

250
7-27-77
SAND77-0369
Unlimited Release
UC-70

DR 1258

Neutron and Gamma-Ray Sources In LWR High-Level Nuclear Waste

Stephen A. Dupree



Sandia Laboratories

SF 2600 Q17-731

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Issued by Sandia Laboratories, operated for the United States Energy Research & Development Administration by Sandia Corporation.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research & Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

Printed in the United States of America

Available from
National Technical Information Service
U. S. Department of Commerce
6285 Port Royal Road
Springfield, VA 22161

Price: Printed Copy \$3.50; Microfiche \$3.00

SAND77-0369
Unlimited Release
Printed June 1977

Distribution
Category UC-70

NEUTRON AND GAMMA-RAY SOURCES
IN LWR HIGH-LEVEL NUCLEAR WASTE

Stephen A. Dupree
Theoretical Division, 5231
Sandia Laboratories
Albuquerque, NM 87115

ABSTRACT

Predictions of the composition of high-level waste from U-fueled LWRs have been used to calculate the neutron and gamma-ray sources in such waste at cooling times of 3 and 10 years. The results are intended for interim application to studies of waste shipping and storage pending the availability of more exact knowledge of fuel recycling and of waste concentration and solidification.

NOTICE
This report was prepared as an account of work sponsored by the United States Government. While the United States and the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, make any warranty, express or implied, or assume any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, it represents that its use would not infringe privately owned rights.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

CONTENTS

	<u>Page</u>
Introduction	7
Projected Composition of LWR High-Level Waste	7
Calculation of Source Strength	8
Conclusions	16
References	16

TABLES

<u>Table</u>		<u>Page</u>
I	Calculated Actinide Activities in High-Level Waste From 1 MT of LWR U Fuel at 3- and 10-Year Decay Following Reprocessing	9
II	Calculated Fission Product Activities in High-Level Waste From 1 MT of LWR U Fuel at 3- and 10-Year Decay Following Reprocessing	10
III	Calculated Fission Product Gamma-Ray Source Intensity per cm^3 of Solidified LWR High-Level Waste at 3- and 10-Year Decay Following Reprocessing	11
IV	Significant Contributing Fission Product Isotopes Calculated for Gamma-Ray Source Intensity in LWR High-Level Waste	12
V	Calculated Actinide Gamma-Ray Source Intensity per cm^3 of Solidified LWR High-Level Wastes at 3- and 10-Year Decay Following Reprocessing	13
VI	Calculated Neutron Source Intensity per cm^3 of Solidified LWR High-Level Waste at 3- and 10-Year Decay Following Reprocessing	14

NEUTRON AND GAMMA-RAY SOURCES IN LWR HIGH-LEVEL NUCLEAR WASTE

Introduction

Expected intensities of neutron and gamma-ray sources present in high-level nuclear waste depend on the history and composition of the particular waste. Characteristics of the reactor in which the waste was generated, the extent to which recycle chemistry is performed on the waste, the degree of concentration or solidification of the residual material, and the cooling time before and after recycling — all contribute to the radiation source intensities in a specific piece of waste material. Any attempt to generalize the neutron and gamma-ray source strengths in high-level nuclear waste must therefore constitute a compromise of some sort.

The purpose of this report is to provide an interim source definition for high-level radioactive waste from light-water reactor (LWR) fuel reprocessing. Pending the availability of more exact knowledge of fuel recycling and of waste concentration and solidification, the results should be useful to programs requiring such sources. The uncertainty in the results presented here — although unknown — is certainly large, at least a factor of 2.

Projected Composition of LWR High-Level Waste

High-level wastes are defined¹ 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 wastes are presumed to contain virtually all the generated fission products, several tenths of a percent of the U and Pu originally present in the spent fuel, and all other actinides generated during the irradiation cycle. Regulations call for these wastes to be solidified within a period of 5 years from the time the liquid is generated (one of the first steps in the fuel recycling) and to be shipped to a government repository within 10 years from the time the liquid is generated.

The present study uses the results of an analysis by Blomeke et al.,² as a basis for defining the neutron and gamma-ray sources in LWR high-level wastes. The source data presented here pertain only to U-fueled LWRs,² and the fuel is assumed to be reprocessed following a 150-day postirradiation decay period.

¹The Blomeke analysis assumed the initiation of Pu recycle in the LWR fuels for 1979. Although this date may now appear premature, the assumption of Pu recycle at some point in the future is certainly reasonable.

