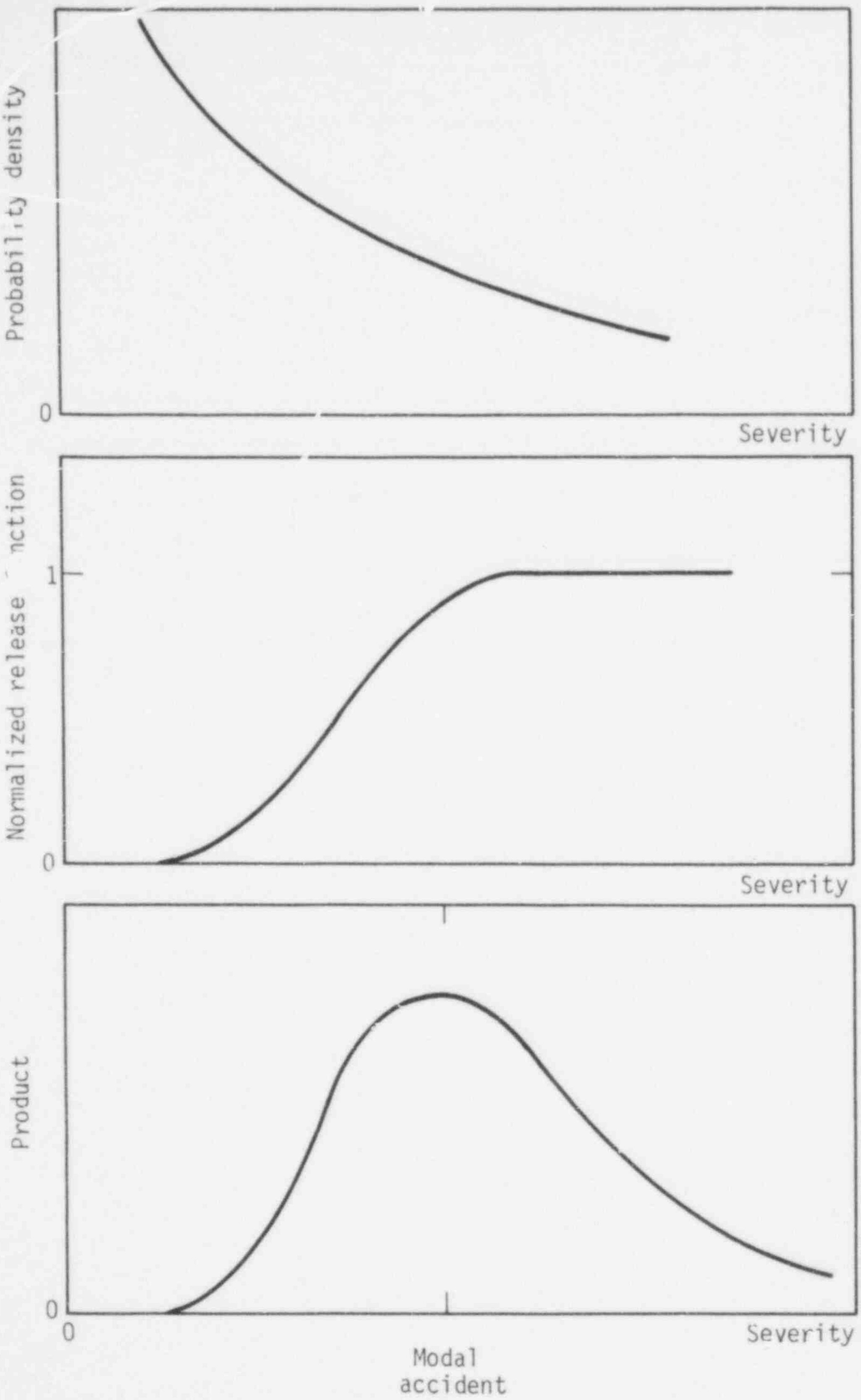

Fig. 6-2. Definition of modal accident.

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Table 6-1. Modal accident conditions.

	Truck	Train
Impact dispersibility		
Impact velocity (mph)	60	50
Puncture dispersibility		
Impact velocity (mph)	40	40
Fire dispersibility		
Temperature (°F)	2000	2000
Time (min)	80	45
Fire volatility		
Temperature (°F)	2000	2000
Time (min)	80	240
Impact dissolution		
Impact velocity (mph)	60	50
Puncture dissolution		
Impact velocity (mph)	40	40
Fire dissolution		
Temperature (°F)	2000	2000
Time (min)	80	240

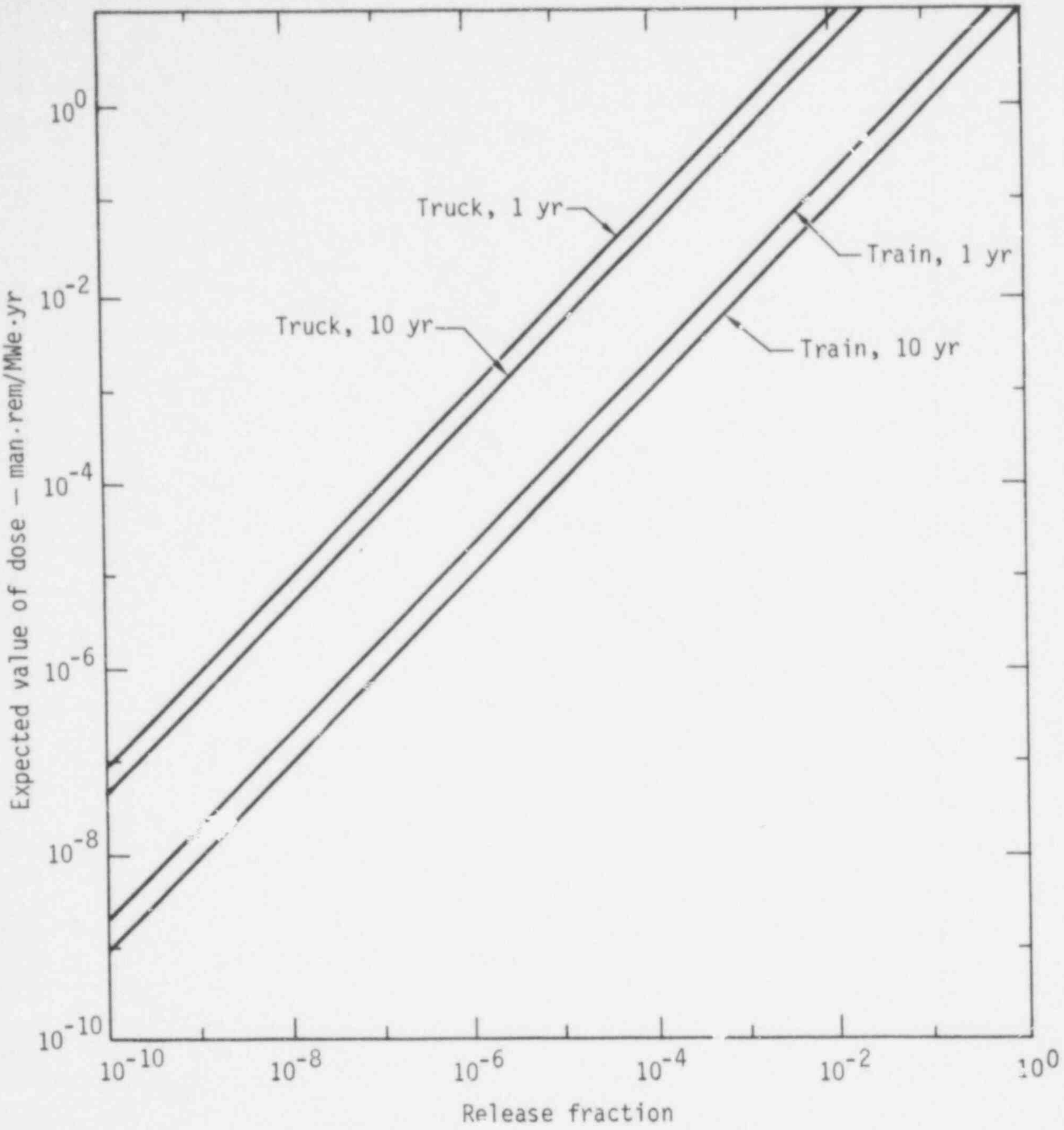

Fig. 6-3. Dependence of dose on release fraction: transportation, dispersion, and impact.

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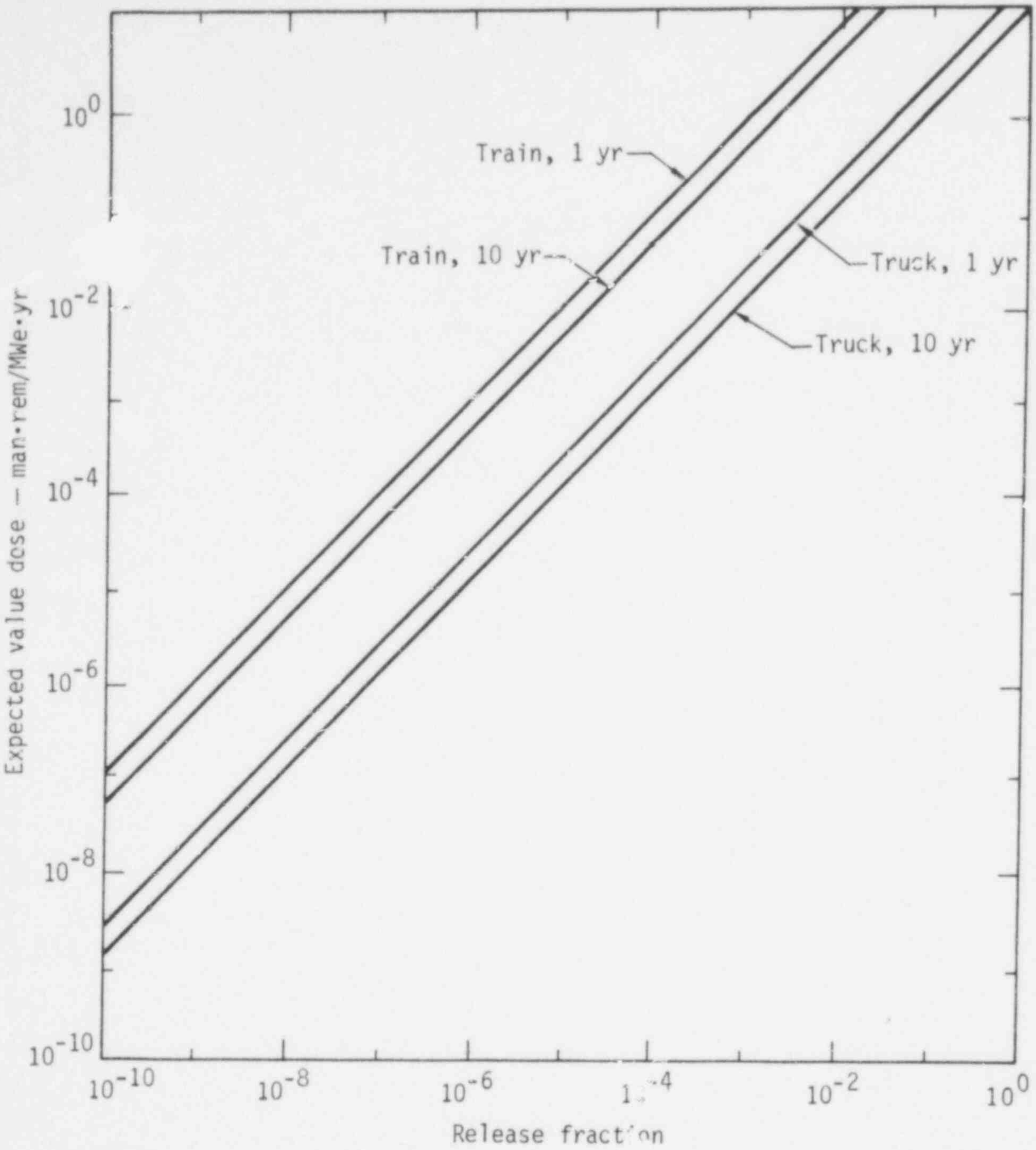

Fig. 6-4. Dependence of dose on release fraction: transportation, dispersion, and fire.

