184
5/26
c

2150
ORNL-4776

MASTER

A RADIOISOTOPE-ORIENTED VIEW OF NUCLEAR WASTE MANAGEMENT

A. F. Rupp

THIS DOCUMENT CONFIRMED AS
UNCLASSIFIED
DIVISION OF CLASSIFICATION
BY Jack H. Kahan / am
DATE 6/5/72

BLANK PAGE

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.95

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, 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.

ORNL-4776
UC-70 - Waste Disposal and Processing

Contract No. W-7405-eng-26

ISOTOPES DEVELOPMENT CENTER

A RADIOISOTOPE-ORIENTED VIEW OF NUCLEAR WASTE MANAGEMENT

A. F. Rupp
Director's Division

MAY 1972

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, 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.

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

UNCLASSIFIED
DATE 10/10/01 BY 1045

fy

CONTENTS

Abstract	1
Introduction	1
High-Level Wastes and the Radioisotope Program	1
High-Level Wastes: A Burial or an Operations Problem?	2
The Problem Nuclides	2
Processing Groups of Fission Products	4
Cesium and Strontium: Processing and Rough Cost Estimate	9
Residue: Processing and Rough Cost Estimates	11
Storage	12
Safety	13
Post-Decade Processing of Residue	13
Discussion	14
References	14

A RADIOISOTOPE-ORIENTED VIEW OF NUCLEAR WASTE MANAGEMENT

A. F. Rupp

ABSTRACT

Drawing upon a 25-year background in separating and packaging kilocurie quantities of fission products for isotopic power and radiation sources in the Oak Ridge National Laboratory Radioisotope Program, a system for the long-range management of high-level fission product waste is presented. This would involve an initial two-year storage of the liquid high-level waste to allow decay of about 70% of the radioactivity, followed by separation into chemically related groups, especially periodic group I-A (^{137}Cs) and II-A (^{90}Sr) (the principal remaining heat producers) and the remainder of the fission products plus the toxic, long-lived transplutonium isotopes. The lightly calcined, packaged residue is given an additional ten-year cooling period and is then chemically processed to remove uranium, plutonium, and transplutonium elements, which are packaged and burned in a nuclear reactor; a small final unburnable residue of toxic nuclides is canned for permanent vault storage or sent to space. Cesium-137 and strontium-90 are reprocessed after 50 to 75 years, and the undecayed portion is recycled. The low-radiation, low-toxicity bulk fission product residues are sent to low-level waste disposal. High decontamination factors are needed only on the final separations.

Vault storage under surveillance on power park or power island premises is suggested to avoid hot shipments. The cost appears to be only moderately greater than for methods for unsupervised environmental storage.

INTRODUCTION

The waste stream from the processing of irradiated uranium attracted radioisotope researchers' attention at the start of the radioisotope program because it appeared to be a copious, cheap source of a wide range of radioisotopes. Some fission products, such as ^{137}Cs and ^{90}Sr , occur in high yield and are unattainable by (n, γ) reactions. A small development program, started in 1948, culminated in the building of the Fission Product Pilot Plant at ORNL in 1955; this early work was reported at the 1955 and 1958 Geneva Conferences.^{1,2} At the Joint Committee of Congress hearings on Industrial Radioactive Waste Disposal in 1959, the advantages that might be obtained in separating certain fission products and packaging them in high-integrity containers were pointed out.³ The arguments concerning specific treatment of high-level fission product waste were reviewed and commented upon in an article entitled "Reactor By-Products."⁴ New attitudes of society toward industry make it clear that economics is no longer the controlling factor in the evaluation of processes. It therefore may be timely to again bring up some ideas that I have had in the past on the treatment of high-level reactor fuel waste. I should point out that this is a speculative paper and not intended to be viewed as an engineering study. However, in my judgment, many parts of the suggested alternative schemes are worthy of study and development.

The essence of this approach to waste management is storage of fission products under surveillance to allow decay, so that the decay products can be stored with little or no custodial care or released to the environment in a controlled way (excepting materials that are suitable for nuclear transformation⁵ or a very small amount of intractable toxic residues that require very special attention). It is suggested that the material undergoing decay in storage can be more safely and economically managed if it is initially separated into chemically related groups and finally processed to obtain low-level waste (LLW) that can be released or stored with minimum custodial care.

HIGH-LEVEL WASTES AND THE RADIOISOTOPE PROGRAM

The work done in the radioisotope program to fabricate separated fission products into large gamma-ray or heat-power sources has tended in some ways to cloud the issue as far as packaging ^{137}Cs or ^{90}Sr purely for storage purposes is concerned. Contentions have been made by opponents that one could never sell enough radioisotope sources to cover expenses and that the market would never be sufficiently large to make much of a dent in the huge output of fission products — both contentions that may be true. However, one should consider that (1) any utilization of fission products is a plus value, since the fission products should be properly packaged in any case, and (2) the

advantages to be gained by proper packaging are worth the cost. It is clear that radioisotope sales can pay for only a portion of waste-processing costs. In this article I should like to comment on the processing of fission products, plutonium and transplutonium elements, and activation products as related to radioactive waste management. Further, I should like to suggest that the spent-fuel-processing job isn't finished if only the fissile materials are returned to the reactors. The remainder of the processing would consist of

1. radioactive decay,
2. induced nuclear transformation,
3. extraterrestrial transport of intractable residues, such as ^{244}Pu and ^{247}Cm , to the sun or distant space orbit, for example.

The kinds of waste to be encountered between now and the year 2000 have been discussed in detail in publications such as "Siting of Fuel Reprocessing Plants and Waste Management Facilities" (ORNL-4451),⁶ which I have used extensively in preparing this paper. I will confine my attention to the power-reactor fuel high-level waste (HLW), including residual plutonium, transplutonium elements, and certain dissolver off-gas elements. Other wastes — intermediate-level (ILW), low-level (LLW), and solid — which in some ways present much more difficult problems, will no doubt be treated in a variety of ways.