Using models for the chemical treatment expected in spent-fuel recycling, including calcine and glassy product formation, Blomeke et al. predicted the volume of recycled waste, excluding cladding, to lie in the neighborhood of 1 to 2 ft³ per metric ton (MT) of LWR fuel charged to the reactor. Despite the uncertainty regarding these numbers, when one considers the likelihood of some dilution of the fully irradiated material with waste from partially irradiated malfunctioning fuel assemblies, equipment decontaminations, and other relatively low-intensity sources, the upper-limit volume of 2 ft³ of solidified high-level waste per metric ton is believed to be the most realistic number now available.⁶

All tritium and noble-gas fission products, as well as 99.9 percent of the I and Br fission products, are assumed to be removed from the waste during the initial reprocessing steps and, accordingly, are omitted from consideration in the source definition. The residue of U and Pu in the final solidified waste is assumed to be 0.5 percent of that initially present in the spent fuel. No cladding or structural material is assumed to be present in the end-product solidified waste.

The LWR fuel is assumed to be enriched to 3.34 percent in ²³⁵U. The power level is assumed to be 30 MW/MT, with a total burnup of 33 MWd/kg of heavy metal. The average neutron flux seen by the fuel during the irradiation period was 2.92×10^{13} n/cm²·s. The actinide and the fission product compositions of waste from 1 MT of LWR fuel⁴ derived from these assumptions are shown in Tables I and II.

It should be pointed out that some familiar isotopes are omitted from these tables because they do not contribute significantly to the radiation source in the waste material in the time frame of interest here.

Calculation of Source Strength

Given the isotopic composition of the waste material of interest, calculation of the gamma-ray source strength is straightforward insofar as the nuclear energy levels and cascade schemes of the pertinent nuclei are known. Determination of the neutron source strength from spontaneous fission is likewise straightforward; however, certain assumptions must be made to obtain an estimate of the neutron source from charged-particle reactions.

For present purposes the gamma emission rates for the various isotopes of Tables I and II were taken from the ORIGEN library.⁴ This library was assembled by the Oak Ridge National Laboratory as a data base for the ORIGEN code. The isotope decay schemes in the library are taken from Lederer et al.⁵ Bremsstrahlung is included by assuming UO₂ as the matrix in which β -decay occurs.

⁶In an update to Reference 1, Kee et al.³ revised the estimate for the volume of waste from 1 MT of fuel charged to the reactor to 1 to 3 ft³, with the upper limit as the recommended value. This, along with revisions in their actinide inventory, indicates that the present neutron results are conservative by approximately a factor of 2, whereas the present gamma-ray results are conservative by approximately 50 percent (both on a per cm³ basis).

TABLE I

Calculated Actinide Activities in High-Level Waste From 1 MT of LWR U Fuel at 3- and 10-Year Decay Following Reprocessing^a

Activity (Ci)		
Isotope	3-Year Decay	10-Year Decay
²³⁴ Th	1.57-3 ^b	1.57-3
²³³ Pa	3.40-1	41-1
^{234m} Pa	1.57-3	1.57-3
²³⁴ U	4.45-3	6.32-3
²³⁶ U	1.44-3	1.44-3
²³⁷ U	1.07-2	7.60-3
²³⁸ U	1.57-3	1.57-3
²³⁷ Np	3.40-1	3.41-1
²³⁹ Np	1.82-1	1.82-1
²³⁸ Pu	9.69+1	9.29+1
²³⁹ Pu	1.62	1.62
²⁴⁰ Pu	3.08	4.33
²⁴¹ Pu	4.46+2	3.20+2
²⁴² Pu	6.91-3	6.93-3
²⁴¹ Am	1.60+2	1.63+2
^{243m} Am	9.02	8.74
²⁴² Am	9.02	8.74
²⁴³ Am	1.82+1	1.82+1
²⁴² Cm	1.68+2	7.17
²⁴³ Cm	3.44	2.96
²⁴⁴ Cm	2.14+3	1.64+3
²⁴⁵ Cm	3.41	3.41-1
²⁴⁶ Cm	6.83-2	6.83-2

^aReprocessing is assumed to take place 150 days after completion of the fuel irradiation period.

^bHead 1.57-3, for example, as 1.57×10^{-3} .