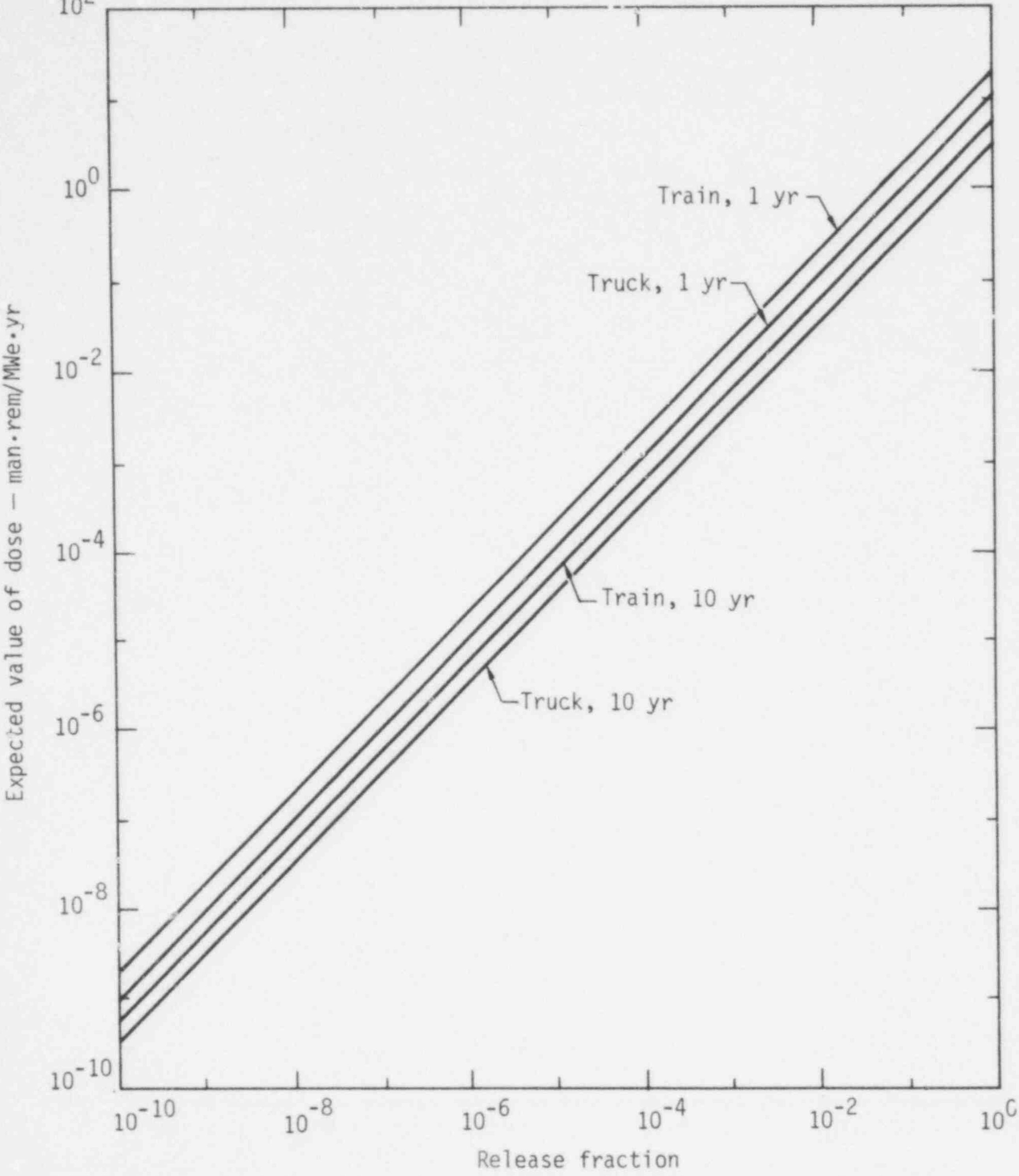

Fig. 6-5. Dependence of dose on release fraction: transportation, volatiles, and fire.

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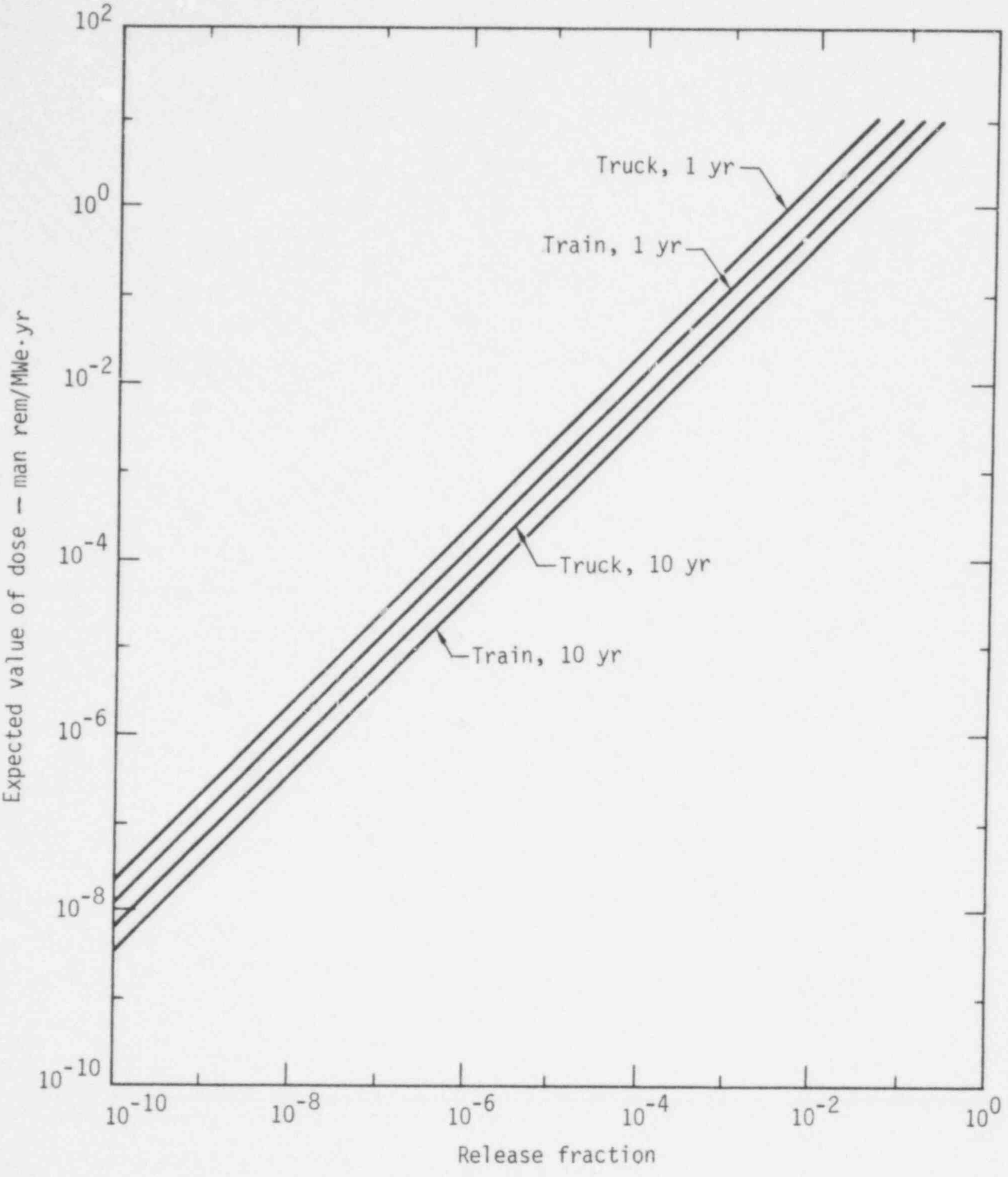

Fig. 6-6. Dependence of dose on release fraction: transportation, dispersion, and puncture.

similar curves for loss of water in the interim storage pool shown in Fig. 6-1. It can be seen that the difference between the dose from 1-year-old waste and 10-year-old waste is considerably smaller for transportation accidents because the age of the waste does not significantly influence the probability of its release.

Insights into acceptable release fractions can also be gained from Figs. 6-3 through 6-6. For example, if 10^{-2} to 1 man·rem per megawatt electric·year is the range of largest acceptable expected population dose per accident mode, maximum acceptable release fractions are on the order of 10^{-5} to 10^{-3} for release by dispersion from impact accidents involving trucks, and 10^{-3} to 10^{-1} for impact accidents involving trains. As in the case of drainage of the interim storage pool, the total expected dose is the sum of those from all the accident modes. For example, if truck transportation of 1-year-old waste is considered, the total expected dose is the sum of those from impact dispersion, fire dispersion, fire volatiles, puncture dispersion, and the water-pathway modes discussed below.

Figures 6-7 through 6-9 illustrate the sensitivity relations for transportation accidents involving water pathways to the biosphere. They differ slightly from Figs. 6-3 through 6-6 in the method of presentation. The expected dose shown is to a single individual. The reader must provide an assumed population living in the vicinity of the affected body of water. For example, if 100 000 people are presumed to live near the affected water, the lines must be raised by five orders of magnitude to represent expected population dose in man·rem per megawatt electric·year. In this case, if for example 1.0 man·rem per megawatt electric·year is the largest permissible expected dose from dissolution following an impact accident, then the maximum acceptable release fraction for truck transportation would be on the order of 5×10^{-3}, and for train transportation, 3×10^{-2}.

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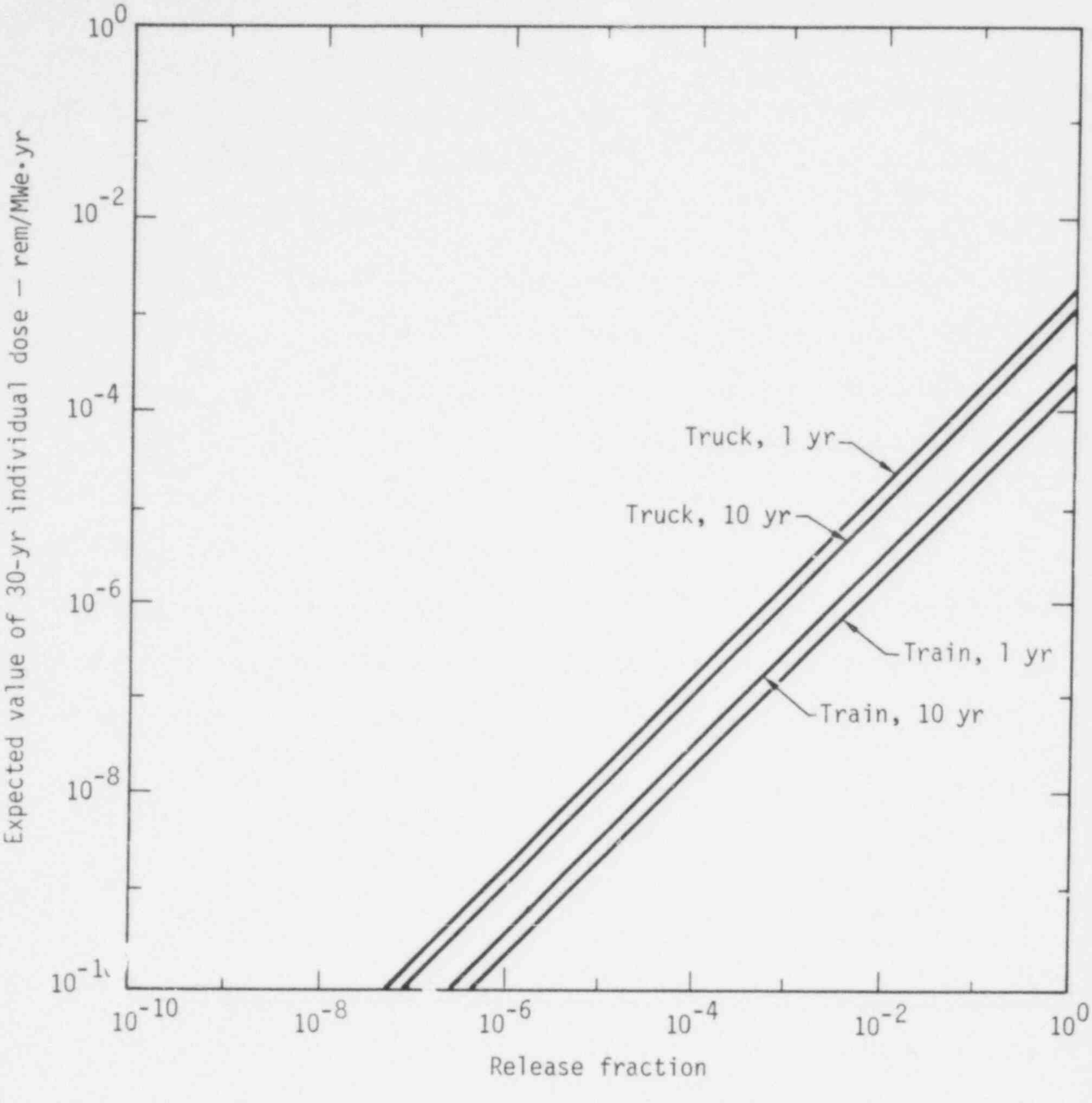

Fig. 6-7. Dependence of dose on release fraction: transportation, dissolution, and impact.

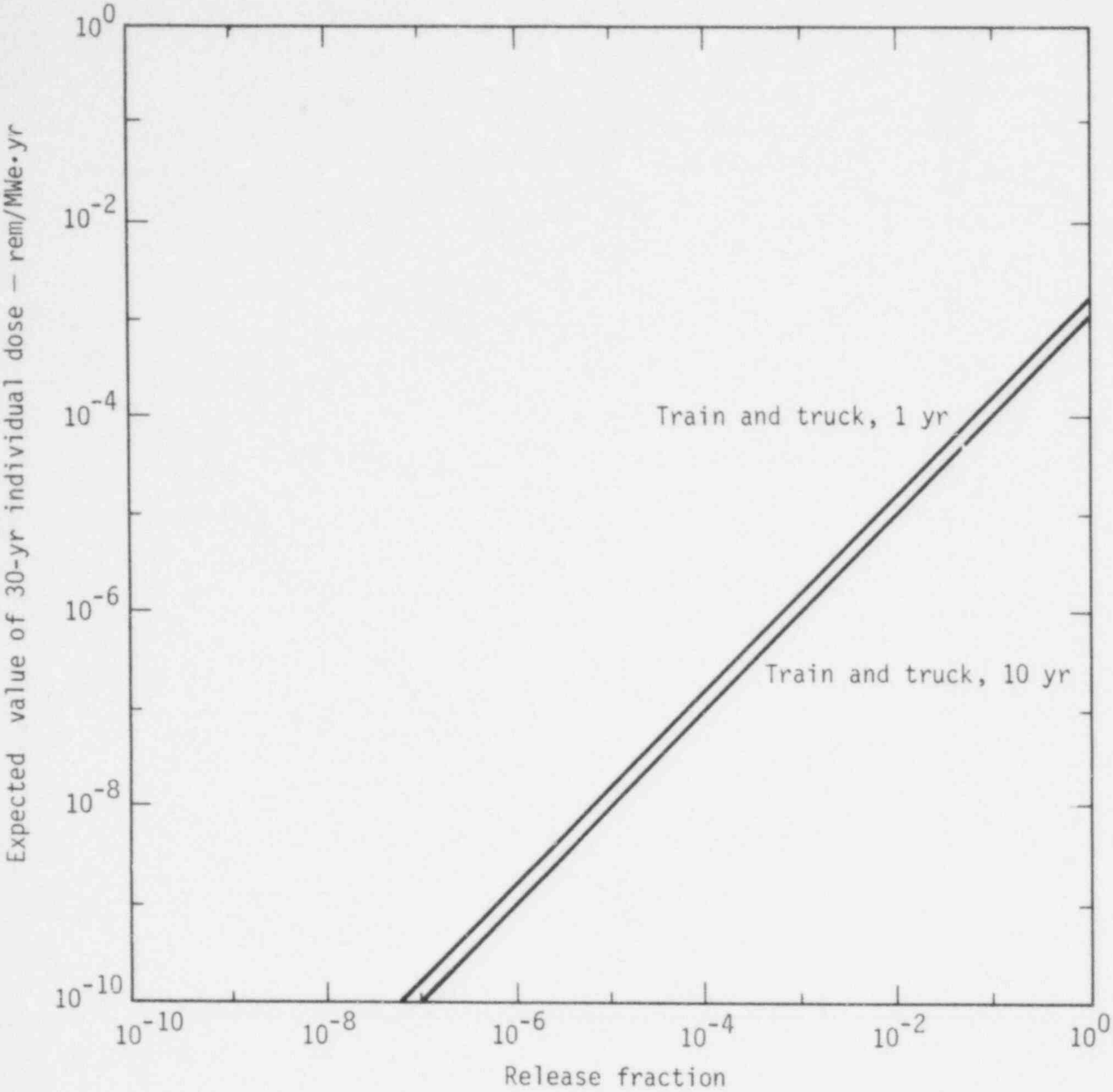

Fig. 6-8. Dependence of dose on release fraction: transportation, dissolution, and fire.

