BYPRODUCT INVENTORIES

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ABSTRACT

An inventory of potentially available nuclear byproducts from defense production activities and from the commercial fuel cycle has been prepared for inclusion in the DOE document, Department of Energy Plan for Recovery and Utilization of Nuclear Byproducts from Defense Wastes, which should be issued in 1983. The byproduct materials included in the inventory are strontium-90, cesium-137, krypton-85, xenon, palladium, rhodium, ruthenium, technetium-99, americium-241, promethium-147, neptunium-237, and curium. The inventory summarizes the amounts of these materials in current wastes and projects the quantities contained in future wastes.

The nuclear byproduct materials of interest include strontium-90, cesium-137, krypton-85, xenon, palladium, rhodium, ruthenium, technetium-99, promethium-147, neptunium-237, americium-241, and curium. These materials are potentially available from currently stored waste and from future processing at Hanford, at the Savannah River Plant (SRP), at the Idaho Chemical Processing Plant (ICPP), and from commercial spent fuel. In addition, technetium-99 is recovered during uranium isotope enrichment and americium-241 is recovered during reprocessing of previously purified plutonium.

Commercial spent fuels represent a major source of byproducts if reprocessing of commercial fuels is restarted. The current inventory of commercial spent fuel, through 1981, is about 8000 metric tons of initial heavy metal (MTIHM).

Although a byproduct may be potentially recoverable from a given source, it does not necessarily follow that recovery is either technically or economically feasible. At present, facilities for recovering, purifying, and packaging of byproducts are limited. Cesium-137 and strontium-90 are recovered, packaged, and stored in water basins at Hanford for waste management purposes. Past emphasis has centered on safe, cost-effective management of cesium and strontium waste, not on the production of optimized heat or radiation sources. The activity levels are less than desirable for most of the capsules and less than minimum useful values for many of the stored waste capsules. In future processing, changing the emphasis to byproduct packaging, and the imposition of appropriate controls will ensure high-quality sources. Cesium-137 recovery, but not packaging, is currently planned at at SRP for waste management purposes. The recovered cesium-137 may be added to the sludge and vitrified, separately vitrified, or converted to a suitable product form and packaged for byproduct use. Neptunium-237 is recovered at SRP and irradiated to plutonium-238. Krypton and xenon are recovered at ICPP with final purification at ORNL.

The quality of the material that can be obtained from a specific source can affect potential recovery for beneficial uses. For example, the cesium-137 content of cesium recovered from spent fuel or stored wastes will depend on the time since the cesium (fuel) was discharged from the reactor. Thus, fuel or wastes stored for long periods may not be a suitable source of high-quality cesium-137. On the other hand, cesium recently discharged from the reactor contains a high concentration of cesium-134, which may adversely affect the quality of the cesium-137 for specific applications. The quality of strontium-90 recovered from fuel or wastes also depends on the time since the strontium was discharged from the reactor. The quality of the strontium-90 obtainable can also be degraded by uncontrolled additions of natural strontium during processing of the spent fuel and waste treatment.

Almost all the byproducts could be recovered from both existing and future wastes at the Idaho Chemical Processing Plant. However, except for krypton and xenon, there are no facilities at ICPP for such recovery. Since the amounts available at ICPP are only about 10 percent of the amounts available at SRP or Hanford, any new recovery capacity should be installed at SRP or Hanford. Accordingly, isotope inventory data from ICPP are not included in the following sections.

Overall, several factors can affect the potential recovery of nuclear byproducts for beneficial use, and all the factors must be considered in evaluating the materials available from the various sources. The reader is cautioned that the inventory data provided are the best currently available, but that they are valid only for the stated assumptions. Refinements in computer calculations, additional sampling and analysis, changes in fuel processing schedules, or changes in byproduct purity requirements will change the inventory.

One point must be explained regarding energy data presented for various radioisotopes. The energy values given are for the entire decay chain. For example, the power values shown for strontium-90 in watts (t) are the combined values for the decay of the strontium-90 and the yttrium-90 daughter.

Strontium-90

Table I gives the estimated inventories of strontium-90 for various sources. Only part of the strontium-90 identified can be recovered and only part of that which can be recovered has any potential for beneficial use. Eighteen percent of the encpapsulated strontium at Hanford (30 of 257 capsules) has a strontium-90 isotopic content higher than 39 percent.[1] Preliminary waste tank analyses at SRP indicate that the strontium has a strontium-90 content of only 12 percent.

