CONF-770512--9

Origin and Characteristics of Low-Level Nontransuranic Waste from the Nuclear Fuel Cycle*

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To be presented at the Symposium on the Management of Low-Level Radioactive Waste at the Georgia Institute of Technology, Atlanta, Georgia on May 23-27, 1977.

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*Research sponsored by the Energy Research and Development Administration under contract with the Union Carbide Corporation.

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ORIGIN AND CHARACTERISTICS OF LOW-LEVEL NONTRANSURANIC WASTE FROM THE NUCLEAR FUEL CYCLE

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Abstract

Low-level nontransuranic wastes are generated in all nuclear fuel cycle operations. While the activity levels and radiotoxicities of these effluents are generally of a lower magnitude than other fuel cycle wastes, their large volumes and their appearance throughout the fuel cycle make their management a very real concern regardless of the fuel cycle option being considered.

For the purposes of this paper, low-level nontransuranic wastes are defined as wastes that contain less than about 10 nCi of long-lived alpha radiation per gram and have gamma radiations low enough to require only minimal biological shielding and remote handling.

Wastes from uranium mining and milling, UF_6 conversion, enrichment, fuel fabrication, and fuel reprocessing are examined with respect to their radionuclide content, volume, and chemical composition. Projections of total quantities through the end of this century are also presented. Fuel cycles based on recycling only uranium, and on recycling both uranium and plutonium, are considered.

1. INTRODUCTION

The sources and characteristics of wastes in the nuclear fuel cycle are especially important from a waste management standpoint. Those wastes containing large quantities of fission product and actinide elements have received considerable attention, and efforts are being made to reduce their volumes and to dispose of them safely. Most of these

wastes, though, are common to only one or two phases of the nuclear fuel cycle. Low-level nontransuranic wastes, on the other hand, are common to every phase, regardless of which fuel cycle option is employed. Typically, these wastes are viewed as nuisance wastes but their large rates of generation make them a major concern.

The term, "low-level nontransuranic," is ill-defined. For the purposes of this paper, low-level nontransuranic waste is defined as waste that contains less than 10 nCi of long-lived alpha radiation per gram and has a gamma radiation level sufficiently low to require only minimal biological shielding and remote handling. The volatile fission products and ^{14}C released during reprocessing are included.

The sources and characteristics of low-level nontransuranic wastes are examined for each phase (except reactor operations) of two LWR options: recycle of uranium only, and recycle of both uranium and plutonium. Projections of total quantities of wastes for these options through the year 2000 are also presented.

2. THE NUCLEAR FUEL CYCLE

The nuclear fuel cycle (Fig. 1) is comprised of seven basic steps: 1) uranium mining, 2) uranium milling, 3) conversion, 4) enrichment, 5) fuel fabrication, 6) reactor operations, and 7) fuel reprocessing. Fuel reprocessing closes the fuel cycle by recovering uranium, or both uranium and plutonium, from the spent fuel and returning them to the fuel cycle. The "uranium-only" recycle option recycles fissile uranium from the spent fuel rods. The amount of fissile uranium not "burned" in spent fuel is significant because fuel exposures are limited by the