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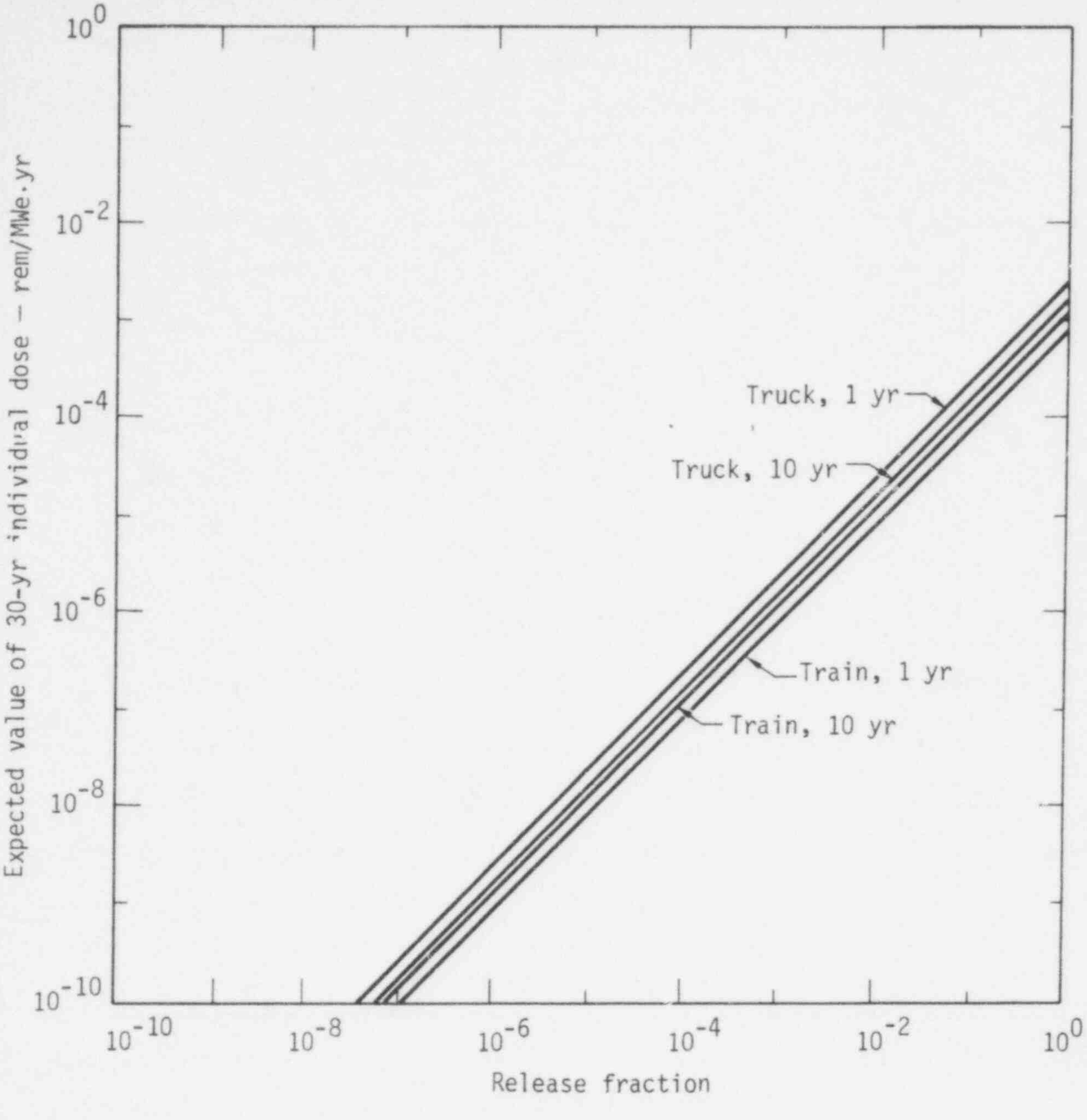

Fig. 6-9. Dependence of dose on release fraction: transportation, dissolution, and puncture.

6.2.5 Summary

This section has provided the sensitivity relations developed for critical accidents that can cause relatively significant expected population doses prior to the sealing of the waste repository. The use of these relations to specify solid-waste characterisitcs, as reflected in the fraction of available material released in a modal accident, was demonstrated.

6.3 SENSITIVITY ANALYSIS OF THE POST-EMPLACEMENT PERIOD

The sensitivity of doses to humans resulting from release of radioactive waste from an underground repository to the physical form of the waste was studied by analyzing functional dependences in the models described in Sec. 5 and Appendix C.

Only the water pathway described earlier was studied. The analysis was carried out to 10^6 years after sealing the repository. Assumptions were (1) that any type of seismic activity would result in leaching of waste into the aquifer, and (2) that the character of the aquifer would remain unchanged.

In the model of Appendix C, water enters the repository at some time after the repository is sealed. This time, whose probability density may be calculated from the geological model of Sec. 5.6, is taken as a parameter. The water then dissolves the radionuclides in the repository. The time required for this dissolution depends on the physical and chemical form of the waste. After flowing through an aquifer, the waste enters surface waters, passes through various ecological systems, and irradiates humans.

The calculations detailed in Appendix C indicate that the sensitivity of dose to dissolution time depends on the relationship between the dissolution time and a "dispersion time" that measures the spreading of a pulse of dissolved radionuclides in the aquifer. The integrated population dose resulting from escape of radioactivity and the "peak dose," which is the greatest dose that will ever be incurred by a member of the general population, behave somewhat differently. The sensitivities given by the model are as follows:

- If the dissolution time is less than the dispersion time (10^4 yr in the baseline case), neither integrated population dose nor peak dose will be sensitive to dissolution time.

- If the dissolution time is greater than the dispersion time, but less than 10^6 years, the integrated population dose will be independent of dissolution time, but the peak dose will be inversely proportional to the dissolution time.

- If the dissolution time exceeds 10^6 years, increases in dissolution time will reduce both integrated population dose and peak dose.

Figure 6-10 shows graphically the relationship between peak dose and dissolution time for a repository containing 10^6 megawatt electric·years of waste, which is the order of magnitude of the total waste generation expected by the year 2000. The peak dose is calculated for a maximum individual.

Some changes in assumptions that might increase the sensitivity of dose to dissolution time are:

- Inclusion of mechanisms that reseal underground fractures in the geological model.

- Use of very high groundwater velocity or very short path length.

- Withdrawal of contaminated water from the aquifer through wells.

6.3.1 Summary

This section has provided the results of sensitivity analysis of the impact of HLSW characteristics on expected radiation dose due to release from

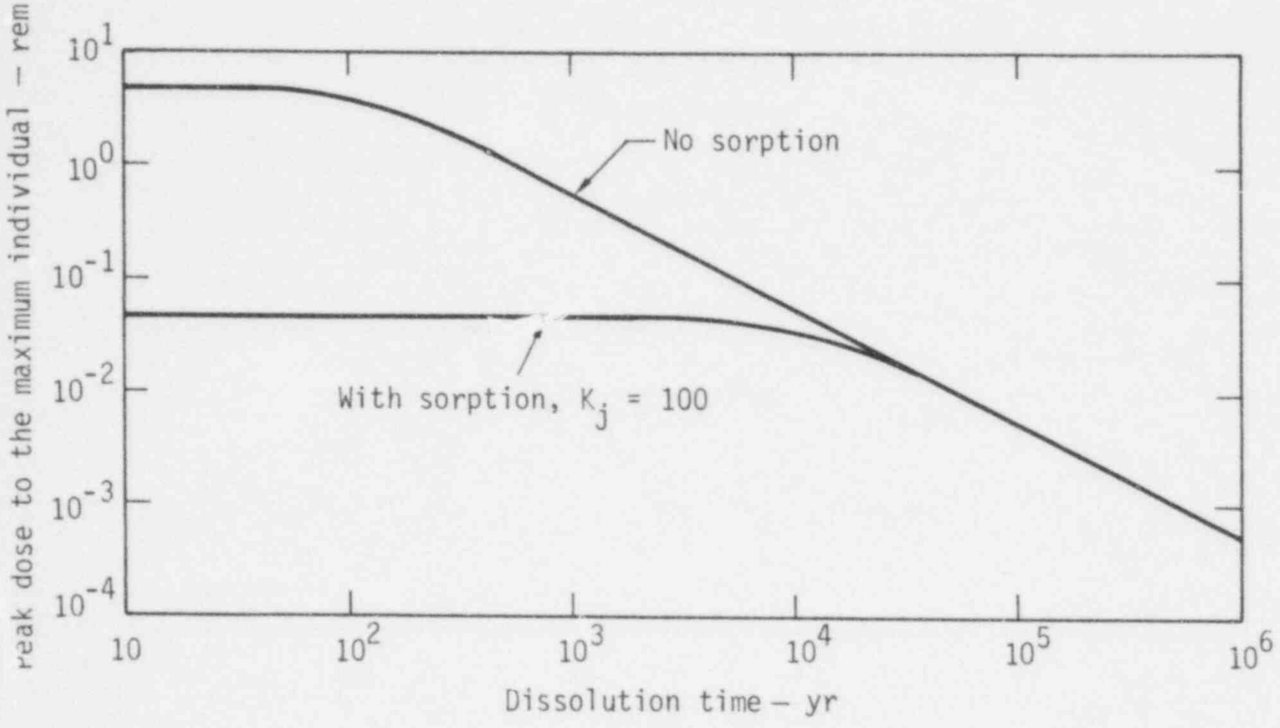

Fig. 6-10. Peak 50-year body dose to maximum individual from a 10^6-megawatt electric·year repository as a function of dissolution time.

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the waste repository. It is shown that there is little difference, in terms of integrated population dose or the maximum individual 50-year dose, over a wide range of dissolution rates. This indicates that the solubility of the solid waste is of little importance after the repository is sealed.

7 DISCUSSION

A preliminary objective of this study was to identify the conditions and environments to which HLSW could be subjected between the time of waste solidification at the FRP and the time the wastes have decayed to a level that would not be hazardous to public health and safety. The purpose of solidification is to render the radioactive constituents in the wastes into a less mobile form than that provided by the concentrated liquid solutions resulting from FRP process operations. By decreasing radionuclide mobility, constraints are placed on the transport mechanisms that could move radionuclides to and through the environment in the event untoward events occur.

The primary objective of this study is to identify the constraints that could be placed on the actual waste matrix such that, when considered with its canister and cask, the risk to public health and safety would be low for wastes subjected to the normal and accident environments identified herein. This report will be supplemented by a second, more detailed report in mid-1977 that will more quantitatively describe the constraints identified herein. Together, these reports will form the basis for a recommendation to NRC of performance criteria for HLSW matrices for use in a forthcoming proposed regulation.

The results obtained to date indicate that the pre-emplacement waste environs may be more limiting in establishing the waste-matrix performance criteria than the post-emplacement environs, considering both normal and potential accident conditions. These results are based upon waste emplacement in a reasonably stable geologic repository but do not rely upon the repository's remaining intact over the potentially hazardous lifetime of the waste.

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Our preliminary evaluations show the transportation phase of the waste-disposal system to be the most potentially hazardous due to both the variety of disruptive interactions and to demographic factors. These concerns may be partially, if not totally, mitigated by the large degree of protection afforded by the Type B casks in which the wastes will be transported. Studies currently in progress, which take into consideration the effect of the cask, will evaluate the potential for release during the transportation phase.

APPENDIX A: BIOLOGICALLY SIGNIFICANT RADIONUCLIDES IN HIGH-LEVEL WASTE

The relative composition of high-level wastes varies greatly as a function of time because of the different decay rates of the constituent radionuclides. Although the waste includes an abundant variety of radioactive species, at any given time only a relative few are potentially significant as biological hazards.

A.1 SOURCE TERM

A list of the major radioactive fission products and actinides that have intermediate or long half-lives is given in Table A-1. Source terms are listed for these nuclides as a function of time after fuel irradiation. Table input has been derived from two sources: for post-irradiation times up to 100 years, data from the *Siting of Fuel Reprocessing Plants* (1970) have been used. For longer times, data have been taken from Gera (1970).