HIGH-LEVEL WASTES: A BURIAL OR AN OPERATIONS PROBLEM?

Processes for extracting uranium and plutonium are now optimized to obtain the best recovery and purification of these elements. Many improvements have been made to produce purer gaseous and liquid process residues. These improvements include mechanical de-jacketing, selective fuel disassembly and chop-leach, controlled oxidation and degassing of fuel to control off-gas volume in removing fission product gases and tritium, and use of nonresidual salting agents. Further improvements have been suggested to avoid addition of nonvolatile chemicals, such as alkali and alkaline earth salts, iron, and manganese, to the process stream. Many of these improvements have been incorporated in the new commercial fuel processing plants. Any improvement that leads to a raffinate that is more amenable to chemical processing is a step in the right direction.

If the by-product nuclides can be put into a relatively inaccessible part of the environment in irretrievable form and essentially forgotten, suggestions for current-

operations-type storage or destruction perhaps make little sense. However, if radioactive wastes are considered to be materials that must be observed and cared for on a continuing basis, some approach such as processing and packaging is necessary. The recovery and packaging of the nuclides under discussion cannot be justified by the radionuclides' market value, even though some exists; rather, the liability incurred by bringing these nuclides into existence must be offset by entries in the cost column. Too little research and development have been done to determine whether the cost of responsibly looking after by-product nuclides is much more, a little more, or about the same as waste "disposal" schemes.

Nuclear electricity installed capacity has been estimated at 153,000 MW(e) by 1980 and 735,000 MW(e) by 2000 (see Table 1); the total amount of fission products and activation products accumulated by the end of the century staggers the imagination. The problem must be broken down into more digestible pieces. As will be pointed out later in this paper, I chose to look at a 1000-MW(e) LWR as a unit; a power park or power island would then be 10 units, and an area fuel processing plant group, say, 100 units.

THE PROBLEM NUCLIDES

The various fission products and transuranium products that occur as by-products in reactor operations are not equally bad from the standpoint of heat, radiation, toxicity, or chemical properties. By regarding the elements individually, or as groups, one can cut down the size of the problem. The most obvious groups are the volatile and nonvolatile elements which are separated in most processing schemes as a matter of course. The fission products of most concern are ^{90}Sr , ^{137}Cs , ^{85}Kr , and ^{129}I . Most of the rest decay fairly rapidly in processing and lag storage (see Table 2).

The toxic plutonium and transplutonium elements are the biggest problem — too long-lived for decay. They must be removed, concentrated, packaged, and perhaps nuclear-burned, possibly in certain power reactor core positions or in special reactors operated for the purpose; ^{242}Pu , ^{244}Pu , ^{247}Cm , and ^{248}Cm appear to be completely intractable residues (Table 3) that can possibly be transported to the sun or outer space or can be stored in the most indestructible form in maximum security surveillance vaults.

Certain other low-toxicity radioisotopes — a few fission products such as 3×10^6 -year ^{135}Cs , 2×10^5 -year ^{99}Tc , 6.5×10^4 -year ^{79}Se , 1.5×10^6 -year ^{93}Zr , and 7.5×10^6 -year ^{107}Pd — would stay with the

Table 1. Projected fuel processing wastes from total U.S. nuclear power economy
(aqueous processing of all fuels)
(from ORNL-4451)

	Calendar year ending				
	1970	1980	1990	2000	2020
Installed capacity, 10^3 MW(e) ^a	14	153	368	735	2210
Volume of waste generated, as liquid ^b					
Annually, 10^6 gal/year	0.017	0.97	2.69	4.60	13.7
Accumulated, 10^6 gal	0.017	4.40	23.8	60.1	238
Volume of waste generated, as solid ^c					
Annually, 10^3 ft ³ /year	0.17	9.73	26.9	46.0	137
Accumulated, 10^3 ft ³	0.17	44.0	238	600	2380
Accumulated radioisotopes ^d					
Total weight, metric tons	1.75	451	2440	6200	24,600
Total activity, MCi	210	18,900	84,500	209,000	666,000
Total heat-generation rate, MW	0.91	81.6	343	807	2520
⁹⁰ Sr, MCi	3.98	962	4640	9550	29,400
¹³⁷ Cs, MCi	5.27	1280	6540	15,600	57,500
¹²⁹ I, Ci	1.85	476	2700	7550	32,200
⁸⁵ Kr, MCi	0.56	124	567	1190	3900
³ H, MCi	0.033	7.29	36.2	89.5	332
²³⁸ Pu, MCi ^e	0.002	1.20	8.28	30.7	166
²³⁹ Pu, MCi ^e	0.00009	0.022	0.235	1.31	8.45
²⁴⁰ Pu, MCi ^e	0.00013	0.0409	0.395	1.91	11.4
²⁴¹ Pu, MCi ^e	0.0295	6.63	47.2	191	909
²⁴² Pu, Ci ^e	0.354	91	910	4870	30,900
²⁴¹ Am, MCi ^e	0.0089	2.31	22.7	121	763
²⁴³ Am, MCi	0.0009	0.232	1.49	5.19	27.0
²⁴⁴ Cm, MCi	0.128	29.9	137	255	700
²⁴² Cm, MCi	0.725	43.2	185	487	1490

^aData from Phase 3, Case 42, Systems Analysis Task Force (April 11, 1968).

^bAssumes that wastes are concentrated to 100 gal per 10^4 MWd(t) and that there is a delay of two years between power generation and waste generation.

^cAssumes 1 ft³ of solidified waste per 10^4 MWd(t).

^dAssumes that LWR fuel is continuously irradiated at a specific power of 30 MW/metric ton to a burnup of 33,000 MWd/metric ton, and that the fuel is processed 90 days after discharge from reactor; LMFBR core continuously irradiated to 80,000 MWd/metric ton at 148 MW/metric ton, axial blanket to 2500 MWd/metric ton at 4.6 MW/metric ton, and radial blanket to 8100 MWd/metric ton at 8.4 MW/metric ton, and that fuel is processed 30 days after discharge.