Table I. Estimated Inventories of Strontium-90 from Various Sources[2-8]

Source	Effective Inventory Date	Effective Ci
Hanford		
- SrF_2 capsules through 7-82	1/83	1.9×10^7
- SrF_2 capsules 7/82 through 86	1/83	3.4×10^7
- Stored wastes[(a)]	1/83	7.0×10^7
- Future wastes	1/91	3.4×10^7
Savannah River Plant		
- Current wastes[(b)]	1/83	1.2×10^8
- Future wastes (through 2000)	1/2001	9.1×10^7
Commercial Spent Fuel		
Accum. through 1981	1/82	3.7×10^8
Accum. through 2020	1/2021	7.9×10^9

(a) Strontium-90 contained in current stored wastes but not considered to be recoverable.
(b) Ninety percent of strontium-90 (1.2×10^8 Ci) contained in current wastes is in 12 storage tanks. The remaining 10% of the strontium is not included in inventory.

The estimated available high-quality strontium-90 ($^{90}Sr/Sr \geq 40\%$) through the year 2000, with applicable assumptions, is shown in Fig. 1.

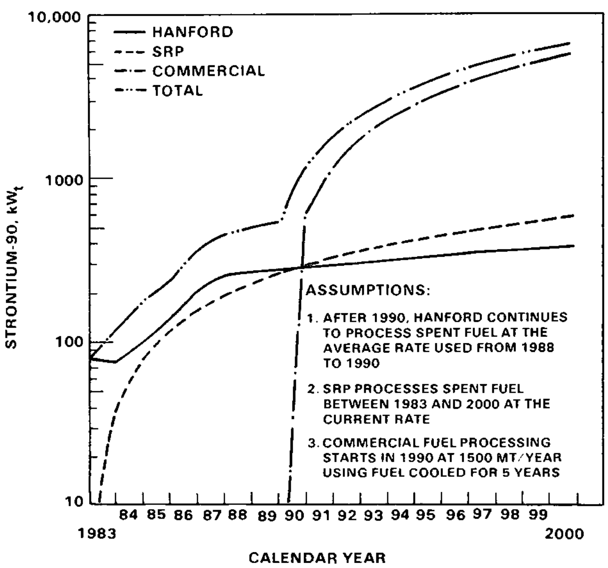

Fig. 1. Cumulative Inventory of Heat Source Grade Strontium-90 ($^{90}Sr/Sr > 40\%$) Available for Recovery from High-Level Wastes

The data show that 80 kW_t of high-quality strontium-90 is available now at Hanford. This is enough material to fuel several power-generation demonstration units, depending upon their size, between 1983 and 1988. Successful performance of these units may then justify construction of the needed facilities at SRP to make significantly more material available. Immediate steps should be taken at SRP to ensure that the strontium-90 in future waste will not be degraded by inert strontium added via process chemicals. When commercial fuel processing is restarted, the need for including strontium recovery facilities should be carefully evaluated. Their inclusion more than doubles the available strontium-90 after 1990.

Cesium-137

The minimum cesium-137 content of fission-product cesium acceptable for use in cesium-137 irradiators has not been specified. If it is assumed that the cesium-137 remains useful for one half-life following reactor discharge, the minimum useful cesium-137 content would be about 20 percent. Very little of the recoverable cesium at Hanford or SRP could have decayed to this level, and the average would be somewhat higher. In the absence of any isotopic analyses on the cesium wastes, it was assumed that all recoverable cesium at both Hanford and SRP could be used. Cesium-137 recovery for byproduct use at SRP will require construction of facilities for product form conversion and for encapsulation.

The estimated inventories of cesium-137 available from all sources are shown in Table II.

Table II. Estimated Inventories of Cesium-137 from Various Sources[1-7,9]

Source	Effective Inventory Date	Ci
Hanford		
- Cesium chloride capsules through 6/82	1/83	6.36×10^7
- Cesium chloride capsules through 6/84	1/83	2.77×10^7
- Stored wastes[(a)]	1/83	2.6×10^7
- Future wastes	1/91	3.87×10^7
Savannah River Plant		
- Current wastes	1/83	1.02×10^8
- Future wastes	1/2001	1.09×10^8
Commercial Spent Fuel		
- Accum. through 1981	1/83	5.1×10^8
- Accum. through 2020	1/2021	1.1×10^{10}

(a) Cesium contained in current waste but not considered recoverable.