The following assumptions pertain to the data in Table A-1:

- Fuel has been irradiated for 3.3 × 10^4 MWe/Mg of uranium at a thermal efficiency of 35.4%.
- The source of ^{129}I is decay of ^{129}Te, ^{129m}Te, and 0.1% of the ^{129}I present in the waste when it is dissolved. The actual quantity of ^{129}I present in high-level waste will be dependent on solidification time (assumed to be 150 da). If waste is not solidified until one year after irradiation, the amount present will be about one-half that shown in Table A-1.

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A.2 BIOLOGICALLY SIGNIFICANT RADIONUCLIDES: AIRBORNE

Tables A-2 and A-3 list the most biologically significant radioactive species evaluated by means of toxicity indices for airborne radioactivity. The toxicity index for each nuclide represents the volume of air that would have to be mixed with that quantity of the isotope formed during the production of one megawatt electric •year of power in order for the radionuclide concentration to be no greater than the maximum permissible concentration in air (MPC_a). Table A-2 gives toxicity indices calculated for the mixture of radionuclides existing one year after fuel irradiation; only those nuclides that have toxicity indices in the two orders of magnitude below that of the most toxic isotope (which is ^{90}Sr for the fission products and ^{244}Cm for the actinides) are included in the list. Table A-3 contains the same information as Table A-2 calculated for the mixture of radionuclides existing 10 years after fuel irradiation. Dose conversion factors have been calculated for all nuclides shown in Tables A-2 and A-3; results are given in Appendix B. Note that biologically significant airborne nuclides have not been identified for post-irradiation times greater than 10 years. Since proposed regulations require that all high-level waste be placed in an underground repository no more than 10 years after it has been generated, there is no credible mechanism whereby such waste can become airborne at times greater than 10 years after fuel irradiation.

It is interesting that all of these biologically significant airborne radioactive species have half-lives less than 10 000 years.

A.3 BIOLOGICALLY SIGNIFICANT RADIONUCLIDES: WATERBORNE

Tables A-4 through A-10 list the most biologically significant radioactive species, which were evaluated using toxicity indices for waterborne radioactivity. These indices are calculated in the same way as the airborne radioactivity indices except that the appropriate MPC_w values have been taken from 10 CFR 20.

A.4 REFERENCES

F. Gera, *Geochemical Behavior of Long-Lived Radioactive Waste*, Oak Ridge National Laboratory, Tenn., Rept. ORNL-TM-4481 (1970).

Siting of Fuel Reprocessing Plants and Waste Management Facilities, Oak Ridge National Laboratory, Tenn., Rept. ORNL-4451 (1970).

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Table A-1. Source terms of biologically significant nuclides (all units are Ci/MWe·yr).

Nuclide	150 da	1 yr	10 yr	10^2 yr	10^3 yr	10^4 yr
^{89}Sr	3.0×10^3	1.7×10^2	0	0	0	0
^{90}Sr	2.4×10^3	2.4×10^3	1.9×10^3	2.0×10^2	4.7×10^{-8}	0
^{90}Y	2.4×10^3	2.4×10^3	1.9×10^3	2.0×10^2	4.7×10^{-8}	0
^{91}Y	5.0×10^3	3.9×10^2	0	0	0	
^{93}Zr	5.9×10^{-2}	5.9×10^{-2}	5.9×10^{-2}	5.9×10^{-2}	5.9×10^{-2}	5.9×10^{-2}
^{93m}Nb	1.2×10^{-3}	2.9×10^{-3}	2.4×10^{-2}	5.9×10^{-2}	5.9×10^{-2}	5.9×10^{-2}
^{95}Zr	8.6×10^3	8.7×10^2	0	0	0	0
^{95}Nb	1.6×10^4	1.9×10^3	5.9×10^{-7}	0	0	0
^{99}Tc	4.4×10^{-1}	4.4×10^{-1}	4.4×10^{-1}	4.4×10^{-1}	4.4×10^{-1}	4.4×10^{-1}
^{106}Ru	1.3×10^4	8.5×10^3	1.7×10^1	0	0	0
^{106}Rh	1.3×10^4	8.5×10^3	1.7×10^1	0	0	0
^{125}Sb	2.5×10^2	2.2×10^2	2.2×10^1	0	0	0
^{126}Sn	1.7×10^{-2}	1.7×10^{-2}	1.7×10^{-2}	1.7×10^{-2}	1.7×10^{-2}	1.7×10^{-2}
^{129}I	2.4×10^{-6}	2.4×10^{-6}	2.4×10^{-6}	2.4×10^{-6}	2.4×10^{-6}	2.4×10^{-6}
^{134}Cs	6.7×10^3	5.5×10^3	2.6×10^2	0	0	0
^{137}Cs	3.3×10^3	3.3×10^3	2.7×10^3	3.3×10^2	3.1×10^{-7}	0
^{144}Ce	2.4×10^4	1.4×10^4	4.7×10^0	0	0	0
^{147}Pm	3.1×10^3	2.7×10^3	2.5×10^2	1.1×10^{-8}	0	0
^{154}Eu	2.1×10^2	2.1×10^2	1.4×10^2	2.9×10^0	0	0
^{210}Pb	0	0	0	2.3×10^{-8}	2.2×10^{-6}	8.8×10^{-5}
^{210}Po	0	0	0	2.3×10^{-8}	2.2×10^{-6}	8.8×10^{-5}
^{226}Ra	0	0	3.4×10^{-9}	3.5×10^{-8}	2.2×10^{-6}	8.8×10^{-5}
^{229}Th	0	0	1.3×10^{-9}	2.3×10^{-8}	2.2×10^{-6}	1.7×10^{-4}
^{230}Th	6.6×10^{-7}	6.6×10^{-7}	6.6×10^{-7}	1.0×10^{-6}	1.2×10^{-5}	1.1×10^{-4}

Table A-1. (Contd.)

Nuclide	150 da	1 yr	10 yr	10^2yr	10^3yr	10^4yr
^{231}Pa	7.7×10^{-7}	7.7×10^{-7}	7.7×10^{-7}	7.7×10^{-7}	8.1×10^{-7}	1.2×10^{-6}
^{233}U	4.6×10^{-7}	4.6×10^{-7}	4.6×10^{-7}	4.6×10^{-6}	4.8×10^{-5}	4.9×10^{-4}
^{237}Np	1.1×10^{-2}	1.1×10^{-2}	1.1×10^{-2}	1.1×10^{-2}	1.2×10^{-2}	1.2×10^{-2}
^{238}Pu	1.1×10^{0}	2.5×10^{0}	3.2×10^{0}	1.6×10^{0}	3.8×10^{-3}	0
^{239}Pu	5.2×10^{-2}	5.2×10^{-2}	5.2×10^{-2}	5.2×10^{-2}	6.4×10^{-2}	1.3×10^{-1}
^{240}Pu	7.6×10^{-2}	8.1×10^{-2}	1.4×10^{-1}	2.7×10^{-1}	2.5×10^{-1}	9.9×10^{-2}
^{241}Pu	1.8×10^{1}	1.7×10^{1}	1.0×10^{1}	1.5×10^{-1}	9.8×10^{-3}	4.6×10^{-3}
^{241}Am	5.4×10^{0}	5.4×10^{0}	5.5×10^{0}	5.1×10^{0}	1.1×10^{0}	4.6×10^{-3}
^{243}Am	5.4×10^{-1}	5.4×10^{-1}	5.4×10^{-1}	5.4×10^{-1}	5.0×10^{-1}	2.3×10^{-1}
^{242}Cm	4.7×10^{2}	1.9×10^{2}	2.2×10^{-1}	1.5×10^{-1}	2.4×10^{-3}	0
^{244}Cm	7.8×10^{1}	7.6×10^{1}	5.4×10^{1}	1.7×10^{0}	0	0

Table A-2. Inventory of biologically significant nuclides if released to air at 1 year after irradiation of 32 MWe·yr/Mg.

Nuclide	$T_{1/2}$	$MPC_a \left(\frac{Ci}{m^3}\right)$	$Q \left(\frac{Ci}{MWe \cdot yr}\right)$	Toxicity index $\left(\frac{m^3}{MWe \cdot yr}\right)$
Fission products				
^{89}Sr	53 da	3×10^{-10}	1.7×10^{2}	6×10^{11}
^{90}Sr	28 yr	3×10^{-11}	2.4×10^{3}	8×10^{13}
^{90}Y	64 hr	3×10^{-9}	2.4×10^{3}	8×10^{11}
^{91}Y	59 da	1×10^{-9}	3.9×10^{2}	4×10^{11}
^{95}Zr	66 da	1×10^{-9}	8.7×10^{2}	9×10^{11}
^{95}Nb	35 da	3×10^{-9}	1.9×10^{3}	6×10^{11}
^{106}Ru	1.0 yr	2×10^{-10}	8.5×10^{3}	4×10^{13}
^{106}Rh[a]	30 sec	1×10^{-9}	8.5×10^{3}	9×10^{12}
^{125}Sb	2.7 yr	9×10^{-10}	2.2×10^{2}	2×10^{11}
^{134}Cs	2.0 yr	4×10^{-10}	5.5×10^{3}	1×10^{13}
^{137}Cs	30 yr	5×10^{-10}	3.3×10^{3}	7×10^{12}
^{144}Ce	280 da	2×10^{-10}	1.4×10^{4}	7×10^{13}
^{147}Pm	2.6 yr	3×10^{-9}	2.7×10^{3}	9×10^{11}
^{154}Eu	16 yr	2×10^{-10}	2.1×10^{2}	1×10^{12}
Fission product total			5.1×10^{4}	2×10^{14}
Actinides				
^{238}Pu	86 yr	7×10^{-14}	2.5	4×10^{13}
^{240}Pu	6.6×10^{3} yr	7×10^{-14}	0.081	1×10^{12}
^{241}Pu	13 yr	3×10^{-12}	17.	6×10^{12}
^{241}Am	460 yr	2×10^{-12}	5.4	3×10^{13}
^{243}Am	8×10^{3} yr	2×10^{-13}	0.54	3×10^{12}
^{242}Cm	160 da	4×10^{-12}	190	5×10^{13}
^{244}Cm	18 yr	3×10^{-13}	76.	2×10^{14}
Actinide totals			2.9×10^{2}	3×10^{14}
Grand totals			5.1×10^{4}	5×10^{14}

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[a] In equilibrium with ^{106}Ru, MPC_w is an estimate.

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Table A-3. Inventory of biologically significant nuclides if released to air at 10 years after irradiation of 32 MWe·yr/Mg.

Nuclide	$T_{1/2}$	$MPC_a \left(\frac{Ci}{m^3}\right)$	$Q \left(\frac{Ci}{MWe \cdot yr}\right)$	Toxicity index $\left(\frac{m^3}{MWe \cdot yr}\right)$
Fission products				
^{90}Sr	28 yr	3×10^{-11}	1.9×10^{3}	6×10^{13}
^{90}Y	64 hr	3×10^{-9}	1.9×10^{3}	6×10^{11}
^{134}Cs	2.0 yr	4×10^{-10}	2.6×10^{2}	7×10^{11}
^{137}Cs	30 yr	5×10^{-10}	2.7×10^{3}	5×10^{12}
Fission product totals			6.8×10^{3}	7×10^{13}
Actinides				
^{238}Pu	86 yr	7×10^{-14}	3.2	5×10^{13}
^{240}Pu	6.6×10^{3} yr	6×10^{-14}	0.14	2×10^{12}
^{241}Am	460 yr	2×10^{-13}	5.5	3×10^{13}
^{243}Am	8×10^{3} yr	2×10^{-13}	0.54	3×10^{12}
^{244}Cm	18 yr	3×10^{-13}	54	2×10^{14}
Actinide totals			63	3×10^{14}
Grand totals			6.8×10^{3}	4×10^{14}

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Table A-4. Inventory of biologically significant nuclides if released to water at 1 year after irradiation at 32 MWe•/Mg.

Nuclide	$T_{1/2}$	$MPC_w \left(\frac{Ci}{m^3}\right)$	$Q \left(\frac{Ci}{MWe \cdot yr}\right)$	Toxicity index $\left(\frac{m^3}{MWe \cdot yr}\right)$
Fission products				
^{89}Sr	53 da	3×10^{-6}	1.7×10^{2}	6×10^{7}
^{90}Sr	28 yr	3×10^{-7}	2.4×10^{3}	8×10^{9}
^{90}Y	64 hr	2×10^{-5}	2.4×10^{3}	1×10^{8}
^{91}Y	59 da	3×10^{-5}	3.9×10^{2}	1×10^{7}
^{95}Zr	66 da	6×10^{-5}	8.7×10^{2}	1×10^{7}
^{95}Nb	35 da	1×10^{-4}	1.9×10^{3}	2×10^{7}
^{106}Ru	1.0 yr	1×10^{-5}	$8. \times 10^{3}$	9×10^{8}
^{106}Rh[a]	30 sec	1×10^{-4}	8.5×10^{3}	9×10^{7}
^{134}Cs	2.0 yr	9×10^{-6}	5.5×10^{3}	6×10^{8}
^{137}Cs	30 yr	2×10^{-5}	3.3×10^{3}	2×10^{8}
^{144}Ce	280 da	1×10^{-5}	1.4×10^{4}	1×10^{9}
^{147}Pm	2.6 yr	2×10^{-4}	2.7×10^{3}	1×10^{7}
^{154}Eu	16 yr	2×10^{-5}	2.1×10^{2}	1×10^{7}
Fission product totals			5.1×10^{4}	1×10^{10}
Actinides				
^{238}Pu	86 yr	5×10^{-6}	2.5	5×10^{5}
^{239}Pu	2.4×10^{4} yr	5×10^{-6}	0.052	1×10^{4}
^{240}Pu	6.6×10^{3} yr	5×10^{-6}	0.081	2×10^{4}
^{241}Pu	13 yr	2×10^{-4}	17.0	9×10^{4}
^{241}Am	460 yr	4×10^{-6}	5.4	1×10^{6}
^{243}Am	8×10^{3} yr	4×10^{-6}	0.54	1×10^{5}
^{242}Cm	160 da	2×10^{-5}	190	9×10^{6}
^{244}Cm	18 yr	7×10^{-6}	76	1×10^{7}
Actinide totals			2.9×10^{2}	2×10^{7}
Grand totals			5.1×10^{4}	1×10^{10}

---

[a] In equilibrium with ^{106}Ru, MPC_a is an estimate.