TABLE II

Calculated Fission Product Activities in High-Level Waste From 1 Mf of LWR U Fuel at 3- and 10-Year Decay Following Reprocessing^a

Activity (Ci)		
Isotope	3-Year Decay	10-Year Decay
⁷⁸ Se	3.08-1	3.98-1
⁹⁰ Sr	7.13+4	6.00+4
⁹⁰ Y	7.13+4	6.00+4
⁹³ Zr	1.89	1.89
^{93m} Nb	4.25-1	3.63-1
⁸⁹ Tc	1.43-1	1.43-1
¹⁰⁶ Ru	5.08+4	4.06+2
¹⁰⁵ Rh	5.08+4	4.06+2
¹⁰⁷ Pd	1.10-1	1.10-1
^{110m} Ag	1.18+2	1.07-1
¹¹⁰ Ag	1.53+1	1.39-2
^{113m} Cd	8.86	6.26
¹²⁵ Sb	3.70+3	6.06+2
^{125m} Te	1.51+3	2.51+2
¹²⁶ Sn	5.46-1	5.46-1
^{126m} Sb	5.46-1	5.46-1
¹²⁶ Sb	5.41-1	5.41-1
¹³⁴ Cs	7.70+4	7.22+3
¹³⁵ Cs	2.86-1	2.86-1
¹³⁷ Cs	9.86+4	8.47+4
^{137m} Ba	5.31+4	7.02+4
¹⁴⁴ Ce	5.19+4	1.01+2
¹⁴⁴ Pr	5.19+4	1.01+2
¹⁴⁷ Pm	4.40+4	6.90+3
¹⁵¹ Sm	1.22+3	1.15+3
¹⁵² Eu	1.03+1	6.86
¹⁵⁴ Eu	6.02+3	4.45+3
¹⁵⁵ Eu	2.01+3	1.37+2

^a Reprocessing is assumed to take place 150 days after completion of the fuel irradiation period.

It, cause the matrix of the waste material is expected to be borosilicate glass or other low-Z material, this assumption produces a conservative over estimate of the gamma-ray source strength at energies below approximately 1 MeV. The gamma source library also includes photon emission from the $^{18}\text{O}(\alpha, n)^{21}\text{N}$ reaction, which is a significant source of neutrons in some types of spent fuel.

The total fission product gamma-ray source calculated for high-level LWR waste at 3 and 10 years after reprocessing is given in Table III. The source is normalized to 1 cm³ of solidified waste, with an assumption of 2 ft³ of waste per metric ton of fuel charged to the reactor. The major contributors to the source in each energy group in the time frame of interest are shown in Table IV. It is apparent that a preponderance of the gamma source is contributed by a very few fission product isotopes over this time period. For a more extensive presentation⁶ of the isotopes important to gamma production in high-level waste as a function of time, see Figure 8 of Cohen.⁷

TABLE III
Calculated Fission Product Gamma-Ray Source Intensity per cm³ of Solidified
LWR High-Level Waste at 3- and 10-Year Decay Following Reprocessing^a

Energy Group ^b	Upper Energy Bound (MeV)	Group Average Energy (MeV)	Photons/cm ³ ·s Emitted in Group	
			3-Year Decay	10-Year Decay
5	3.5	3.25	1.4+5	1.1+3
6	3.0	2.75	4.4-6	3.6-4
7	2.6	2.38	5.6-7	4.5-5
8	2.2	1.99	3.6+8	1.7-8
9	1.8	1.55	1.9+9	1.6-8
10	1.35	1.10	6.5+9	2.3+9
11	0.9	0.63	2.0+11	6.8+10
12	0.4	0.30	1.5-10	3.0-9
	0.2			

^a Reprocessing is assumed to take place 150 days after completion of the fuel irradiation period.

^b Fission product gamma source in Groups 1-4 is zero (see Table V for energy structure).

⁶Cohen apparently used the same assumptions as Blomeke et al.

TABLE IV

Significant Contributing Fission Product Isotopes Calculated for Gamma-Ray Source Emission in LWR High-Level Waste^a

Energy Group ^b	Upper Energy Bound (MeV)	Isotope	Photons cm ⁻² s Emitted in Group	
			3-Year Decay	10-Year Decay
5	3.5	¹⁰⁶ Rh	1.4+5	1.1+3
6	3.0	¹⁰⁶ Rh	4.4+6	3.6+4
7	2.6	¹⁰⁶ Rh	5.6+7	4.5+5
8	2.2	¹⁴⁴ Pr	2.8+8	5.5+5
		¹⁰⁶ Rh	7.6+7	6.1+5
		⁹⁰ Y	6.3+5	5.3+5
9	1.8	¹³⁴ Cs	1.5+9	1.4+8
		¹⁰⁶ Rh	2.5+7	2.0+6
		¹⁴⁴ Pr	1.2+8	2.4+5
		⁹⁰ Y	2.1+7	1.8+7
		¹⁵⁴ Eu	2.8+9	2.1+9
10	1.35	¹⁰⁶ Rh	1.7+9	1.4+7
		¹³⁴ Cs	1.6+9	1.5+8
		¹³⁴ Cs	1.2+11	1.1+10
11	0.9	^{137m} Ba	6.4+10	5.4+10
		¹⁰⁶ Rh	5.1+9	4.1+7
12	0.4	¹⁴⁴ Ce	3.4+9	6.6+6
		¹⁴⁴ Pr	2.6+9	5.0+6
		⁹⁰ Y	2.3+9	1.7+9
		¹³⁷ Cs	6.6+8	5.6+8

^a Reprocessing is assumed to take place 150 days after completion of the fuel irradiation period.