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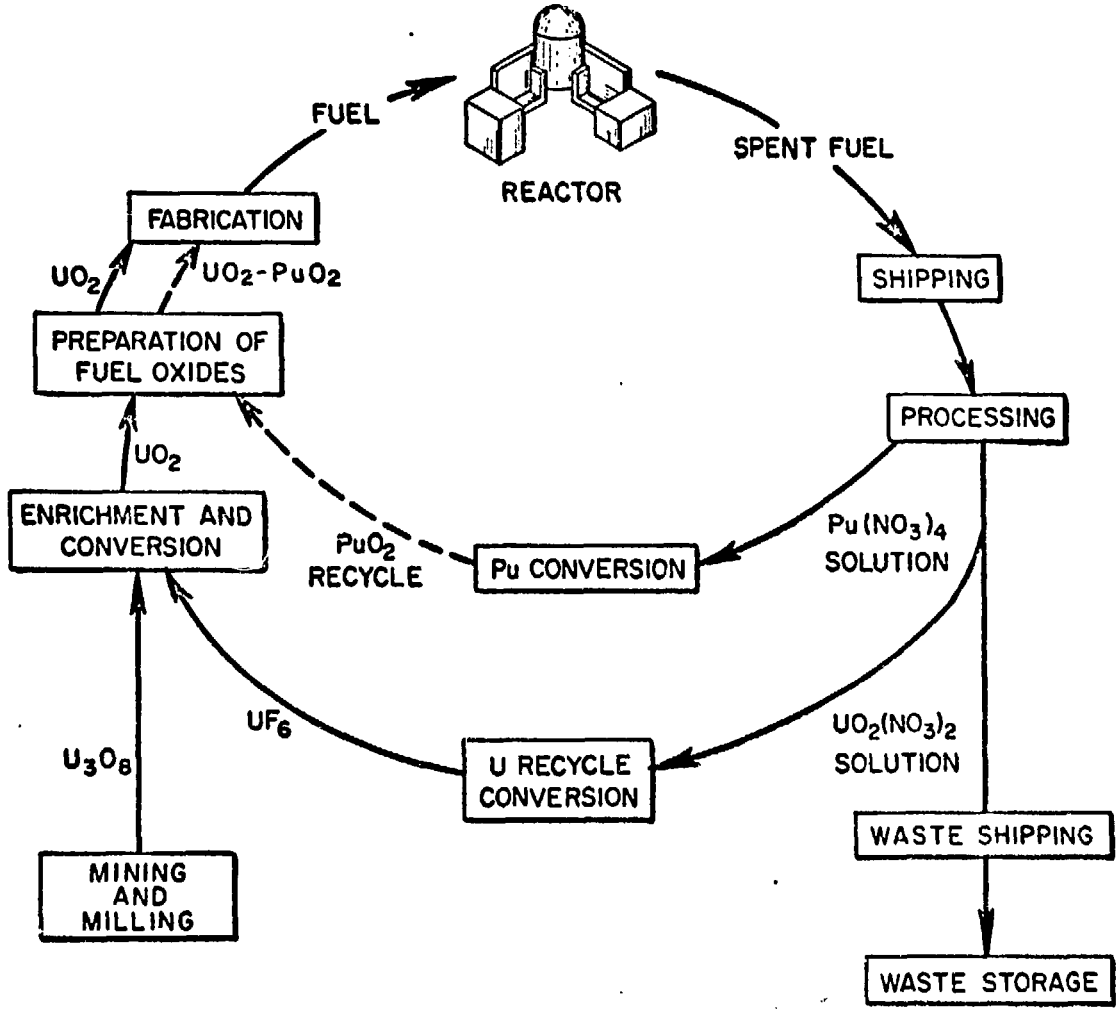
FUEL
REACTOR
SPENT FUEL
FABRICATION
UO_2
UO_2-PuO_2
SHIPPING
PREPARATION OF FUEL OXIDES
PROCESSING
UO_2
ENRICHMENT AND CONVERSION
PuO_2 RECYCLE
Pu CONVERSION
$Pu(NO_3)_4$ SOLUTION
UF_6
U_3O_8
U RECYCLE CONVERSION
$UO_2(NO_3)_2$ SOLUTION
WASTE SHIPPING
MINING AND MILLING
WASTE STORAGE

buildup of neutron poisons and by radiation damage. In the second recycle option, plutonium is recycled with the uranium because of its fissile value.

A brief synopsis of the nuclear fuel cycle follows with the intention of summarizing the basic operations involved so that the sources and characteristics of the wastes can be more easily examined and understood.