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Table A-5. Inventory of biologically significant nuclides if released to water at 10 years after irradiation at 32 MWe•yr/Mg.

Fission products Nuclide	$T_{1/2}$	$MPC_w \left(\frac{Ci}{m^3}\right)$	$Q \left(\frac{Ci}{MWe \cdot yr}\right)$	Toxicity index $\left(\frac{m^3}{MWe \cdot yr}\right)$
^{90}Sr	28 yr	3×10^{-7}	1.9×10^{3}	6×10^{9}
^{90}Y	64 hr	2×10^{-5}	1.9×10^{3}	9×10^{7}
^{134}Cs	2.0 yr	9×10^{-6}	2.6×10^{2}	3×10^{7}
^{137}Cs	30 yr	2×10^{-5}	2.7×10^{3}	1×10^{8}
Fission product total			6.8×10^{3}	6×10^{9}
Actinides				
^{238}Pu	86 yr	5×10^{-6}	3.2	6×10^{5}
^{241}Am	460 yr	4×10^{-6}	5.5	1×10^{6}
^{243}Am	8×10^{3} yr	4×10^{-6}	0.54	1×10^{5}
^{244}Cm	18 yr	7×10^{-6}	54	8×10^{6}
Actinide totals			63	1×10^{7}
Grand totals			6.9×10^{3}	6×10^{9}

735 077

Table A-6. Inventory of biologically significant nuclides if released to water at 100 years after irradiation of 32 MWe•yr/Mg.

Nuclide	$T_{1/2}$	$MPC_w\left(\frac{Ci}{m^3}\right)$	$Q\left(\frac{Ci}{MWe \cdot yr}\right)$	Toxicity index $\left(\frac{m^3}{MWe \cdot yr}\right)$
Fission products				
^{90}Sr	28 yr	3×10^{-7}	2.0×10^{2}	7×10^{8}
^{90}Y	64 hr	2×10^{-5}	2.0×10^{2}	1×10^{7}
^{137}Cs	30 yr	2×10^{-5}	3.3×10^{2}	2×10^{7}
Fission product totals			7.3×10^{2}	7×10^{8}
Actinides				
^{238}Pu	86 yr	5×10^{-6}	1.6	3×10^{5}
^{241}Am	460 yr	4×10^{-6}	5.1	1×10^{6}
^{243}Am	8×10^{3} yr	4×10^{-6}	0.54	1×10^{5}
^{244}Cm	18 yr	7×10^{-6}	1.7	2×10^{5}
Actinide totals			8.9	2×10^{6}
Grand totals			7.4×10^{2}	7×10^{8}

Table A-7. Inventory of biologically significant nuclides if released to water at 1000 years after irradiation of 32 MWe•yr/Mg.

Fission products Nuclide	$T_{1/2}$	$MPC_w \left(\frac{Ci}{m^3}\right)$	$Q \left(\frac{Ci}{MWe \cdot yr}\right)$	Toxicity index $\left(\frac{m^3}{MWe \cdot yr}\right)$
^{93}Zr	1.5×10^6 yr	8×10^{-4}	5.9×10^{-2}	7×10^1
^{93m}Nb	14 yr	4×10^{-4}	5.9×10^{-2}	1×10^2
^{99}Tc	2.1×10^5 yr	3×10^{-4}	4.5×10^{-1}	2×10^3
^{126}Sn[a]	1×10^5 yr	2×10^{-5}	1.7×10^{-2}	9×10^2
^{129}I	1.7×10^7 yr	6×10^{-8}	2.4×10^{-6}	4×10^1
Fission product totals			5.9×10^{-1}	3×10^3
Actinides				
^{237}Np	2.1×10^6 yr	3×10^{-6}	1.2×10^{-2}	4×10^3
^{239}Pu	2.4×10^4 yr	5×10^{-6}	6.4×10^{-2}	1×10^4
^{240}Pu	6.6×10^3 yr	5×10^{-6}	2.5×10^{-1}	5×10^4
^{241}Am	4.6×10^2 yr	4×10^{-6}	1.1×10^0	3×10^5
^{243}Am	8.0×10^3 yr	4×10^{-6}	5.0×10^{-1}	1×10^5
Actinide totals			1.9	5×10^5
Grand totals			2.5	5×10^5

---

[a] MPC assumed to be equal to the smallest of all other isotopes Sn. Present in secular equilibrium with ^{126}Sn are ^{126m}Sb and ^{126}Sb. If release pathway does not partition Sb and Sn, considerations should be given to these additional isotopes.

735 079

Table A-8. Inventory of biologically significant nuclides if released to water at 10^4 years after irradiation of 32 MWe·yr/Mg.

Fission products Nuclide	$T_{1/2}$	$MPC_w \left(\frac{Ci}{m^3}\right)$	$Q \left(\frac{Ci}{MWe \cdot yr}\right)$	Toxicity index $\left(\frac{m^3}{MWe \cdot yr}\right)$
^{93}Zr	1.5×10^6 yr	8×10^{-4}	5.9×10^{-2}	7×10^1
^{93m}Nb	14 yr	4×10^{-4}	5.9×10^{-2}	1×10^2
^{99}Tc	2.1×10^5 yr	3×10^{-4}	4.4×10^{-1}	2×10^3
$^{126}Sn^{a,b}$	1×10^5 yr	2×10^{-5}	1.6×10^{-2}	9×10^2
^{129}I	1.7×10^7 yr	6×10^{-8}	2.4×10^{-6}	4×10^1
Fission product totals			5.7×10^{-1}	3×10^3
Actinides (plus other alpha-emitters)[c]				
^{210}Pb	2.0×10^1 yr	1×10^{-7}	8.8×10^{-5}	9×10^2
^{210}Po	3.8×10^{-1} yr	7×10^{-7}	8.8×10^{-5}	1×10^2
^{226}Ra	1.6×10^3 yr	3×10^{-8}	8.8×10^{-5}	3×10^3
^{237}Np	2.1×10^6 yr	3×10^{-6}	1.2×10^{-2}	4×10^3
^{239}Pu	2.4×10^4 yr	5×10^{-6}	1.7×10^{-1}	3×10^4
^{240}Pu	6.6×10^3 yr	5×10^{-6}	9.9×10^{-2}	2×10^4
^{241}Am	4.6×10^2 yr	4×10^{-6}	4.6×10^{-3}	6×10^2
^{243}Am	8.0×10^3 yr	4×10^{-6}	2.3×10^{-1}	6×10^4
Actinide totals			5.2×10^{-1}	1×10^5
Grand totals			1.1×10^0	1×10^5

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[a] MPC is an estimate.

[b] Existing in equilibrium with ^{126}Sn are ^{126m}Sb and ^{126}Sb.

[c] Besides ^{210}Pb, ^{210}Po, and ^{226}Ra, other isotopes of the 4n+1 and 4n+2 decay series are present in the source in secular equilibrium (almost) with 1.7×10^{-4} Ci/MWe·yr of ^{229}Th and 1.0×10^{-4} Ci/MWe·yr of ^{230}Th.

Table A-9. Inventory of biologically significant nuclides if released to water at 10^5 years after irradiation of 32 MWe•yr/Mg.

Nuclide	$T_{1/2}$	$MPC_w \left(\frac{Ci}{m^3}\right)$	$Q \left(\frac{Ci}{MWe \cdot yr}\right)$	Toxicity index $\left(\frac{m^3}{MWe \cdot yr}\right)$
Fission products				
^{93}Zr	1.5×10^6 yr	8×10^{-4}	5.7×10^{-2}	7×10^1
^{93m}Nb	1.4×10^1 yr	4×10^{-4}	5.7×10^{-2}	1×10^2
^{99}Tc	2.1×10^5 yr	3×10^{-4}	3.3×10^{-1}	1×10^3
$^{126}Sn^{a,b}$	1×10^5 yr	2×10^{-5}	9.1×10^{-3}	5×10^2
^{129}I	1.7×10^7 yr	6×10^{-8}	2.4×10^{-6}	4×10^1
Fission product totals			4.5×10^{-1}	2×10^3
Actinides (plus other alpha-emitters)[c]				
^{210}Pb	2.0×10^1 yr	1×10^{-7}	7.0×10^{-4}	7×10^3
^{210}Pb	3.8×10^1 yr	7×10^{-7}	7.0×10^{-4}	1×10^3
^{226}Ra	1.6×10^3 yr	3×10^{-8}	7.0×10^{-4}	2×10^4
$^{229}Th^a$	7.3×10^3 yr	7×10^{-6}	3.7×10^{-3}	5×10^2
$^{230}Th^c$	8.0×10^4 yr	2×10^{-6}	6.9×10^{-4}	3×10^2
^{233}U	1.6×10^5 yr	3×10^{-5}	4.0×10^{-3}	1×10^2
^{237}Np	2.1×10^6 yr	3×10^{-6}	1.1×10^{-2}	4×10^3
^{239}Pu	2.4×10^4 yr	5×10^{-6}	1.8×10^{-2}	4×10^3
Actinide totals			3.9×10^{-2}	4×10^4
Grand totals			4.9×10^{-1}	4×10^4

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[a] The MPCs are estimates — lowest for all other isotopes of that element.

[b] Existing in equilibrium with ^{126}Sn are ^{126m}Sb and ^{126}Sb.

[c] Besides ^{210}Pb, ^{210}Po, and ^{226}Ra, other isotopes of the 4n+1 and 4n+2 decay series are present in the source in secular equilibrium with ^{229}Th and ^{230}Th.

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Table A-10. Inventory of biologically significant nuclides if released to water at 10^6 years after irradiation of 32 MWe•yr/Mg.

Nuclide	$T_{1/2}$	$MPC_w \left(\frac{Ci}{m^3}\right)$	$Q \left(\frac{Ci}{MWe \cdot yr}\right)$	Toxicity index $\left(\frac{m^3}{MWe \cdot yr}\right)$
Fission products				
^{93}Zr	1.5×10^6 yr	8×10^{-4}	3.9×10^{-2}	5×10^1
^{93m}Nb	1.4×10^1 yr	4×10^{-4}	3.9×10^{-2}	1×10^2
^{129}I	1.5×10^7 yr	6×10^{-8}	2.3×10^{-6}	4×10^1
Fission product totals			7.8×10^{-2}	2×10^2
Actinides (plus other alpha-emitters)[a]				
^{210}Pb	2.0×10^1 yr	1×10^{-7}	1.7×10^{-4}	2×10^3
^{210}Po	3.8×10^{-1} yr	7×10^{-7}	1.7×10^{-4}	2×10^2
^{226}Ra	1.6×10^3 yr	3×10^{-8}	1.7×10^{-4}	6×10^3
^{229}Th[a]	7.3×10^3 yr	7×10^{-6}	9.1×10^{-3}	1×10^3
^{230}Th	8.0×10^4 yr	2×10^{-6}	1.7×10^{-4}	9×10^1
^{231}Pa	3.3×10^4 yr	9×10^{-7}	1.0×10^{-5}	1×10^1
^{233}U	1.6×10^5 yr	3×10^{-5}	9.0×10^{-3}	3×10^2
^{237}Np	2.1×10^6 yr	3×10^{-6}	8.5×10^{-3}	3×10^3
Actinide totals			2.7×10^{-2}	1×10^4
Grand totals			1.1×10^{-1}	1×10^4

[a] Besides ^{210}Pb, ^{210}Po, and ^{226}Ra, other isotopes of the 4n + 1, 4n + 2, and 4n + 3 decay series are present in the source in secular equilibrium with ^{229}Th, ^{230}Th, and ^{231}Pa. MPC is an estimate — the lowest of all other isotopes of Th.

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APPENDIX B: DOSE CONVERSION FACTORS

B.1 AIRBORNE RADIOACTIVITY

Only those radioactive species having relatively short half-lives ($T_{1/2} < 10\ 000$ yr) were assumed to contribute significantly to radiation exposures resulting from airborne transport. Doses to members of the exposed population were considered to result from three processes:

- inhalation of radioactivity from the passing cloud,
- exposure to the gamma radiation from radioactivity deposited from the cloud onto the surface of the ground,
- ingestion of radioactivity that has entered into food chains after having been deposited on the ground from the cloud (only the forage-cow-milk-person pathway was considered).

In all cases, a short-term airborne release ("puff") rather than a long-term seep ("plume") of radioactivity is assumed. The following discussions present some features of the dose conversion factors.

B.1.1 Inhalation Dose Conversion Factors

The dose delivered over a 50-year period after radioisotope intake was determined for the critical organs of a "standard man" (*Recommendations of the International Commission on Radiological Protection*, 1959) for each of the nuclides of interest (see Table B-1) under the assumption of a short-term exposure to airborne radioactivity.

Uptake of the radioactivity was assumed to occur over a one-day period by inhalation; the total amount of air inhaled during this period is taken as

Table B-1. Dose conversion factors[a] for biologically significant nuclides in air: inhalation.