The estimated cumulative inventory of recoverable cesium-137 is shown in Fig. 2. These data show that 90 MCi of cesium-137 is currently available at Hanford for irradiator utilization. A "typical" irradiator would use 2 to 10 megacuries, so the current inventory is sufficient to fuel 10 to 40 "typical" irradiators. If facilities are constructed at SRP for the product form conversion and packaging of cesium-137, the cumulative amount of cesium-137 recoverable from Hanford operations and from SRP by 1990 will be about 250 megacuries. This amount alone will support a substantial irradiation industry. Beyond 1990 the available cesium-137 could be greatly increased by including cesium-137 recovered from commercial power reactor fuel.

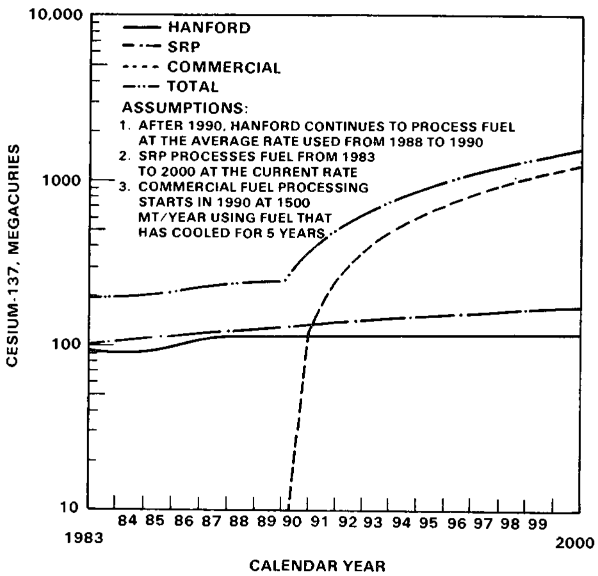

Fig. 2. Cumulative Inventory of Cesium-137 Available from High-Level Wastes for Beneficial Use

Krypton-85/Xenon

The potentially recoverable amounts of krypton-85 and xenon are given in Tables III and IV.

TABLE III. Estimates of Krypton-85 Potentially Recoverable from Future Fuel Processing[2,6]

Source	Inventory Date	^{85}Kr, at. %	10^6 Ci	Kr, kg
- Future Hanford fuels	1/91	6	5	210
- Future SRP fuels	1/2001	3.7[b]	14	965
- Commercial spent fuel - fuel in storage at the end of 1981	1/82	5.6	42	1,910
- Fuels accumulated thorough the year 2020[a]	1/2021	3.0[b]	630	53,500

(a) Assumes no interim processing.
(b) These values represent krypton accumulated through dates shown; thus the krypton-85 is low.

Table IV. Estimates of Xenon Potentially Recoverable from Future Fuel Processing[2,6,9]

Source	Effective Inventory Date	10^3 kg
- Future Hanford fuels	1/91	3
- Future SRP fuels	1/2001	6.1
Commercial spent fuel - fuel in storage at the end of 1981	1/82	28
- Fuel accumulated through the year 2020[a]	1/2021	1,790

(a) Assumes no interim processing.

At present, krypton and xenon are recovered from fuel processing operations at ICPP and are purified at ORNL. The current shortfall in the supply of krypton-85 is caused by limited recovery capacity, not by availability. The current estimated annual need for krypton-85 is 20 to 30 kCi, or about 3 percent of the annual release of krypton-85 from SRP. Construction of improved recovery facilities at ICPP or of a recovery plant at Hanford or SRP should meet all market needs.

Platinum Group Metals

The current defense wastes at Hanford and SRP contain an estimated 1400 kg of palladium, 2000 kg of rhodium, 6800 kg of ruthenium, and 3300 kg of technetium.[2-6,9,10] Continued processing through the year 2020 at currently anticipated schedules will increase these inventories by 40 to 50 percent. Spent commercial fuel is by far the most significant source of fission-product platinum group metals. By the year 2021 this fuel will contain an estimated 180,000 kg of palladium, 66,000 kg of rhodium, 333,000 kg of ruthenium, and 120,000 kg of technetium. Utilization of these materials will require 1) development of processes and construction of facilities that will adequately separate them from the other fission products, and 2) proper management of the radioactive palladium, rhodium, and ruthenium isotopes.

Neptunium

The potential sources of neptunium-237 are shown in Table V.

Neptunium-237 is currently recovered at SRP and is irradiated for the production of plutonium-238. Current needs are met by this source, and recovery of neptunium-237 is not planned when the Hanford Purex plant restarts.