2.1 Uranium Mining

Uranium ore is presently mined in this country by two methods. The first method is open pit or surface mining, which is used when the ore is near the surface and beneath relatively friable rock. The second method, underground mining, is used when open pit mining is not feasible. Economically recoverable U.S. deposits of uranium ore are found in flat-lying sedimentary rock, primarily in the west and southwest. The uranium ore usually assays from 0.15 to 0.30 percent U_3O_8.

2.2 Milling

The purpose of the milling operation is to refine or concentrate the uranium in the ore. Actual milling operations vary from one mill to the other because of the variations in the ores; however, there are processes that are common to most types of uranium milling. The ore is initially pulverized and is transferred as a slurry to tanks where it is contacted with sulfuric acid and an oxidizing agent to dissolve (or leach) the uranium. The dissolved ore is pumped to a solvent-extraction system where the uranium is purified and concentrated to a product solution. Following this step, the uranium is subjected to various separa-

tion and consolidation processes such as precipitation, centrifugation, and calcination. The end product is concentrated (75 to 85%) U_3O_8.

2.3 UF_6 Conversion

The uranium concentrate from the milling operation is the feed material for the conversion process which continues to refine the uranium while converting it to the volatile UF_6. The two methods used in commercial conversion operations are the dry hydrofluor and the wet solvent extraction--fluorination processes.

The dry hydrofluor process carries the uranium concentrate feed through successive reduction, hydrofluorination, and fluorination in fluidized-bed reactors, after which the crude uranium hexafluoride is further purified and recovered by fractional distillation.

The wet process differs from the dry process in that aqueous-organic extraction is used to prepare a high-purity uranium oxide feed prior to the reduction, hydrofluorination, and fluorination steps.

2.4 Enrichment

Enrichment of uranium in the ^{235}U isotope is achieved by gaseous diffusion. This entails passing gaseous UF_6 through thousands of stages containing diffusion barriers which cause isotopes to be separated on the basis of their different masses. The product stream generally consists of several percent ^{235}U in ^{238}U, while the stream which is depleted in ^{235}U is commonly called the "tails". Since ^{238}U is a great potential energy resource, we do not consider enrichment tails to be a waste in a conventionally defined sense.

2.5 Fuel Fabrication

The current method for the fabrication of LWR fuel (2 to 4% ^{235}U) is the wet ammonium diuranate (ADU) process. In the ADU process, the UF_6 is first hydrolyzed to form UO_2F_2. The UO_2F_2 is then reacted with ammonium hydroxide to precipitate ammonium diuranate, and this is calcined to UO_2. After pelletizing, sintering, grinding, washing, and drying, the UO_2 is ready to be loaded into Zircaloy fuel rods.

Two fabrication processes are available for mixed oxide fuel fabrication. The mixed UO_2-PuO_2 powders can be produced by coprecipitation of ammonium diuranate and plutonium hydroxide by the addition of ammonia to a mixture of the uranyl and plutonium nitrate solutions. The second process involves the mechanical blending of UO_2 and PuO_2 powders followed by sintering and grinding steps. The plutonium oxide powder is formed from the conversion of plutonium nitrate to the oxide. This conversion process involves the formation of an insoluble plutonium oxalate and calcination of the oxalate to the oxide. The uranium dioxide powder is made up of natural or depleted uranium which has gone through the conventional ADU process.

2.6 Fuel Reprocessing

There are essentially six steps in the reprocessing of spent fuel. They consist of: 1) disassembly of the fuel assemblies and chopping of the fuel elements; 2) nitric acid leaching of the fuel from the cladding; 3) preparation of the nitric acid leach solution for the solvent-extraction process; 4) solvent-extraction, which separates the uranium and plutonium from the fission products and from each other; 5) puri-

fication of the uranium and plutonium products; and 6) conversion of the uranium stream to UF_6, and the plutonium stream to PuO_2.