Nuclide	Critical-organ dose conversion factors (rem·m^3/Ci·yr)	Critical organ	Whole-body equivalent dose conversion factor (rem·m^3/Ci·yr)	Whole-body dose conversion factor (rem·m^3/Ci·yr)
Fission products				
^{89}Sr	3.0×10^{9}	Bone	5.0×10^{8}	8.4×10^{7}
^{90}Sr	3.3×10^{11}	Bone	5.5×10^{10}	1.6×10^{9}
^{91}Y	2.4×10^{9}	Bone	4.1×10^{8}	6.5×10^{7}
^{95}Zr	1.4×10^{9}	Lung	4.7×10^{8}	1.2×10^{8}
^{95}Nb	4.7×10^{8}	Lung	1.6×10^{8}	3.3×10^{7}
^{106}Ru	8.4×10^{9}	Lung	2.8×10^{9}	2.1×10^{7}
^{125}Sb	1.8×10^{9}	Lung	6.0×10^{8}	2.7×10^{7}
^{127m}Te	1.2×10^{9}	Lung	4.0×10^{8}	1.2×10^{7}
^{129m}Te	1.4×10^{9}	Lung	4.8×10^{8}	3.2×10^{7}
^{134}Cs	4.0×10^{9}	Lung	1.3×10^{9}	4.1×10^{8}
^{137}Cs	3.3×10^{9}	Lung	5.5×10^{8}	2.4×10^{8}
^{144}Ce	8.8×10^{9}	Bone	1.5×10^{9}	4.7×10^{8}
^{147}Pm	1.4×10^{9}	Bone	2.3×10^{8}	5.0×10^{7}
^{154}Eu	2.2×10^{10}	Bone	3.7×10^{9}	1.4×10^{9}
Actinides				
^{238}Pu	4.2×10^{13}	Bone	6.9×10^{12}	1.0×10^{12}
^{240}Pu	4.8×10^{13}	Bone	8.0×10^{12}	1.2×10^{12}
^{241}Pu	9.0×10^{11}	Bone	1.5×10^{11}	1.9×10^{10}
^{241}Am	1.5×10^{13}	Bone	2.5×10^{12}	9.9×10^{11}
^{243}Am	1.5×10^{13}	Bone	2.5×10^{12}	9.7×10^{11}
^{242}Cm	3.8×10^{11}	Liver	1.3×10^{11}	2.5×10^{10}
^{244}Cm	9.2×10^{12}	Bone	1.5×10^{12}	5.6×10^{11}

[a] For dose delivered over a 50-yr period following a brief period of inhalation of radioactivity.

735 084

20 m^3 (*Recommendations of the ICRP*, 1959). The dose conversion factors given in the tables of this appendix were calculated with the Oak Ridge INREM code (Turner *et al.*, 1968; Killough and McKay, 1976). Basically, this code uses the single-exponential model used by the International Commission to calculate "maximum permissible concentrations in air and water" of radioactivity, where, in the latter reference, chronic exposure is assumed.

Since the method of calculating the critical-organ dose commitment resulting from exposure to a given amount of radioactivity ingested over a finite period of time is outlined in detail by Turner *et al.* (1968) and by Killough and McKay (1976), this aspect of calculating the dose conversion factors used for inhalation will not be further amplified here. However, the method for converting the critical-organ dose conversion factor to the "whole-body equivalent" dose conversion factor warrants further discussion.

This latter conversion is based on the implicit equivalence of doses to various human organs as exemplified by the maximum permissible dose equivalents for occupational workers recommended by the International Commission (*Recommendations of the ICRP*, 1959) (see Table B-2). It can be seen that, for instance, a dose of 30 rem/yr is permissible to the skin, as opposed to 5 rem/yr to the whole body. Thus, one might infer that a dose of 1 rem to the skin incurs a risk to the exposed individual only one-sixth as great as that if the 1-rem dose had been delivered to the whole body. Consequently, the "whole-body equivalent" of a 1-rem dose to the skin would be 0.17 rem. This procedure has been followed to calculate the whole-body equivalent dose conversion factors in Table B-1. Again, the critical-organ dose conversion factors shown in Table B-1 have been calculated with the Oak Ridge INREM code (Turner *et al.*, 1968; Killough and McKay, 1976).

735 085

Table B-2. Maximum permissible dose equivalents recommended by the International Commission on Radiological Protection for occupational workers.

Organ	Maximum permissible dose equivalent in one year (rem)
Red bone marrow, whole body, and gonads	5
Skin, thyroid, and bone[a]	30
All other single organs	15

[a] Note that the National Council on Radiation Protection and Measurement currently specifies an annual maximum permissible dose of 15 rem for these organs as well (NCRPM, 1971).

B.1.2 Deposition Dose Conversion Factor: External Dose

Fifty-year integrated gamma-ray doses to the whole body resulting from the uniform deposition of the nuclides (see Table B-3) have been calculated, with the assumption that the "whole body" is being exposed at a point three feet above an infinite plane source of the radioactive species. The method of calculation has been discussed in detail (Higgins, 1963) and includes a correction for the air attenuation of the gamma radiation.

As noted above, only gamma energy released in the first 50 years after deposition of the radioactivity is considered in the dose conversion factors shown in Table B-3; for the shorter half-life nuclides, such as ^{89}Sr, essentially all this dose is delivered in the first year after deposition. For the very long-lived species, such as ^{243}Am, the dose rate is essentially constant over the period of interest. The gamma-ray energies used in these calculations for the nuclides of interest were obtained from Lederer *et al.* (1967).

It should be noted that these dose conversion factors have not been corrected for "weathering." The process of weathering will slowly move the radioactivity into the soil, thus increasing the gamma-ray attenuation and decreasing the dose delivered to a point three feet above the ground surface. Clearly, this process will not be important for the shorter-lived nuclides such as ^{89}Sr, but for the longer-lived isotopes (especially those having relatively low-energy gamma rays) the dose conversion factors in Table B-3 could be high by a factor of 2 or 3. However, no data exist whereby a reasonable correction for this effect can be estimated and justified.

735 087

Table B-3. Dose conversion factors[a] for radioactivity released to the atmosphere and deposited on the ground: external exposure.

Nuclide	Whole-body dose conversion factor (rem·m^2/Ci)
Fission product	
^{89}Sr	3.0
^{90}Sr	—
^{91}Y	1.3×10^{2}
^{95}Zr	2.9×10^{4}
^{95}Nb	1.8×10^{4}
^{106}Ru	6.9×10^{4}
^{125}Sb	2.4×10^{5}
^{127m}Te	7.2×10^{3}
^{129m}Te	6.8×10^{3}
^{134}Cs	8.3×10^{5}
^{137}Cs	3.1×10^{6}
^{144}Ce	9.6×10^{3}
^{147}Pm	—
^{154}Eu	1.0×10^{6}
Actinides	
^{238}Pu	2.9×10^{4}
^{240}Pu	8.6×10^{4}
^{241}Pu	2.5×10^{5}
^{241}Am	5.1×10^{5}
^{243}Am	3.1×10^{6}
^{242}Cm	3.0×10^{4}
^{244}Cm	3.5×10^{4}

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[a] For dose delivered over a 50-yr period following a single deposition of radioactivity on the ground.

B.1.3 Deposition Dose Conversion Factor: Internal Dose

Dose conversion factors have been calculated that give the internal dose (to a critical organ) delivered over a 50-year period following the deposition of a number of radioactive species whose concentrations are known for a given area. For the purpose of this approach, it has been assumed that the predominant pathway for the ingestion of radioactivity is through the forage-cow-milk-person food chain. Since the method used has been discussed in some detail by Ng and Thompson (1966) and more recently by Ng *et al*. (1976), this report will only indicate briefly the approach employed in the estimation of the dose conversion factors.

The dose to a given organ by means of the forage-cow-milk pathway is calculated as the product of the total activity ingested through milk per unit deposition and the 50-year dose per unit of activity ingested. The total activity ingested through milk (in μCi) is given by,

$$\int_0^{50\text{ yr}} I^*dt = \frac{Jr(\text{UAF})F(0)f^*_m}{\lambda_p} ,$$

where J = rate of milk consumption (liters/da), r = retention factor on forage, (UAF) = utilized area factor (i.e., the effective area of pasture grazed daily by the cow [m^2/da]), F(0) = deposition at time zero (μCi/m^2), f^*_m = transfer coefficient to milk (i.e., the fraction of the nuclide ingested daily by the cow that is secreted in milk per liter [da/liter]), and λ_p = effective rate of removal from forage (da^{-1}).

The dose per unit deposition through milk of Table B-4 are those calculated by Ng *et al*. (1976) for the adult. Assumptions are J = 1, r = 0.5,

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Table B-4. Dose[a] conversion factors for radioactivity released to the atmosphere and deposited on the ground: internal exposure by means of the forage-cow-milk-person pathway.

Nuclide	Critical-organ dose conversion factor (rem•m^2/Ci)	Critical organ[b]	Whole-body equivalent dose conversion factor (rem•m^2/Ci)	Whole-body dose conversion factor (rem•m^2/Ci)
Fission product				
^{89}Sr	8.1×10^3	Bone	1.3×10^3	1.1×10^3
^{90}Sr	1.9×10^5	Bone	3.3×10^4	1.5×10^4
^{91}Y	8.3×10^2	G.I.[c]	2.8×10^2	2.7
^{95}Zr	3.2×10^3	G.I.	1.1×10^3	2.1×10^1
^{95}Nb	9.5×10^2	G.I.	3.2×10^2	7.3
^{106}Ru	7.0×10^1	G.I.	2.3×10^1	1.4
^{125}Sb	3.4×10^2	G.I.	1.1×10^2	4.9
^{127m}Te	3.4×10^3	G.I.	1.1×10^3	5.0×10^1
^{129m}Te	6.9×10^3	G.I.	2.3×10^3	8.9×10^1
^{134}Cs	2.0×10^5	Whole body	2.0×10^5	2.0×10^5
^{137}Cs	1.5×10^5	Whole body	1.5×10^5	1.5×10^5
^{144}Ce	2.2×10^3	G.I.	7.2×10^2	3.4
^{147}Pm	1.0×10^2	G.I.	3.5×10^1	5.3×10^{-1}
^{154}Eu	5.9×10^2	G.I.	2.0×10^2	1.7×10^{-1}
Actinides				
^{238}Pu	2.3×10^1	Bone	3.8	6.7×10^{-1}
^{240}Pu	2.6×10^1	Bone	4.3	7.4×10^{-1}
^{241}Pu	5.6×10^{-1}	Bone	9.4×10^{-2}	1.4×10^{-2}
^{241}Am	1.8×10^5	Bone	3.0×10^4	5.1×10^3
^{243}Am	1.7×10^5	Bone	2.9×10^4	5.0×10^3
^{242}Cm	3.6×10^3	Bone	6.0×10^2	1.1×10^2
^{244}Cm	9.0×10^4	Bone	1.5×10^4	2.7×10^3

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[a] Dose delivered over a 50-yr period following a single deposition of radioactivity on forage.

[b] Inferred from *Recommendations of the ICRP* (1959).

[c] Gastrointestinal tract.

and (UAF) = 45. The half-residence time for nuclide particles on forage is assumed to be 14 days, which leads to the following expression for λ_p:

$$\lambda_p = \frac{\ln 2}{T_r} + \frac{\ln 2}{14} ,$$

where T_r is the half-life for radioactive decay, expressed in days. The transfer coefficient to milk f^*_m and the 50-year dose to the critical organs and whole body per unit of actively ingested $D'_{50\ yr}$ (μ rem/μCi) are listed in Ng *et al.* (1976).

Internal dose conversion factors were calculated both for the critical organ and for the whole body. Also, the whole-body equivalent dose conversion factors were calculated by the method discussed in Sec. B.1.1 and are shown in Table B-4.

B.2 WATERBORNE RADIOACTIVITY

Only the relatively long-lived radioactive species were assumed to contribute significantly to radiation exposures resulting from ground water transport. However, for the sake of completeness, a few of the shorter-lived species are included in this compilation. Critical-organ doses to members of the population who drink contaminated water can be calculated with dose conversion factors based on the values of the MPC_w, given in Appendix B, Table 2, of the *Nuclear Regulatory Commission Standards for Protection Against Radiation* (1960, 1976). These dose conversion factors (both for critical organs and the whole body), given in Table B-5, are calculated under the assumption of a long-term exposure (integrated over 50 yr) to water containing the

Table B-5. Dose conversion factors for biologically significant nuclides in water.

Nuclide	MPC_w[a] (Ci/m^3)	Critical[b] organ	Annual dose[b,c] (rem)	Critical-organ dose conversion factor[d] ($rem \cdot m^3/Ci$)	Whole-body equivalent dose conversion factor[d] ($rem \cdot m^3/Ci$)	Whole-body MPC_w,[e] (Ci/m^3)	Whole-body dose conversion factor[d] ($rem \cdot m^3/Ci$)
<u>Fission product</u>							
^{90}Sr[f]	3×10^{-7}	Bone	3	3×10^{8}	5×10^{7}	1×10^{-6}	1×10^{7}
^{93}Zr	8×10^{-4}	G.I.[g]	1.5	9×10^{4}	3×10^{4}	6×10^{-1}	4×10
^{99}Tc	3×10^{-4}	G.I.	1.5	3×10^{5}	8×10^{4}	1×10^{-2}	3×10^{3}
^{126}Sn	2×10^{-5}	G.I.	1.5	4×10^{6}	1×10^{6}	2×10^{-3}	1×10^{4}
^{129}I	6×10^{-8}	Thyroid	0.5	4×10^{8}	4×10^{8}	8×10^{-6}	3×10^{6}
^{137}Cs[f]	2×10^{-5}	Whole body	0.5	7×10^{5}	7×10^{5}	2×10^{-5}	7×10^{5}
<u>Actinides</u>							
^{210}Pb[h]	1×10^{-7}	Kidney	1.5	7×10^{8}	3×10^{8}	1×10^{-7}	3×10^{8}
^{210}Pb[h]	7×10^{-7}	Spleen	1.5	1×10^{8}	4×10^{7}	8×10^{-6}	3×10^{6}
^{226}Ra[h]	3×10^{-8}	Bone	3	5×10^{9}	8×10^{8}	6×10^{-8}	4×10^{8}
^{237}Np	3×10^{-6}	Bone	3	5×10^{7}	8×10^{6}	1×10^{-5}	3×10^{6}
^{238}Pu[f]	5×10^{-6}	Bone	3	3×10^{7}	5×10^{6}	4×10^{-5}	6×10^{5}
^{239}Pu	5×10^{-6}	Bone	3	3×10^{7}	5×10^{6}	3×10^{-5}	8×10^{5}
^{240}Pu	5×10^{-6}	Bone	3	3×10^{7}	5×10^{6}	3×10^{-5}	8×10^{5}
^{241}Am	4×10^{-6}	Kidney	1.5	2×10^{7}	6×10^{6}	1×10^{-5}	3×10^{6}
^{243}Am	4×10^{-6}	Bone	3	4×10^{7}	6×10^{6}	1×10^{-5}	3×10^{6}
^{244}Cm[f]	7×10^{-6}	Bone	3	1×10^{7}	2×10^{6}	2×10^{-5}	7×10^{5}

[a] From NCRPM (1960, 1976).