^eAssumes that 0.5% of the plutonium in the spent fuel is lost to waste.

Table 2. Waste nuclide groups

Isotopes	Half-life	Process and remarks
⁹⁰ Sr	28 years	Storage-decay (and burn?)
¹³⁷ Cs, ¹³⁵ Cs	30 years, 3×10^6 years	Storage-decay
⁸⁵ Kr	10.6 years	Storage-decay
¹²⁹ I	1.7×10^7 years	Storage (and burn?)
Most other fission products	Short (or low toxicity)	Storage-decay
Pu, transplutonium	Medium-long	Storage, burn, ETT ^a
²⁴² , ²⁴⁴ Pu and ²⁴⁷ , ²⁴⁸ Cm	Very long	Storage or ETT ^a

^aExtraterrestrial transport.

Table 3. Intractable residue

Element	Half-life (years)	σ_c (barns)	σ_f
^{242}Pu	386,900	18.5	0
^{244}Pu	8.28×10^7	1.6	0
^{247}Pu	1.64×10^7	60	0
^{248}Cm	352,000	5.2	0

bulk of the decayed residue, consisting essentially of stable isotopes. Others, such as ^{129}I , might be nuclear-burned.

PROCESSING GROUPS OF FISSION PRODUCTS

Again using the data in ORNL-4451 for the Diablo Canyon Reference LWR, I have rearranged some of the data into the Periodic Table element groups in Table 4. The weights per tonne* at 1 and 10 years and the radioactivity levels at 90 days and at 1, 10, and 100 years are given. Data on radioactivity and heat output for certain nuclides are given in Table 5.

I refer to the flowsheet (Fig. 1) entitled "Complete Fuel Processing," in which the word "complete" is used to indicate that not only the uranium and plutonium are processed to return them to reactor fuel, but also the fission products and neutron products are processed in various ways to allow safe, compact storage or, in some cases, destruction or use of a portion of the materials.

The first part of the flowsheet indicates an improved fuel process which discharges a minimum volume of gas containing iodine, xenon, krypton, and small amounts of tritium. Cladding residue containing activation products and casual fission product contamination is also discharged. (Tritium, cladding residue, intermediate-level waste, low-level waste, and solid waste are not discussed in this paper.) The balance of the fission products, uranium-plutonium residues, and transplutonium elements contaminated with only a small amount of corrosion products are transferred to water-cooled storage tanks, where the first and quite important process occurs — decay of the short-lived radioactivity. During a two-year decay period, 70% of the radioactivity (and consequent heat production) is lost. Much of the nitric acid and organic residue is decomposed by the radioactivity; water loss is adjusted as desired by temperature and off-gas control; the

vapors and gases could be returned to the nitrogen oxide-nitric acid recovery system. The cooled weak nitric acid fission product solution is now sent to chemical processing to remove groups I-A and II-A, which contain ^{137}Cs and ^{90}Sr , the principal remaining heat producers.

The bulk of the remaining fission products is group III-B, lanthanide rare earths plus a much smaller weight of actinides. The most important radionuclide heat producer after the two-year cooling period is 284-day ^{144}Ce ; 7.9-year ^{154}Eu is the controlling gamma emitter.* Other beta-gamma nuclides in this group are 33-day ^{141}Ce , 2.6-year ^{147}Pm , 93-year ^{151}Sm , and 5-year ^{155}Eu , which are either relatively short-lived (^{141}Ce , ^{155}Eu) or weak beta emitters (^{147}Pm , ^{151}Sm); all are of relatively low toxicity. The important elements of this group from a hazards standpoint are the plutonium and transplutonium elements, primarily because of their toxicity. However, in plutonium-recycle fuel situations, the actinides become heavy heat producers and will have to be separated at least partially to join the ^{137}Cs and ^{90}Sr in high-heat storage.

As indicated in Fig. 1, following the two-year cooling period and the removal of groups I-A and II-A, the balance is dried, lightly calcined (so that future redissolution is not too difficult), and packaged in relatively large sealed containers for carefully controlled storage for a second decay period of about ten years. In plutonium-recycle fuel, most of the actinides will also have to be removed, as previously noted.

Storage space may become a problem after a number of years, so the various tubes could be sent to a processing plant to keep space available as the economics at the time dictate. Instead of reprocessing, one could also ship the decayed high-bulk, low-heat, high-toxicity tubes to a long-range repository. The cesium-strontium tubes could be reprocessed after 50 to 100 years to remove the inert barium and zirconium decay products and to return the remnant ^{137}Cs and ^{90}Sr to the process stream.

After the ten-year decay of the residue, the bulk of which is group III-B, there are a few other radionuclides present, none of which appear to pose much of a problem from either the standpoint of radioactivity or toxicity. Groups IV-B, VI-B, VII-B, and VIII include

*Values for ^{154}Eu from Table 4 may be high by a factor of 4, based upon available comparable fission yields of ^{154}Eu vs ^{137}Cs , for example. Also, as noted in Table 5, shorter half-life would lower values in decayed residue.

*Metric ton.

Table 4. Data on isotopes and radioactivity in 33,000 MW Diablo Canyon reference LWR fuel
(from ORNL-4451)