3. SOURCES AND CHARACTERISTICS OF LOW-LEVEL NONTRANSURANIC WASTES

The sources and characteristics of low-level nontransuranic waste for each phase of the nuclear fuel cycle are summarized in Table 1.

3.1 Mining Wastes

Low-level nontransuranic wastes from mining operations are virtually nonexistent. Most of the waste produced is simply rock and earth overburden. However, there is some radioactivity released due mainly to the presence of the decay daughters of uranium. Calculations for a model mine indicate that 0.46 Ci of ^{222}Rn per metric ton of uranium would be released to the environment (ERDA76a).

3.2 Milling Wastes

Wastes from milling operations appear as gases, liquids, and solids. The liquid and solid wastes are generally combined and constitute the tailings.

Gaseous effluents are present throughout the life of the mill and are generated for the "radioactive life" of the tailings. Ore and yellow-cake dust, containing uranium and its daughters, are generated during all solids processing and handling operations. In addition, some radon is released during the milling operations, but the primary source of radon release is the tailings, especially before stabilization. Tailings dust and radon emanating from the tailings pile remain after the mill operations are terminated. The ^{222}Rn and its decay daughters

Table 1. Sources and characteristics of low-level nontransuranic waste

Source	Original waste form	Major radioisotopes	Activity (Ci/MTHM[a])	Final waste form
1. Mining	Gaseous	^{222}Rn	0.46	Gaseous
2. Milling	Gaseous	^{222}Rn	1.97	Gaseous
	Airborne particulates	U, ^{226}Ra, ^{230}Th	1.31×10^{-4}	Airborne particulates
	Liquid	U, ^{226}Ra, ^{230}Th	1.30×10^{-2}	7.4×10^{5} liters of liquid waste waste per MTU[b]
	Solid	U, ^{226}Ra, ^{230}Th	4.15	317 m^3 of tailings (solid) per MTU
3. UF_6 conversion (fresh)	Airborne particulates	U	1.38×10^{-5}	Airborne particulates
	Liquid	U, ^{226}Ra, ^{230}Th	1.07×10^{-6}	1.3×10^{4} liters of liquid waste per MTU
	Solid	U, ^{226}Ra, ^{230}Th	9.40×10^{-3}	6.9 m^3 of solid waste (including CaF_2) per MTU
4. UF_6 conversion (recycle)	Airborne particulates	Fission products	8.73×10^{-6}	Airborne particulates
		^{238}Pu, ^{241}Pu	2.46×10^{-6}	
		U	3.32×10^{-5}	
	Solid	Fission products	0.57	~7.0 m^3 of solid waste (including CaF_2 and carbonate leached ash) per MTU
		^{238}Pu, ^{241}Pu	4.12×10^{-5}	
		U	5.03×10^{-4}	
5. Enrichment (fresh)	Gaseous and airborne particulates	U	1.96×10^{-5}	Gaseous and airborne particulates
	Liquid	U	1.11×10^{-6}	675 liters of liquid waste per MTU
6. Enrichment (fresh + 10% recycle)	Gaseous and airborne particulates	U	1.98×10^{-5}	Gases and airborne particulates
		^{237}Np	8.33×10^{-11}	
		^{239}Pu	1.67×10^{-13}	
		Fission products	2.29×10^{-4}	

Table 1. (Continued)