[b] Inferred from *Recommendations of the ICRP* (1959).

[c] Steady state exposure at MPC_w.

[d] For integral dose from 50 yr of exposure.

[e] For annual dose of 0.5 rem; inferred from *Recommendations of the ICRP* (1959).

[f] Assuming an initial concentration equal to MPC_w and allowing for radioactive decay.

[g] Gastro-intestinal tract.

[h] In equilibrium with parent ^{230}Th.

735 092

radionuclides listed in the first column. The appropriate values of MPC_w are also listed in Table B-5 for the radioactivities of interest (both for the critical organs and the whole body), as are the annual doses that would be delivered to the critical organs by exposure to the indicated concentrations of radioactivity. (These doses were inferred by using the MPC_w values found in *Recommendations of the ICRP* [1959].) The critical organs were also identified through use of these references. Once again, the whole-body equivalent dose conversion factors were determined using the approach outlined in Sec. B.1.1.

735 093

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APPENDIX C: HYDROLOGIC MODEL

GENERAL CHARACTERISTICS

The water-release model of Burkholder (1975) was taken as a point of departure for modeling flow of radionuclides from repository to biosphere. The model assumes that radionuclides flow through an aquifer of constant cross-sectional area. To make this model more realistic, the following modifications were made:

- Start of leaching was assumed to begin at the time of the first earthquake or breach of the repository. This corresponds to the time of first transition from Group I in the Markov chain model.
- The dispersion coefficient was assumed to be equal to the flow velocity multiplied by 50 meters.
- The velocity of water flow through the aquifer was assumed to be 0.1 foot per day rather than 1.0 foot per day.
- Buildup of radionuclides in topsoil, sediment, and the human population was explicitly modeled.

All of these modifications tend to make the model less conservative than the model used by Burkholder (1975).

FLOW IN AN AQUIFER

Consider the motion of a nuclide through an aquifer after it has been released from a repository, neglecting radioactive decay for the moment. The waste moves at velocity ν/K_j, where ν is the velocity of the water in the aquifer and K_j is the sorption retardation factor for the nuclide in question. In the aquifer, hydraulic dispersion causes narrow pulses to spread. Only

735 096

longitudinal spreading need be considered; lateral spreading will not affect the time of release into surface waters. We therefore treat motion in one dimension only and describe it by a diffusion-type differential equation (Grove, 1970):

$$\frac{\partial c}{\partial t} = - \frac{\nu}{K_j} \frac{\partial c}{\partial x} + \frac{\alpha\nu}{K_j} \frac{\partial^2 c}{\partial x^2} , \qquad \text{(C-1)}$$

with $\alpha\nu = D$, the dispersion coefficient. This equation has a well-known solution by the method of Green's function, with

$$G(x,t;\xi,\tau) = \sqrt{\frac{K_j}{2\pi\alpha\nu(t - \tau)}} \; \exp\left\{ - \frac{\left[x - \xi - \frac{\nu}{K_j} (t - \tau) \right]^2}{\frac{2\alpha\nu}{K_j} (t - \tau)} \right\} . \qquad \text{(C-2)}$$

The Green's function, G, is the response of the aquifer to a unit impulse input at ξ,τ. If the input into the aquifer is a pulse $c_{in}(\xi,\tau)$, the output from the aquifer will be

$$c_{out} = \iint G \, c_{in} \, d\xi d\tau \quad . \qquad \text{(C-3)}$$

For ease of calculation, assume that the input pulse, which represents the course of dissolution of waste in the repository, has a Gaussian form:

$$c_{in}(\xi) = Q_j \frac{\lambda_\ell K_j}{\nu \pi^{1/2}} \; \exp\left\{ \frac{-\lambda_\ell^2 \xi^2 K_j{}^2}{\nu^2} \right\} \quad (\tau = 0) \; ,$$

where Q_j is the total amount of nuclide j in the repository and λ_ℓ^{-1} is the dissolution time of the waste. This choice involves the implication that waste begins to leave the repository before containment is breached. However, it will be shown that the final answer is not sensitive to the particular pulse shape chosen.

Performing the integral in Eq. C-3 gives

$$c_{out}(t) = \frac{Q_j}{\sqrt{\pi\left(\frac{2\alpha\nu t}{K_j} + \frac{\nu^2}{\lambda_\ell^2 K_j^2}\right)}} \exp\left\{\frac{-\left(z - \frac{\nu t}{K_j}\right)^2}{\frac{\nu^2}{K_j^2\lambda_\ell^2} + \frac{2\alpha\nu t}{K_j}}\right\} .$$

The substitution $t = K_j z/\nu$ gives the maximum release rate

$$c_{out}^{max} = \frac{Q_j}{\sqrt{\pi\left(2\alpha z + \frac{\nu^2}{\lambda_\ell^2 K_j^2}\right)}} . \qquad \text{(C-4)}$$

In the limit where

$$\frac{\nu^2}{\lambda_\ell^2 K_j^2} << 2\alpha z \quad , \qquad \text{(C-5)}$$

the maximum release rate will be independent of the dissolution time λ_ℓ^{-1}. In the opposite limit, the maximum release rate will be inversely proportional to the dissolution time.

735 098

The condition of Eq. C-5 is equivalent to

$$\lambda_{\ell}^{-1} << T_D = \sqrt{2\alpha z} \; \frac{K_j}{\nu} \qquad \text{(C-6)}$$

where T_D (the dispersion time) is defined as the time over which dispersion spreads a pulse.

For the baseline case without ion exchange (α = 50 m, z = 10 mi, K_j = 1, and ν = 11.1 m/yr), this formula gives $T_D = 10^2$ year. If ion exchange is included with K_j = 100, $T_D = 10^4$ year.

The behavior of release rates in the two limiting cases does not depend on the shape of the input pulse.

Consider first the case where the condition of Eq. C-5 is satisfied. The input pulse will then be relatively narrow. Its structure will be entirely obliterated by the spreading due to hydraulic dispersion, and the shape of the output pulse will depend only on how much waste is in the input pulse. This situation is represented mathematically by a Green's function that is much wider than the input pulse. The input pulse may be approximated by a delta function. The output will have the same time dependence as the Green's function, with its amplitude proportional to the total amount of waste in the input pulse.

If, on the other hand, the input pulse is wide and smooth, hydraulic dispersion will have little effect and the output will have approximately the same shape as the input. This situation corresponds to an input pulse much wider than the Green's function. The amplitude of the output in this case will simply be equal to the amplitude of the input.

This argument shows that the behavior of a pulse of radionuclides in an aquifer is governed by the relationship between the initial width of the pulse, λ_{ℓ}^{-1}, and the length of time across which hydraulic dispersion spreads the pulse, T_D. If the pulse is initially much longer than the spreading time, the output pulse will have the same maximum amplitude as the input pulse. If the pulse is initially much shorter than the spreading time, however, the maximum amplitude of the output will depend on the total quantity of waste contained in the pulse and not on the rate at which waste enters the aquifer at any instant.

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735 101

APPENDIX D: METEOROLOGY AND DEMOGRAPHY

D.1 METEOROLOGY

In the event of an accidental release of solid radioactive waste at a fuel reprocessing plant (FRP), along a transportation route, or at a FWR, a portion of the waste might become airborne and thereby affect populations downwind. Therefore, it is necessary to estimate the dispersion and deposition of such debris. For our calculations, we assume a unit-curie release at one meter above the ground. The initial source is assumed to have a one meter radius and half-thickness. At the FRP or the FWR, it is possible to have either a continuous (plume) release, an instantaneous (puff) release, or perhaps something in between these two extremes. A transportation accident would most likely result in an instantaneous release.

The Gaussian diffusion equations for these two types of releases at the surface are given below. If the plume is sector-averaged over 22.5°,

$$\chi/Q = \left(\frac{2}{\pi}\right)^{3/2} \frac{4}{\sigma_z \, \bar{u} \, x} \exp \left(- \frac{1}{2} \, \frac{h^2}{\sigma_z{}^2}\right), \qquad \text{(D-1)}$$

where χ = surface air concentration on plume centerline (Ci/m^3), Q = source term (Ci/sec), $\bar{u}$ = mean surface wind speed (m/sec), σ_z = vertical standard deviation of wind (m), x = distance downwind (m), and h = height of plume center above ground (m).

For a puff,

$$\chi/Q = \frac{1}{2 \, \pi^{3/2} \, \sigma_{x_i} \, \sigma_{y_i} \, \sigma_{z_i}} \exp - \left(\frac{1}{2} \, \frac{h^2}{\sigma_{z_i}{}^2}\right), \qquad \text{(D-2)}$$

where Q is in curies and the subscripts signify instantaneous values of the standard deviation in the x (downwind), y (crosswind), and z (vertical) directions. Since a plume meanders about an average trajectory at an angular variation dependent on the atmospheric stability while a puff is influenced only by the winds along its trajectory, the σ values for a plume are larger than those for a puff in similar conditions of stability and windspeed.

In addition to surface concentration, surface deposition has been incorporated into the calculations. This modification of the diffusion equations involves the use of a deposition velocity that represents the rate of particulate impaction with the surface due to turbulent eddies near the ground. A deposition velocity of 0.01 meters per second has been assumed, and the above equations have been modified to include plume depletion as deposition occurs.

For the purpose of an accident analysis, we have considered both an average and a "worst" case. The average was made by assuming a Pasquill-Gifford "D" stability with a five-meter-per-second wind speed. To determine the worst case, we have estimated the upper envelope of concentration for both plumes and puffs in the following stability categories and windspeeds:

Stability category	Windspeed, m/sec
A	1
B	3
C	4
D	5
E	2
F	1

From 0.1 to 100 kilometers from the source, average- and worst-case analytical equations for average, unit-source continuous and instantaneous surface releases (plume and puff, respectively) are given in Table D-1. Graphs of

Table D-1. Analytical expressions for estimating relative centerline concentrations[a] from a solid nuclear waste accident; χ/Q is relative concentration (yr/m^3), x is distance downwind (m), plume or puff height is 1 m, and deposition velocity is 3×10^5 m/yr.

Type of estimate	Plume	Puff
Average-case χ/Q		
(D stability, $\bar{u}$ = 5 m/sec)	$5.6 \times 10^{-8}\ x^{-1.7}$	$3.8 \times 10^{-4}\ x^{-3.7}$
Worst-case χ/Q		
(upper envelope)	$1.3 \times 10^{-6}\ \chi^{-2}$	$4.1 \times 10^{-3}\ x^{-3.9}$

[a] Equivalent expressions for the relative surface deposition rate (in units of m^{-2}) may be obtained by multiplying the coefficients of the expressions in the table by 3×10^5 m/yr, the deposition velocity.

these functions are shown in Figs. D-1 and D-2. Similar equations for the deposition rate (in m^{-2}) can be obtained by multiplying Eqs. (D-1) and (D-2) by 0.01, the deposition velocity in meters per second.

Concentrations obtained from the worst-case expression for a plume have been compared with NRC guidance for nuclear power stations (*Preparation of Environmental Reports*, 1976, p. 7-1). At a distance of 100 meters from an accident, the expression for a plume (Table D-1) yields a χ/Q about a factor of 5 greater than the value obtained from *Preparation of Environmental Reports*. The concentrations from both methods are the same at six kilometers. At 100 kilometers, the Table D-1 expression gives a concentration about a factor of 3 less than that indicated by *Preparation of Environmental Reports*.

D.2 DEMOGRAPHY

In an estimate of the total population dose (out to 100 km) from an accident involving solid waste, population is best expressed by analytic

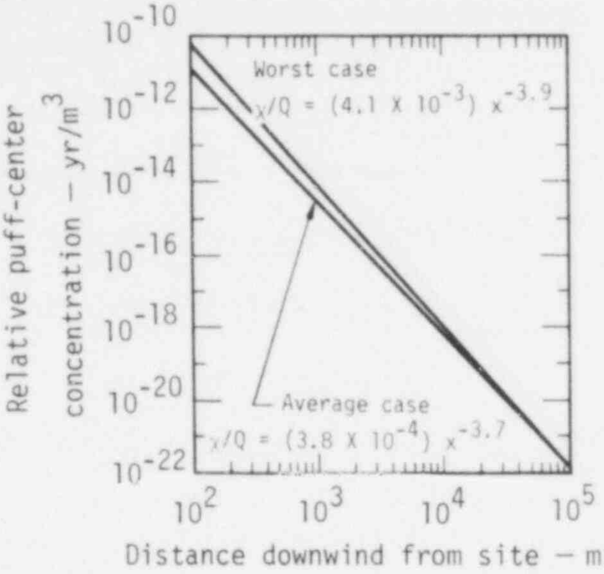


Fig. D-1. Graph of analytical expressions for a plume. Release height is one meter; sector averaging was used.