Table V. Estimated Inventories of Neptunium-237 from Various Sources[2,3,7]

Source	Neptunium-237 Inventory Date	kg
Hanford		
- Current wastes[a]	1/83	140
- Future recovery[b]	1/91	200
Savannah River Plant		
- Current wastes[a]	1/83	20
- Future processing[c]		45 to 65 per year[d]
Commercial Spent Fuel		
- Accumulated through 1981	1/82	2,100
- Accumulated through 2020[e]	1/2021	66,400

(a) Neptunium-237 contained in insoluble sludge, not considered available for recovery for beneficial use.
(b) Neptunium-237 contained in spent N-Reactor fuel to be processed.
(c) Neptunium contained in spent defense fuels to be processed.
(d) Depends on mode of reactor operation selected.
(e) Assumes no interim processing.

Curium-244

Curium-244 may be used in selected applications where the spontaneous fission can be tolerated. There are no significant amounts in defense wastes, but current commercial spent fuel contains 93 kg (262 kW_t), and by the year 2020 the amount will increse to 1800 kg (5200 kW_t).

Promethium-147 and Americium-241

The current requirements for americium-241 are satisfied by recovery from processing of previously purified plutonium. Optimistically projected needs for promethium-147 and americium-141 represent less than 10 percent of the amounts of these materials that could be recovered at SRP or Hanford production sites. Some additional processing equipment and facilities would need to be installed.

The potential sources of americium-241 and promethium-147 are shown in Tables VI and VII.

Table VI. Estimated Inventories of Americium-241 and Americium-243 from Various Sources, kg[2-4,7,10]

Source	Effective Inventory Date	Am-241	Am-243
Hanford			
- Current wastes	1/83	13	0.63
- Future wastes	1/94	44-60[a]	3.6
Savannah River Plant			
- Current wastes	1/83	15	
- Future wastes	1/2001	16	
Commercial Spent Fuel			
- Accumulated through 1981	1/82	1,160	370
- Accumulated through 2020[b]	1/2021	97,400	11,700

(a) Americium-241 available from future wastes depend on fuel processing schedule used.
(b) Assumes no interim processing.

Table VII. Estimated Inventories of Promethium-147 from Various Sources[2-4,6]

Source	Effective Inventory Date	kg	10^6 Ci
Hanford			
- Current wastes	1/83	9	8.3
- Future wastes	1/91	39	36
Savannah River Plant			
- Current wastes	1/83	120	110
- Future wastes	1/2001	206	190
Commercial Spent Fuel			
- Accumulated through 1981	1/82	460	430
- Accumulated through 2020[a]	1/2021	3,300	3100

(a) Assumes no interim processing.

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REFERENCES

1. Data (^{90}Sr capsule computer printout) supplied by Rockwell Hanford Operations, Richland, Washington, June 30, 1982.

2. Department of Energy. September 1982. Spent Fuel and Radioactive Waste Inventories, Projections and Characteristics. DOE/NE-0017/R1, Washington, D. C.

3. Letter from J. R. Wetch, Rockwell Hanford Operations, Richland, Washington, July 30, 1982, to E. J. Wheelwright, Pacific Northwest Laboratory.

4. Rockwell Hanford Operations. 1980. Technical Aspects of Long-Term Management Alternatives for High-Level Defense Waste at the Hanford Site. RHO-LD-141, Richland, Washington.

5. Energy Research and Development Administration. 1977. Alternatives for Long-Term Management of Defense High-Level Radioactive Waste - Hanford Reservation, Richland, Washington. ERDA-77-44.

6. Teletype from W. R. Cornman, E. I. duPont de Nemours & Co., Savannah River Plant, Aiken, South Carolina, to E. J. Wheelwright, Pacific Northwest Laboratory.

7. Energy Research and Development Administration. 1977. Alternatives for Long-Term Management of Defense High-Level Radioactive Waste - Savannah River Plant, Aiken, South Carolina. ERDA-77-42/1, Washington, D. C.

8. Data Supplied by Rockwell Hanford Operations, Richland, Washington, July 12, 1982.

9. Data (^{137}CsCl capsule computer printout) supplied by Rockwell Hanford Operations, Richland, Washington, June 30, 1982.

10. W. W. Schulz, M. M. Beary, R. A. Watrous, R. G. Johnston, and J. V. Panesko. 1982. Inventories of and Technology for Recovery of Americium, Promethium, Rhodium, and Palladium Values at Hanford: A Preliminary Assessment. RHO-LD-170, Rockwell Hanford Operations, Richland, Washington.