^b Fission product gamma source in Groups 1-5 is zero.

Although the fission products contribute almost all of the gamma source in LWR high-level waste, the actinides are the source of some gamma radiation due to spontaneous fission and other minor reactions. The gamma-ray sources from the actinides, including daughter products, are given in Table V. Except for the fact that the actinides contribute the only gamma rays present above 3.5 MeV (Groups 1-4), their contribution to the total gamma-ray source is negligible and Table III may be considered to be the total gamma source. The actinide gamma source is shown here only for completeness.

TABLE V

Calculated Actinide Gamma-ray Source Intensity per cm^3 of Solidified LWR High-Level Wastes at 3- and 10-Year Decay Following Reprocessing^a

Energy Group	Upper Energy Bound (MeV)	Group Average Energy (MeV)	Photons $\cdot \text{cm}^{-3} \cdot \text{s}^{-1}$ Emitted in Group	
			3-Year Decay	10-Year Decay
1	5.5	5.25	2.2-1	1.7-1
2	5.0	4.70	3.5-1	2.7-1
3	4.5	4.22	7.4-1	5.6-1
4	4.0	3.70	1.2-2	8.9-1
5	3.5	3.25	1.8-2	1.4-2
6	3.0	2.75	2.9-2	2.2-2
7	2.6	2.38	6.3-2	4.8-2
8	2.2	1.99	1.3-3	9.6-2
9	1.8	1.55	2.4-3	1.8-3
10	1.35	1.17	2.4-6	2.4-6
11	0.9	0.63	9.6-6	9.6-6
12	0.4	0.32	2.0-6	2.0-6
	0.25 ^b			

^a Reprocessing is assumed to take place 150 days after completion of the fuel irradiation period.

^b Note that the lower bound of group 12 differs from that of Tables III and IV. Omitted from this table are 7.6×10^7 photons/ $\text{cm}^3 \cdot \text{s}$ emitted by the actinides at energies less than 250 keV.

Neutron production by the actinides predicted for LWR high-level waste at 3 and 10 years after reprocessing is shown in Table VI. Contributions of the U and Np isotopes and of ^{245}Cm , ^{246}Cm , and ^{239}Pu to the total neutron production rate are insignificant in this time frame and therefore have been omitted from Table VI. Contributions due to spontaneous fission and (α, n) reactions are shown separately. The number of prompt neutrons emitted per spontaneous fission were taken from Devillers and Blum.⁷ The (α, n) reaction rate, taken from Bell,⁴ assumes the presence of an oxide matrix for the $^{15}\text{O}(\alpha, n)^{21}\text{Ne}$ reaction in which half of the slowing down experienced by the alpha particles is due to the oxygen. For a more extensive presentation of the isotopes contributing to the neutron source as a function of time, see Figure 11 of Chen.⁶

TABLE VI

Calculated Neutron Source Intensity per cm^3 of Solidified LWR High-Level Waste at 3- and 10-Year Decay Following Reprocessing^a

Isotope	Spontaneous Fission Neutron Production Rate (neutrons/ $\text{cm}^3 \cdot \text{s}$)		$^{18}\text{O}(\alpha, n)^{21}\text{N}$ Neutron Production Rate (neutrons/ $\text{cm}^3 \cdot \text{s}$)	
	3-Year Decay	10-Year Decay	3-Year Decay	10-Year Decay
^{238}Pu	2.5-1	2.4-1	1.6	1.5
^{240}Pu	2.1-1	3.0-1	4.1-2	5.7-2
^{241}Pu	0	0	4.8	3.4
^{241}Am	0	0	2.6	2.6
$^{242\text{m}}\text{Am}$	0	0	1.2-1	1.2-1
^{242}Am	0	0	1.2-1	1.2-1
^{243}Am	0	0	2.6-1	2.6-1
^{242}Cm	1.7+1	7.4-1	3.8	1.6-1
^{243}Cm	0	0	6.7-2	5.8-2
^{244}Cm	<u>5.1+3</u>	<u>3.9+3</u>	<u>4.2+1</u>	<u>3.2+1</u>
	5.1-3	3.9+3	5.5+1	4.6+1

Total neutron production rate (neutrons/ $\text{cm}^3 \cdot \text{s}$)

3-year decay	5.2+3
10-year decay	3.9+3

^a Reprocessing is assumed to take place 150 days after completion of the fuel irradiation period.