Isotope	Half-life or natural abundance	Grams/tonne at –		Curies/tonne at –			
		1 year	10 years	90 days	1 year	10 years	100 years
Group I-A							
⁸⁵ Rb	72.8%	95	108				
⁸⁷ Rb	5 × 10 ¹⁰ years	239	239				
¹³³ Cs	100%	1010	1010				
¹³⁴ Cs	2 years	134	6	220,000	175,000	8,330	Nil
¹³⁵ Cs	2 × 10 ⁶ years	326	326				
¹³⁷ Cs	30 years	1210	980	110,000	110,000	85,000	11,000
Group II-A							
⁸⁸ Sr	82.56%	351	251				
⁹⁰ Sr	28 years	534	427	76,900	75,500	60,400	6,560
¹³⁴ Ba	2.42%	123	251				
¹³⁶ Ba	7.81%	26	26				
¹³⁷ Ba	11.32%	71	298				
¹³⁸ Ba	71.66%	1210	1210				
Group III-B							
⁸⁹ Y	100%		465				
¹³⁹ La	99.91%		1270				
¹⁴⁰ Ce	88.48%		1460				
¹⁴¹ Ce	33 days		0	205,000	570	0	0
¹⁴² Ce	11.07%		1180				
¹⁴⁴ Ce	290 days		0	892,000	456,000	150	0
¹⁴¹ Pr	100%		1200				
¹⁴² Nd	27.13%		21				
¹⁴³ Nd	12.20%		808				
¹⁴⁴ Nd	23.80%		1340				
¹⁴⁵ Nd	8.30%		706				
¹⁴⁶ Nd	17.18%		711				
¹⁴⁸ Nd	5.72%		378				
¹⁵⁰ Nd	5.60%		183				
¹⁴⁷ Pm	2.6 years		8	104,000	85,100	78	0
¹⁴⁷ Sm	15.07%		163				
¹⁴⁸ Sm	11.27%		251				
¹⁴⁹ Sm	13.84%		6				
¹⁵⁰ Sm	7.47%		315				
¹⁵¹ Sm	73 years		39	1,150	1,150	1,070	521
¹⁵² Sm	26.63%		92				
¹⁵⁴ Sm	22.53%		37				
¹⁵³ Eu	52.23%		130				
¹⁵⁴ Eu	16 years ^a		31	6,870	6,650	4,500	91
¹⁵⁵ Eu	1.8 years		0.1	6,790	5,090	162	0
¹⁵⁴ Gd	2.15%		18				
¹⁵⁶ Gd	20.59%		84				
¹⁵⁸ Gd	24.78%		13				
¹⁵⁹ Tb	100%		1.7				

Table 4 (continued)

Isotope	Half-life or natural abundance	Grams/tonne at		Curies/tonne at			
		1 year	10 years	90 days	1 year	10 years	100 years
Group III-B, actinides							
²³⁹ Pu	24,400 years	5380		330	330		
²⁴⁰ Pu	6600 years	2170		478	478		
²⁴¹ Pu	13.2 years	975		116,000	111,000		
²⁴² Pu	380,000 years	349		1.4	1.4		
²⁴³ Pu	5 hr						
²⁴⁴ Pu	7 × 10 ⁷ years						
²⁴⁵ Pu	11 hr						
²⁴⁶ Pu	11 days						
²⁴¹ Am	462 years	301	0.01	172	301		
^{242m} Am	152 years			4	4		
²⁴³ Am	7370 years	90	0.056	17	17		
²⁴⁴ Am	10 hr	0					
²⁴² Cm	163 days	1.8		19,300	6,000		
²⁴³ Cm	35 years	0.1		4	4		
²⁴⁴ Cm	18.1 years	30		2,500	2,430		
Group IV-B							
⁹⁰ Zr	51.46%		143				
⁹¹ Zr	11.23%		609				
⁹² Zr	17.11%		663				
⁹³ Zr	1.5 × 10 ⁶ years		735	1.9	1.9	1.9	1.9
⁹⁴ Zr	17.4%		789				
⁹⁵ Zr	95 days	0		524,000	27,000	0	0
⁹⁶ Zr	2.8%		830				
Group V-B							
⁹⁵ Nb	35 days		0				
Group VI-B							
⁹⁵ Mo	15.7%		770				
⁹⁶ Mo	16.5%		39				
⁹⁷ Mo	9.45%		838				
⁹⁸ Mo	23.75%		849				
¹⁰⁰ Mo	9.62%		971				
Group VII-B							
⁹⁹ Te	2 × 10 ⁵ years		835	14	14	14	14
Group VIII							
¹⁰⁰ Ru	12.70%		56				
¹⁰¹ Ru	16.98%		776				
¹⁰² Ru	31.34%		768				
¹⁰³ Ru	39.8 days		0	255,000	2,070	0	0
¹⁰⁴ Ru	18.27%		538				
¹⁰⁶ Ru	1 year		0.2	459,000	273,000	550	0
¹⁰³ Rh	100%		392				
¹⁰⁴ Pd	9.3%		246				
¹⁰⁵ Pd	22.6%		294				
¹⁰⁶ Pd	27.2%		448				
¹⁰⁷ Pd	7 × 10 ⁶ years		236				
¹⁰⁸ Pd	26.8%		156				
¹¹⁰ Pd	13.5%		34				
Group I-B							
¹⁰⁹ Ag	48.65%		60				

Table 4 (continued)

Isotope	Half-life or natural abundance	Grams/tonne at –		Curies/tonne at			
		1 year	10 years	90 days	1 year	10 years	100 years
Group II-B							
¹¹⁰ Cd	12.39%		41				
¹¹¹ Cd	12.75%		17				
¹¹² Cd	24.07%		9				
¹¹⁴ Cd	28.86%		1.2				
¹¹⁶ Cd	7.58%		4				
Group III-A							
¹¹⁵ In	95.77%		1.2				
Group IV-A							
¹¹⁶ Sn	14.07%		2.7				
¹¹⁷ Sn	7.54%		3.9				
¹¹⁸ Sn	23.98%		4.0				
¹¹⁹ Sn	8.62%		4.2				
¹²⁰ Sn	33.03%		4.4				
¹²² Sn	4.78%		5.1				
¹²⁴ Sn	6.11%		7.6				
Group V-A							
¹²¹ Sb	57.25%		4.5				
¹²³ Sb	42.75%		5.5				
¹²⁵ Sb	2.7 years		0	8,500	6,990	693	0
Group VI-A							
¹²⁵ Te	7.01%		11				
¹²⁶ Te	18.72%		20				
¹²⁸ Te	31.72%		134				
¹³⁰ Te	34.46%		426				
⁷⁸ Se	23.52%		2.6				
⁷⁹ Se	6.5 × 10 ⁴ years		5.6				
⁸⁰ Se	49.82%		10.3				
⁸² Se	9.19%		32.5				
Group VII-A							
¹²⁷ I	100%		40				
¹²⁹ I	1.7 × 10 ⁷ years		233		0.04	0.04	0.04
⁸¹ Br	49.5%		15				
Inert gases							
⁸³ Kr	11.48%		41				
⁸⁴ Kr	57.02%		111				
⁸⁵ Kr	10.6 years		16	11,300	10,800	6,050	18
⁸⁶ Kr	17.43%		192				
¹³⁰ Xe	4.07%		11				
¹³¹ Xe	21.13%		409				
¹³² Xe	26.96%		1150				
¹³⁴ Xe	10.54%		1530				
¹³⁶ Xe	8.95%		2320				