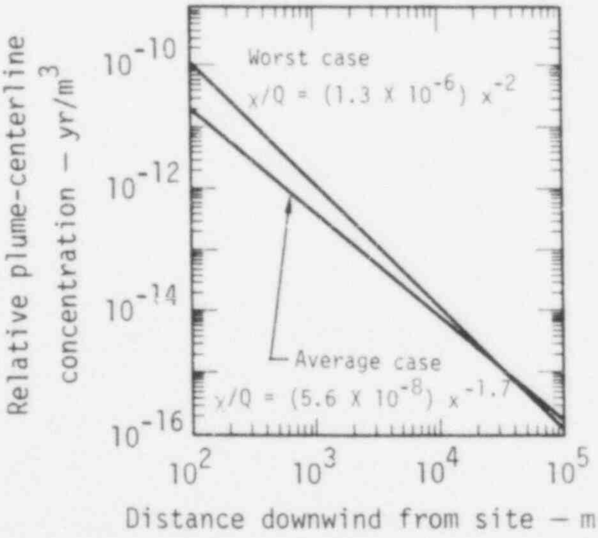


Fig. D-2. Graph of analytical expressions for a puff. Release height is one meter.

equations. Consideration of population density (the number of persons per unit of area) simplifies the formulation of such equations.

It is reasonable to assume that the rural population density ρ will be nearly uniform. In and around towns and cities, the population density can be approximated by Gaussian curves (of the form given by: a exp[-bx]). General expressions for such population densities are:

1. From the accident site to the nearest city ($0 < x < x_c$),

$$\rho = (\rho_c - \rho_s) \exp [-b_1 (x - x_c)^2] + \rho_s; \tag{D-3}$$

2. From the city to outlying rural areas ($x > x_c$),

$$\rho = (\rho_c - \rho_r) \exp [-b_2 (x - x_c)^2] + \rho_r, \tag{D-4}$$

where b_1 and b_2 are coefficients determining the shape of the Gaussian curves, and where x = distance downwind from the accident (m), x_c = distance from the accident to the city (m), ρ_s = population density at the accident site (persons/m^2), ρ_c = population density in the city (persons/m^2), and ρ_r = rural population density (persons/m^2).

D.2.1 Fuel Reprocessing Plant Demography

It is reasonable to assume that future FRPs will be located in relatively sparsely populated regions. Such plants will likely have small towns (population less than 10 000) within easy commuting distance.

Since it represents the state of the art in reprocessing plants, the Barnwell Nuclear Fuel Plant (BNFP) (see *Environmental Statement...Barnwell*, 1974), located on the eastern edge of the Savannah River Plant (SRP) in South Carolina, was selected as a typical site for defining the constants in Eqs. (D-1) and (D-2). The BNFP has a special advantage of having the sparsely populated SRP to the west. Therefore, only the population to the east was considered. The BNFP environmental statement was used in determining the regional demography. The constants substituted into Eqs. (D-1) and (D-2) (ρ

in units of persons/m^2) are: $b_1 = 0.08$, $b_2 = 0.05$, $x_c = 1.2 \times 10^4$ m (distance from FRP to Barnwell), $\rho_s = 5 \times 10^{-6}$ persons/m^2, $\rho_c = 1 \times 10^{-4}$ persons/m^2 (average population density of Barnwell), and $\rho_r = 2.2 \times 10^{-5}$ persons/m^2 (average population density of U.S.).

The specific equations for this site are then:

1. Within 12 kilometers of the FRP,

$$\rho = 9.5 \times 10^{-5} \exp[-0.08(0.001x - 12)^2] + 5 \times 10^{-6}, \qquad \text{(D-5)}$$

2. Beyond 12 kilometers,

$$\rho = 7.8 \times 10^{-5} \exp[-0.05(0.001x - 12)^2] + 2.2 \times 10^{-5}. \qquad \text{(D-6)}$$

Figure D-3 is a plot of population-density Eqs. (D-5) and (D-6) for an FRP; however, in this figure ρ has units of persons per square kilometer.

D.2.2 Transportation Demography

Urban Site — The population distribution in the Dallas-Fort Worth area was modeled as a typical demography for transportation accidents with solid radioactive waste in urban areas. This particular region was selected because:

- The rail lines and truck routes through Dallas-Fort Worth could be used to transport solid nuclear waste to a sparsely populated FWR that may be located in the southwestern United States.

- Central Texas has a high occurrence of tornadoes. Although it is not likely, a large tornado could pick up and rupture a solid-waste transportation cask.

- Dallas-Fort Worth was the tenth largest metropolitan area in the U.S. in 1973 (*Statistical Abstract*, 1975).

For the purposes of this study, we assume an accident in the center of the metropolitan area (area of maximum population density). Also, we assume that the population density falls off in a Gaussian fashion to the rural area. The 1973 population density of the Dallas-Fort metropolitan area (*World*

735 107

Almanac, 1976) was 114 persons per square kilometer. The total metropolitan area is 21 700 square kilometers. Within the city limits of Dallas, the 1970 population density was 1230 persons per square kilometer, while the area was 688 square kilometers (*Statistical Abstract*, 1975). If we assume that the city limits form a circle, it would have a radius of 15 kilometers; the radius of the metropolitan area under the same assumption would be 83 kilometers. Using these assumptions, one can determine the Gaussian equation for the population density ρ in units of persons per square meter as:

$$\rho = 1.26 \times 10^{-3} \exp(-1 \times 10^{-9} x^2) \ , \tag{D-7}$$

where x is the radial distance in meters from the center of the city. Figure D-4 shows a plot of Eq. (D-7) with ρ in persons per square kilometer. The coefficient of 1.26×10^{-3} was chosen so that the curve between 0 and 1.5×10^4 meters (radius of the city limits) gives equal areas of the city above and below the average population density of 1.23×10^{-3} persons per square meter.

Rural Site — The demography of a rural transportation accident can be assumed uniform over distance. Then, $\rho = \rho_r$ at all distances, where ρ_r is the 1973 U.S. average population density of 2.2×10^{-5} persons per square meter.

D.2.3 Federal Waste Repository Demography

The population distribution (*Selected Census...NTS*, 1973) around the Nevada Test Site (NTS) has been used in this study as typical of that around a FWR site. The city of Las Vegas dominates the population density within a few hundred kilometers of NTS. Therefore, since the wind blows toward Las Vegas less than 5% of the time, its population has been omitted in order to obtain an "expected value" population dose. Hence, the population was averaged over 16-kilometer bands, excluding the population of Las Vegas. With the NTS population data fitted to a Gaussian curve by Eqs. (D-3) and (D-4), the equations for population density ρ in units of persons per square meter are:

1. Within 45 kilometers,

$$\rho = 3.8 \times 10^{-7} \exp[-4 \times 10^{-3}(0.001x - 45)^2], \qquad \text{(D-8)}$$

2. Beyond 45 kilometers,

$$\rho = 3.8 \times 10^{-7} \exp[-4 \times 10^{-4}(0.001x - 45)^2] + 2.0 \times 10^{-7} . \qquad \text{(D-9)}$$

Note that the population of the FWR has been assumed virtually zero. It is expected that only a few guards would be on duty during normal circumstances. Figure D-5 shows a graph of these equations with ρ in persons per square kilometer.

D.3 AREA INTEGRALS OF THE PRODUCT OF CONCENTRATION, AREA, AND POPULATION DENSITY

If we obtain the integral of $\chi/Q \cdot A \cdot \rho$ (where A is the area under consideration), we need then only multiply by the appropriate dose conversion factors to obtain the total population dose (in man•rem) out to 100 kilometers over the swath of the plume or puff. Since the three quantities χ/Q, A, and ρ are interrelated with respect to distance or, equivalently, time, one must integrate their product over either distance or time to obtain population dose. Let this integral be denoted as $\sum$.

For a plume, Eq. (D-1) incorporates sector-averaging over a 22.5° sector. The integral is,

$$\sum = \int_{10^2}^{10^5} \frac{\chi}{Q} \rho \, dA, \qquad \text{(D-10)}$$

735 109

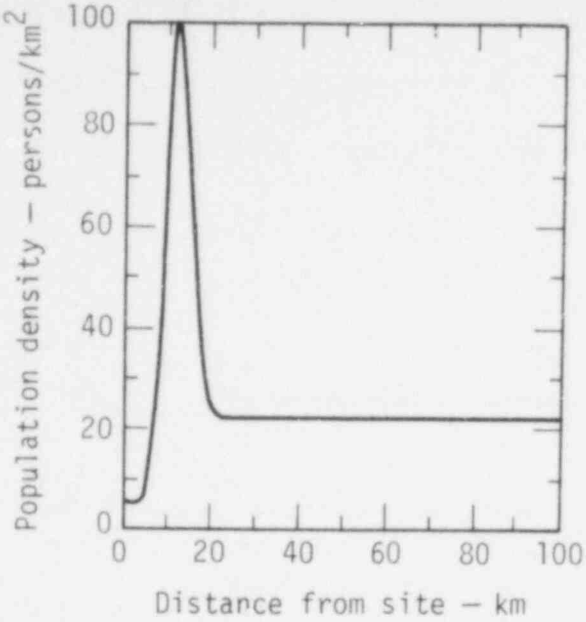
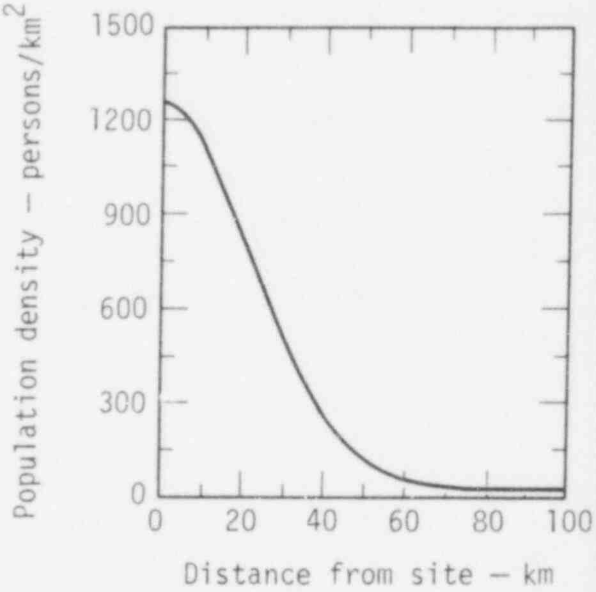


Fig. D-3. Population density vs distance from a hypothetical accident at a fuel reprocessing plant with a town of 5000 12 kilometers away.

Fig. D-4. Population density vs distance from a hypothetical transportation accident at the center of an urban area.



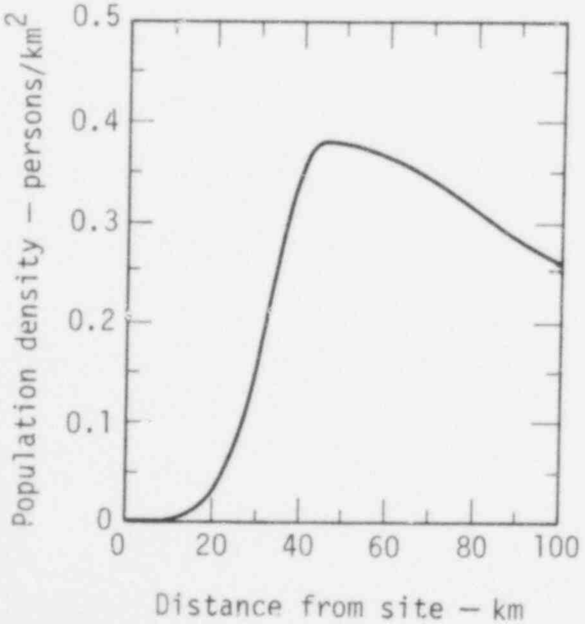


Fig. D-5. Population density vs distance from a hypothetical accident at a federal nuclear waste repository.

735 110

where $dA = \frac{\pi}{8} \cdot dx$, x in meters. Substitution of the appropriate χ/Q expression for Table D-1 and the population-density equation from the previous subsection yields an integral equation with distance downwind as the sole variable. Numerical integration out to 10^5 meters results in the plume values shown in Table D-2.

For the puff, the approach is similar, except that the integral is different (one cannot sector-average a puff):

$$\sum = \int \left(\frac{\bar{\chi}}{Q} A\right) \rho \, dt \; . \tag{D-11}$$

Since $dt = \frac{1}{u} dx$ (u represents wind speed), the equation becomes:

$$\sum = \frac{1}{\bar{u}} \int_{10^2}^{10^5} \left(\frac{\bar{\chi}}{Q} A\right) \rho \; dx \; . \tag{D-12}$$

We have considered an expanding puff whose radius at a given x is equal to two standard deviations ($\sigma_x = \sigma_y$) and derived the average concentration χ/Q over

Table D-2. Integrated values of ($\frac{\chi}{Q} \cdot \rho \cdot A$) for plumes and puffs in various accident senarios. Units are man$\cdot$yr/m^3.

	Plume		Puff	
Accident scenario	Average value	Worst value	Average value	Worst value
FRP	4.9×10^{-11}	1.1×10^{-10}	9.6×10^{-11}	(Same as average value in all scenarios)
Transportation				
Rural	4.5×10^{-11}	7.6×10^{-11}	8.7×10^{-11}	
City	1.6×10^{-9}	3.4×10^{-9}	2.9×10^{-9}	
FWR	3.2×10^{-13}	5.7×10^{-13}	6.7×10^{-13}	

735 111

A at each distance x. Since area and concentration both use the same σs at distance x, it is convenient to evaluate their product for all xs. Hence, for the average value (D stability, u = 5 m/sec),

$$\frac{\bar{\chi}}{Q} A = 4.3\, x^{-0.65}, \tag{D-13}$$

where $\bar{\chi}/Q$ is the average concentration of radioactivity (in units of sec/m^3) at a distance x, and over an area A of $\pi(2\sigma)^2$. To within a factor of 2, this expression for a puff also gives the concentration for the upper envelope of all stability categories. Numerical integration of the equation that results from substituting into Eq. (D-12) results in the puff values given in Table D-2. The products of the values in this table and the appropriate dose conversion factors in Appendix B yield estimates of population doses in man•rem.

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