The energy spectrum of the neutrons from spontaneous fission is reasonably well known and, for practical purposes, can be assumed to be equal to a ^{235}U prompt fission neutron spectrum (presented in numerous references). The neutrons produced by the (α, n) reactions are somewhat harder than an average prompt fission neutron. Their energies generally lie in the range of approximately 2 to 4 MeV; in the present case, however, because the (α, n) neutrons constitute only a small fraction of the total neutron source, it is safe to assume that all neutrons have a prompt fission neutron energy spectrum. From Table VI it is clear that ^{244}Cm may be expected to be the dominant source of neutrons in LWR high-level waste at the cooling times of interest.

Figures 8 and 11 of Reference 6 show the total gamma-ray power emitted and the neutrons/s emitted from all high-level nuclear waste generated in 400 GWe-years of operation of LWRs. In a fully developed nuclear power economy this amount of power is expected to be generated each year by nuclear reactors.

Current plans indicate that the high-level waste will be placed in metal canisters for transport to a permanent disposal site. Cohen's waste storage canister constitutes approximately 7.85 ft^3 , or about $2.2 \times 10^6 \text{ cm}^3$, of solidified waste. Figure 11 of Reference 6 shows the neutron emission rate for each of these canisters. The 400 GWe-years of reactor operation is expected to generate approximately 4000 such canisters. Therefore, the present results can be compared directly with those of Cohen. Upon converting the gamma source of Table III to watts and scaling to 4000 canisters, one obtains 1.9×10^7 watts of gamma power at 3 years and 5.5×10^6 watts of gamma power at 10 years. Likewise, converting the neutron source of Table VI to neutrons per 7.85 ft^3 canister gives 1.2×10^9 neutrons/s at 3 years and 8.7×10^8 neutrons/s at 10 years. All these numbers are in good agreement with the results in Reference 6.

Conclusions

The LWR high-level waste radiation sources presented here are based on preliminary predictions of future light-water-reactor U fuel reprocessing. As such, they are subject to large uncertainties and should be treated as estimates only. Nevertheless, it is hoped that they will be of use to the fuel-cycle community in studies of waste transport and storage.

Only results from 3- and 10-year cooling time following reprocessing are presented. Because many different isotopes with widely divergent half-lives are present in the waste, it is improper to attempt to extrapolate to later times on the basis of the data presented here. Those interested in considering longer cooling times should see Figures 8 and 11 of Reference 6, which provide data out to 10^6 years after reprocessing. Gamma spectral changes should be significant over this period, with the average gamma energy decreasing markedly, whereas changes in the neutron energy spectrum should be small. However, the uncertainty in the total source strengths will increase with cooling time because, with time, the sources will depend increasingly on the precise mixture of the transuranic isotopes generated during the fuel-irradiation period and will depend decreasingly on the fission product mix. The former is known with significantly less accuracy than the latter.

REFERENCES

1. "Siting of Fuel Reprocessing Plants and Related Waste Management Facilities," Federal Register, 35, No. 222, 17330 (November 14, 1970).
2. J. O. Blomeke, C. W. Kee, and J. P. Nichols, Projections of Radioactive Wastes To Be Generated by the U. S. Nuclear Power Industry, ORNL-TN-3965, Oak Ridge National Laboratory, Oak Ridge, TN (February 1974).
3. C. W. Kee, A. G. Croff, and J. O. Blomeke, Updated Predictions of Radioactive Wastes To Be Generated by the U. S. Nuclear Power Industry, ORNL/TM-5427, Oak Ridge National Laboratory, Oak Ridge, TN (December 1976).
4. M. J. Bell, ORIGEN - The ORNL Isotope Generation and Depletion Code, ORNL - 4828, Oak Ridge National Laboratory, Oak Ridge, TN (May 1973). The ORIGEN library is distributed along with the code as a code package CCC-217 by the Radiation Shielding Information Center, Oak Ridge, TN.
5. C. M. Lederer, J. M. Hollander, and S. Perlman, Table of Isotopes (J. Wiley and Sons, New York, 1967).
6. Bernard L. Cohen, "High-Level Radioactive Waste from Light-Water Reactors," Rev Mod Phys, 49, 1 (1977).
7. C. Devillers and P. Blum, "Neutron Multiplication and Shielding Problems in PWR Spent-Fuel Shipping." Reprint of paper presented at ANS/ENS International Conference, Washington, D. C., November 14-19, 1976. Abstract published in Trans Am Nucl Soc, 55, 491 (1976).