^aThe $T_{1/2}$ on ¹⁵⁴Eu is now reported as 7–10 years.

Note: For some of the nuclides in this table from ORNL-4451 there are newer, and presumably better, half-life values:

⁹⁰ Sr	28.5 years	¹²⁹ I	1.57 × 10 ⁷ years
¹⁴⁴ Ce	284 days	⁸⁵ Kr	10.7 years
¹⁵⁴ Eu	7–10 years	²⁴¹ Am	433 years
¹⁵⁵ Eu	4.96 years	²⁴¹ Pu	14.8 years

Table 5. Activity and heat output from certain nuclides
(per tonne of fuel)

Isotope	Half-life	At one year		At two years		At ten years	
		Ci	kW	Ci	kW	Ci	kW
^{134}Cs	2.05 years	175,000	1.83	125,000	1.31	8,330	0.09
^{137}Cs	30.0 years	105,000	0.56	103,000	0.54	85,300	0.45
^{90}Sr	28.0 years	75,000	0.53	73,700	0.53	60,400	0.42
^{144}Ce	284 days	456,000	3.79	187,000	1.55	150	
^{147}Pm	2.62 years	85,100	0.04	65,300	0.03	7,870	
^{154}Eu	16.0 years ^a	6,650	0.06	6,370	0.06	4,500	0.04
^{154}Eu	7.9 years ^a	6,430	0.06	5,890	0.06	2,920	0.03
^{95}Zr	65 days	27,900	0.43				
^{103}Ru	39.8 days	2,070	0.01				
^{106}Ru	1 year	273,000	2.66	136,500	1.33	550	0.01
^{125}Sb	2.7 years	6,990	0.02	5,400	0.02	693	
^{85}Kr	10.8 years	10,800	0.02	10,100	0.02	6,050	0.01

^aEurorium-154 half-life is in doubt; said to be 7 to 10 years.

Fig. 1. Complete fuel processing.

most of the rest of the weight of the residue containing the radionuclides 1.5×10^6 -year ^{93}Zr , 2×10^5 -year ^{99}Tc , 1-year ^{106}Ru , and 7.5×10^6 -year ^{107}Pd ; of these only ^{106}Ru causes a radiation problem. Radioisotopes in groups III-A to VI-A are 2.7-year ^{125}Sb and 6.5×10^4 -year ^{79}Se , which are minor radiation or toxicity problems.

Removal of the toxic Pu, Np, Am, and Cm from the redissolved residue in a semibatch process after the total decay period of about 12 years is a different kind of problem than trying to remove these elements with high decontamination factors from the blazing hot, continuously processed original dissolver solution. It becomes more of a problem in chemistry than one in high-level radiation process engineering. There are a number of ways of going about removing these elements, but in this conceptual paper, it suffices to say that I believe it can be done without too much cost.

Group VII-A is important in that it includes iodine. However, all the short-lived iodines decay away quickly, leaving only 1.7×10^7 -year ^{129}I , diluted with stable ^{127}I . This can be canned and permanently stored or nuclear-burned to 12.3-hr ^{130}I and 25-min ^{128}I and thence allowed to decay to stable xenon.

The last group, inert gases, contains only 10.6-year ^{85}Kr as a significant radionuclide. As indicated, stable xenon is fractionated from krypton, purified, and sold commercially. The ^{85}Kr is stored directly in pressure cylinders, or possibly enriched so as to require only one-tenth as many storage cylinders. However, the enrichment process would probably require permission to release a small amount of ^{85}Kr to the atmosphere in the depleted krypton gas stream, and the economic feasibility is questionable. The cost of krypton removal, concentration, and packaging for storage is not included in my cost summary (Table 6), as I do not have good data on it at the present time; commercial organizations

are preparing to offer krypton packaging in the near future. Hopefully, xenon sales may eventually recover most of the cost.

CESIUM AND STRONTIUM: PROCESSING AND ROUGH COST ESTIMATE

Hanford is already processing waste to some extent in a way similar to that shown in Fig. 1. They are separating and packaging ^{137}Cs and ^{90}Sr and drying out the sludge in big underground tanks, which correspond more or less to the lightly calcined residue capsules except that they are huge and not as amenable to transportation, handling, inspection, and reprocessing of their contents after 10 to 15 years' decay.

There is some practical knowledge of the probable costs of at least parts of the Fig. 1 process gained from years of fission product processing in the ORNL Fission Products Development Laboratory and Hanford's more recent fission product separations.

Using the 1000-MW(e) Diablo Canyon reactor as the unit, I chose to look at a miniplant integrated with the reactor site to process the fuel that comes out of it and take care of the fission products. This would, of course, appear to be a very uneconomic procedure, although it is roughly analogous to a homogeneous-reactor-integrated fuel-processing situation. Diablo Canyon will discharge each year about 30 tonnes (one-third of the reactor charge) of spent fuel. The fuel will cool about 90 days and normally could be shipped to a processing plant; however, in my hypothetical case we run it through the associated miniplant, remove and decontaminate uranium, plutonium, and neptunium only to the economic optimum, and ship it to a big finishing plant instead. Setting aside the interesting possibilities of the reactor or power park on-site recovery of partially decontaminated uranium and plutonium, let's speculate about how much of a job it would be to take care of the fission products. The 1AW waste stream from the 30 tonnes would be contained in about 1000 gal of solution. After two years' decay, the tank contents would be ready for processing while another tank is being filled for the cooling period. Therefore, we can assume that we have the entire year to process the fission products from the 30 tonnes. Group I-A contains about 90 kg of cesium and rubidium, of which only about 1 kg is rubidium (see Table 4). At 1-year decay, there still is more 2-year ^{134}Cs (5.2×10^6 Ci) than 30-year ^{137}Cs (3.3×10^6 Ci); radioactivity of 2×10^6 -year ^{135}Cs is negligible.

Group II-A contains about 43 kg of barium and about 27 kg of strontium, the only significant radioactivity being 28-year ^{90}Sr (2.3×10^6 Ci).

Table 6. Rough cost summary

Item	Cost (mills/kWhr)
Two-year cool, acid waste	0.0033 ^a
Packaged ^{137}Cs	0.0024
Packaged ^{90}Sr	0.0018
Packaged curium	^b
Packaged residue	0.0060
Vault, capital carrying charges	0.017
Vault, operations	0.0070
Total	0.0375

^aORNL-3128.

^bNo data available, but probably about 0.002.

In Hanford's waste-processing scheme (Fig. 2), cesium and strontium are removed in rather good purity, to some extent with possible end uses for heat and radiation sources in mind and also to save storage space. However, I have a simpler approach in mind than the Hanford-ORNL Fission Products Development Laboratory process now being used to produce rather pure ^{137}Cs and ^{90}Sr . Since there appears to be no need for high chemical purity for the group I-A (Cs), group II-A (Sr), and residue for storage, an adaptation of the original ammonia process should be good enough; a highly simplified flowsheet is shown in Fig. 3. The early actinide removal step that may be required for plutonium-recycle fuels is not included. Judging from rather informal Hanford estimates, purified $^{137}\text{CsCl}$ should cost about \$0.50 per curie and $^{90}\text{SrCO}_3$, about \$0.03 per curie. One could calculate a cost for the important cesium-strontium removal of about \$234,000 for the hypothetical on-site unit we are studying, equivalent to \$7900 per tonne and about 0.028 mill/kWhr. This is obviously more than one would want to pay for this operation, which is only one part

of complete fuel processing. Viewed as either a power park incremental operation or as part of a processing plant operation, it is difficult to see how the cost could be that high. A small processing unit utilizing three men on single shifts could hardly use up a year to process 150 kg of crude cesium and strontium, especially if source-grade material need not be produced. Further, while cesium-strontium processing has limiting factors in scale-up, certainly one should be able to drastically reduce costs by using well-engineered process equipment on a routine basis.

The containers for ^{90}SrO and $^{137}\text{CsCl}$ would be made from 10-ft-long, nominal-diameter $3\frac{1}{2}$ -in. (actual 3.55 in.) standard stainless steel pipe with welded ends. The outside protecting capsule would be made from double extra strong welded steel pipe (5.56 in. OD, 4.06 in. ID, 0.75 in. wall). Allowing for the impurities in $^{137}\text{CsCl}$ (Rb, K, and Na) and in ^{90}SrO (Ba and Ca), the capsules would hold about 2 MCi of ^{90}Sr or 1.5 MCi of ^{137}Cs . The densities allowed for are obtained by tamping or step-pressing the dried powder into the tubes. The cost of the tubes plus protective

Fig. 2. Hanford HLW management program.

Fig. 3. Ammonia FP process.

capsules is about \$1000 each. Tube loading, testing, and handling would cost another \$1000. Referring to our 30-tonne per year Diablo Canyon example, one tube would be required for ^{90}Sr (2.3 MCi) and two for ^{137}Cs per year; estimated cost to the storage vault is shown in Table 7.

RESIDUE: PROCESSING AND ROUGH COST ESTIMATES

The residue, which is primarily fission products minus groups I-A and II-A, is subjected to drying and light calcination, except in the cases where plutonium-recycled fuel has caused an excessive buildup of longer-lived heat producers such as ^{241}Am , ^{243}Am , and ^{244}Cm (most of the plutonium and neptunium will have been removed in the main extraction process). Fortunately, only a fair separation of americium-curium actinide rare earths from the large quantity of lanthanide rare earths is required. There are several ways in which a crude separation such as this can be done; however, it is an area where research and development are needed.

The quantities per year in our 1000-MW(e) example are shown in Table 8. The primary processing work for the residue consists in drying of the main stream and

incorporation of returned precipitates from the group I-A scavenging of molybdenum, ruthenium, and minor elements. Again, only fair separations are required. The solids are calcined sufficiently to prevent gas buildup in storage from radiation decomposition of H_2O , NH_3 , CO_3^{2-} , OH^- , and NO_3^- ; hopefully the calcine would be light enough to allow easy dissolution for processing after ten years' storage. The 25 kW of heat production would cause little trouble. The total quantity of residue elements is about 375 kg, or about 450 kg of oxides; at a tamped density of about 3 kg/liter (50%), this storage would require 148 liters of volume (5.2 ft^3). Three 6-in.-diam stainless steel capsules 10 ft long would be adequate. Extra heavy steel pipe would be used for the protective capsule. Estimated cost for the capsules would be about \$1500; packaging operations should cost no more than for the cesium-strontium tubes, about \$1000.

The cost of the residue processing operations can be only very roughly estimated at this point, as we do not have direct experience. However, experience with similar operations indicates that one should be able to perform these operations for less than \$50,000 in a processing unit with a relatively small capital equipment investment. This is about \$1700 per tonne, or 0.006 mill/kWh(e) upon delivery of the storage capsules to the vault.

Table 7. Estimated cost to storage vault

	Hanford's process cost		Group processing ^a	
	¹³⁷ Cs	⁹⁰ Sr	¹³⁷ Cs	⁹⁰ Sr
Cost (dollars)				
Processing	165,000	69,000	16,500	13,500
Capsules	2,000	1,000	2,000	1,000
Packaging	2,000	1,000	2,000	1,000
Total	169,000	71,000	20,500	15,500
Cost per tonne (dollars)	5,630	2,360	684	515
Cost per kWh(e) (mills)	0.02	0.008	0.0024	0.0018

^aUnrefined cesium and strontium.

Table 8. Weight of residue residues

Element group	Quantity (kg)	Heat output (kW)
Rare-earth lanthanides	330	16
Americium and curium	6.4	4
Zirconium	11	Nil
Molybdenum	10	Nil
Technetium	2.4	Nil
Ruthenium and palladium	12	5
Ag, Cd, Sn, Sb, Te, and Se	2.4	Nil
Total	374 approx	25 approx

STORAGE

The storage vault for the ¹³⁷Cs, ⁹⁰Sr, and residue tubes would need about 150 spaces for cesium, about 75 for strontium, and about 30 for residue, each space occupying approximately 1 ft². The depth required would be enough to accommodate tubes a maximum of 10 ft long, plus plenum chambers for incoming and exiting cooling air, plus top deck shielding, which would total about 40 ft. The vault is designed to accommodate one hundred and fifty 1.5-MCi tubes of ¹³⁷Cs, the equilibrium amount of ¹³⁷Cs, at which point the radioactivity is decaying as fast as it is being produced in the 1000-MW(e) reactor being used as the unit in this study; similarly, seventy-five 2.3 MCi tubes of ⁹⁰Sr would be adequate. This point would be reached in about 75 years, which might be an appropriate time to start reprocessing the cesium and strontium so that the vault space could be reused for new fission products (possibly coming from a new reactor). Approximately 30 residue containers would be accumulated each decade, at the end of which time reprocessing could be started if so desired. The accumulated radioactivity load would be 143 MCi of ¹³⁷Cs, 100 MCi of ⁹⁰Sr, and 1 MCi of miscellaneous residue

radioisotopes at the 75-year ¹³⁷Cs equilibrium time, with no corrections made for decay. The residue tubes accumulate for only ten years, however. The maximum accumulated heat load, uncorrected for decay during the loading period, would be about 3.5 MW. It is seen that a fairly small vault (about 225 ft² in this case) could easily outlast the reactor it is servicing.

The overall vault structure area would probably not be more than about 500 ft², including the service hot cell. For a power park with ten reactors, a vault building 50 by 100 ft should suffice; the maximum radioactivity load would be about 2450 MCi and the heat load about 35 MW.

The storage vault would be steel-lined to protect the concrete. The compartments for the various tubes would be made of heavy steel plate, designed so that a group of tubes could be constantly monitored to detect any signs of radioactivity leakage. In case of a leak, the tubes in the group could be taken out to a cell for inspection, repair, or recanning.

A 250-ft² structure 40 ft deep would have a gross cubage of 10,000 ft³; at \$100 per cubic foot, a rough estimated cost of \$1 million is derived. The capital carrying costs at 15% per year would be \$150,000, or 0.017 mill/kWhr.

Operating costs for a storage vault would depend greatly upon whether it was an incremental operation associated with a reactor station, power park, or waste processing plant. However, for the unit example [1000-MW(e) reactor], one man-year of operation and one man-year of maintenance should be more than adequate, inasmuch as the resources of the nuclear site would be available as needed. Monitoring could be handled through the central instrument panel. An overall estimate of \$50,000 per year assigned to these functions would result in a cost of about 0.007 mill/kWhr. These estimates for a small individual unit servicing only one reactor are probably quite high, since

the capital, and particularly the operational, costs would be greatly reduced if a vault installation serving a 10-reactor power park or a fuel processing plant serving a 100-reactor group were considered. However, I would prefer not to see too large a concentration of vaults — certainly not one serving the entire United States, for example — regardless of the economies that might be gained.

SAFETY

When vault storage has been evaluated by others, it has been rejected as (1) unsafe, and (2) too costly. In my opinion, both of these arguments are open to question and further study; they considered only the underground vaults that were sealed, that contained gross fission products in sealed direct-evaporation pots, and that depended upon rejection of heat through the earth. Such storage is simply burial of a group of sealed fission product pots in a special king-sized concrete container. This bears little resemblance to my proposal shown in Fig. 1.

The main point of my contention is that the fission products need to be looked after, much as we do for the radioisotopes inside the nuclear reactor, which are decaying and burning away from the instant they are created. If we can care for the equilibrium fission products, intimately mixed in fissile material with gigawatts of potential power, it seems reasonable that we can safely handle kilowatt amounts of fission products in decay tanks and capsule storage vaults.

The vault would be doubly contained and would be associated with a reinforced concrete chimney for the cooling air and an emergency radiative heat rejector. Banks of low-pressure-drop filters would be used to remove possible particulate matter from input and exit cooling air. Complete monitoring, checking, and repair services would ensure that the capsules remained in good condition. Warm, dry conditions would limit corrosion to a negligible minimum. Rather detailed knowledge on the possible corrosion of stainless steel by fission product oxides and other compound forms is available in the Isotopes Program, some covering decades of storage.

It is doubtful whether natural disasters, such as hurricanes or earthquakes, would fatally damage a properly designed vault. Loss of air cooling would be a matter of concern, but the emergency radiative heat rejection would ensure that the capsules would not melt down and release their contents. Obviously, a detailed safety analysis would be required; I am confident that it could pass a rational, competent review.

POST-DECADE PROCESSING OF RESIDUE

If we accumulate three tubes of residue (about 375 kg of elements, Table 8) per year from our 1000-MW(e) reactor, we will build up about 30 tubes during a decade, or 300 tubes containing 37,500 kg for a 10-reactor power park. We can either process them and make room for new tubes or build new vault space.

What would be the incentives to process the residue? There might be at least two: (1) the processing costs may be less than the capital and operating costs of increased storage; (2) it may be desirable to avoid further accumulation of transuranium isotopes. Utilizing our figures for vault capital and operating costs, it appears that we should have to keep the processing costs (separations, packaging, decontaminated residue disposal, nuclide destruction, special canning, restorage, or extraterrestrial disposal of transplutonium elements) below about 0.024 mill/kWhr. Without the necessary research and development, good cost figures are not available; past experience with similar problems suggests that we could come reasonably close to this figure. However, reason 2 is a more cogent one: keeping down the total inventory volume of toxic nuclear materials.

Redisolution of the calcined, aged residue could be difficult, especially if self-sintering occurred during storage. Ordinary group chemical separations, for example, oxalate precipitation of the rare earths, would be used as preliminary steps. Referring again to Table 8, we see that the primary problem is to separate 6 kg of americium and curium (plus residual amounts of uranium and plutonium, not shown) from 330 kg of lanthanide rare earths. The ten-year-decayed rare-earth radioactivities are shown in group Iii-B of Table 4 in curies per tonne; on the 30-tonne/year basis (375 kg total residue) the radioactivity is:

Isotope	Curies	Kilograms of total element
^{144}Ce	4,500	79.0
^{147}Pm	2,340	0.2
^{151}Sm	32,100	27.0
^{153}Eu	135,000	4.8
^{154}Eu		
^{155}Eu	4,860	

The balance of the lanthanide group is composed of stable La, Pr, Nd, and Y. The nitric acid solution is differentially extracted, first with about 30% tributyl phosphate to remove uranium, plutonium, and neptunium at appropriate nitric acid concentration; then extraction of americium and curium is done with 100% tributyl phosphate at high nitric acid concentration.

Process parameters are adjusted to favor the separation of the transuranium elements at the expense of having part of the lanthanides go with them – the objective being to decontaminate the lanthanides as much as possible. Cerium and europium may be removed by oxidation-reduction precipitation or mercury electrode processes for further decay. The lanthanides may then be further decontaminated by high-pressure ion exchange. The low-radioactivity, low-toxicity bulk residue is now intermediate-level waste, or possibly low-level waste, and is handled according to systems that may be developed for that class of waste.

The uranium-plutonium-neptunium cut is sent back to the fuel reprocessing unit or calcined to oxide (or other fuel form), canned, and put into a reactor for nuclear burning. The quantity should be quite small (about 0.5% X 30 tonnes, about 150 g/year).

DISCUSSION

How would future generations look at all this? Quite possibly they would applaud having the fission products and transuranium elements disposed of, out of sight and mind, deep in salt mines, granite caverns, or Plowshare excavations, but perhaps not, especially if some unforeseen geological happening allowed the waste to gain access to the biosphere. Better ways of coping with fission products and transuranium waste may have been found by that time; readily retrievable, conveniently packaged material from the storage vaults could then be directed into new processes for improved storage or disposal, or for use of contained values.

The radioisotope-oriented waste management described in this paper is fully compatible with most other methods. At various points, for example, one could interrupt the process flow and consign the extremely rugged cesium, strontium, curium, or residue capsules to a salt mine, if that is eventually fully approved.

Viewed another way, the process described above could be considered an interim storage stage that goes on indefinitely. Thus, time can be taken to examine every possible "ultimate" disposal scheme that arises during the ensuing decades, and the stored fission products would be readily available in good chemical condition to direct into new processes.

Without a doubt, the greatest hue and cry would come from critics who oppose the collection of radioactivity at various power park sites, even though in maximum security form and under strict surveillance.

The approach suggested in this paper centers around the old axiom: divide and conquer. In summary: we avoid shipping hot fuel around the country; radioactive

species are separated according to heat generation, toxicity, etc. (not necessarily the "clean separation" said by some to be required), so that proper individual attention can be given; long-lived toxic nuclides are destroyed or sent into space if found feasible; limited amounts of material are allowed per storage vault, after the manner of explosive-storage magazines, to limit the effects of accidents; and monitoring and general surveillance permit detection of possible leaks for early repair to stop accidents before they can happen.

Finally, I should mention the possible value of the by-products, as I brought out in refs. 3 and 4 and C. A. Rohrmann covered in more detail in refs. 7 and 8. In all three categories of radioisotope use – (1) electromagnetic radiation, (2) heat and power, (3) stable (decayed) nuclides – there are good possibilities for eventual utilization in fairly large quantities. Beneficial use of the by-products should be helpful in defraying a portion of the expense of waste disposal and enhance the image of fission nuclear energy's unavoidable companion, radioisotopes.

REFERENCES

1. A. F. Rupp, "Large Scale Production of Radioisotopes," *Proc. Int. Conf. Peaceful Uses At. Energy*, 14, 68 (1956).
2. E. Lamb et al., "Fission Product Pilot Plant and Other Developments in the Radioisotope Program at the Oak Ridge National Laboratory," *Proc. Int. Conf. Peaceful Uses At. Energy*, 20, 38–44 (1953).
3. *Hearings Before the Special Subcommittee of Joint Committee on Atomic Energy*, 86th Congress, 3, 2341, U.S. Government Printing Office, Washington, D.C. (1959).
4. USAEC Division of Technical Information, *Reactor Technology Selected Reviews – 1964*, pp. 477–528, Clearinghouse for Federal Scientific and Technical Publications, Springfield, Va. (1964).
5. H. C. Claiborne, "High-Level Radioactive Waste Disposal by Transmutation," *Trans. Am. Nuclear Soc.* (to be published).
6. Oak Ridge National Laboratory, *Siting of Fuel Reprocessing Plants and Waste Management Facilities*, ORNL-4451 (1970).
7. C. A. Rohrmann, "Values in Spent Fuel From Power Reactors," *Isotop. Radiat. Technol.* 6(1), 19–41 (1968).
8. C. A. Rohrmann, "Fission Product Xenon and Krypton – An Opportunity for Large-Scale Utilization," *Isotop. Radiat. Technol.* 8(3), 253–60 (1971).