Source	Original waste form	Major radioisotopes	Activity (Ci/MTHM[a])	Final waste form
	Liquid	U	$1.1[illegible] \times 10^{-6}$	675 liters of liquid waste per MTU
		^{237}Np	$1.6[illegible] \times 10^{-9}$	
		^{239}Pu	$3.3[illegible] \times 10^{-12}$	
		Fission products	3.40×10^{-4}	
7. Fuel fabrication (fresh + 10% recycle)	Airborne particulates	U, ^{234}Th, ^{234}Pa	$5.6[illegible] \times 10^{-6}$	Airborne particulates
	Liquid	U, ^{234}Th, ^{234}Pa	$1.2[illegible] \times 10^{-3}$	$5.7 \times 10^{[illegible]}$ liters of liquid waste per MTU
	Solid	U	7.34×10^{-3}	4.6×10^{-3} of solid waste (including CaF_2) per MTU
8. Fuel fabrication (mixed oxide)	Airborne particulates	U	$5.8[illegible] \times 10^{-12}$	Airborne particulates
		Pu, Am	2.08×10^{-7}	
	Solid	U, Pu, Am	3.47	~2265 (98.2) m^3 of solid waste per MT of plutonium (uranium) processed
9. Reprocessing	Gaseous	^{3}H	436-526[c]	~0.008-0.01[c], ~0.01-0.013[d] m^3 of $Ca(OH)_2$ per MTHM
			544-665[d]	
	Gaseous	^{127}I, ^{129}I	0.655-0.936[c]	20 liters of liquid scrubber waste per MTHM; ~0.0011-0.0013[c], ~0.0016-0.0019[d] m^3 of concrete per MTHM
		^{131}I	0.686-0.992[d]	
	Gaseous	^{85}Kr	7330-9100[c]	~0.05-0.06[c,d] cylinders (50 liters, 2200 psig) per MTHM
			5000-6020[d]	
	Gaseous	^{14}C	0.539-0.598[c]	$^{14}CO_2$
			0.293-0.314[d]	
	Solid	Pu, Am	0.53	~1586 (15.0) m^3 of solid waste per MT of plutonium (uranium) processed
		Fission products		

[a]Metric ton of heavy metal.
[b]Metric ton of uranium.
[c]First number is for a BWR-U; second number is for a PWR-U.
[d]First number is for a BWR-mixed oxide; second number is for a PWR-mixed oxide.

account for more than 99.99% of all the activity present in the gaseous effluents and are released at the rate of 1.97 Ci per metric ton of uranium (Se75). However, the release rate can be reduced if the tailings pile is stabilized with an earthcover or if the ^{230}Th and ^{226}Ra are chemically removed.

Large amounts of liquid waste are generated in milling operations, but the contamination is not so severe as in the other stream. About 1.5 tons of liquid waste are produced for every ton of ore processed, and the radionuclide concentrations vary from 3 to 10 pCi of uranium per milliliter, 0.4 to 11 pCi of radium per milliliter, and 11 to 500 pCi of thorium per milliliter (Se75). Typically, the total activity in all wastes from milling operations is 1.30×10^{-2} Ci per metric ton of uranium (ERDA76a). The usual means of release of the liquid effluents from the tailings pond retention area is by seepage.

The source of the tailings is at the point in the milling process where the leach liquor is separated from the undissolved solid matter. Greater than 90% of the uranium is present in the leach liquor, and the remainder of the uranium and its daughters are in the tailings. Process water and chemicals are routed along with the tailings to a retention pond for evaporation and storage.

The tailings pile is generated at the rate of 317 m^3 per metric ton of uranium (Ke76). At the time of discharge, it is contaminated with uranium and daughters to the level of 4.15 Ci per metric ton of uranium, and both the average uranium concentration and recovery efficiency are decreasing from historical values of about 0.2% U_3O_8 in

the ore and a 95% recovery. This decline will cause an increase in the quantity of the tailings per unit of uranium recovered.

3.3 UF_6 Conversion Wastes

3.3.1 Fresh Uranium

The conversion of virgin U_3O_8 to UF_6 is presently accomplished by the aforementioned dry Hydrofluor and wet solvent extraction--fluorination processes. The dry process is characterized by gaseous and solid waste streams while the wet process generates, primarily, a liquid waste stream. The dry process, emphasized here because it has fewer environmental impacts, operates with very low uranium losses [99.5% of the uranium input is recovered as UF_6 (ERDA76a)], and all solid process wastes undergo uranium recovery steps before being routed to waste streams for final disposition.

The gaseous effluents, as in all fresh uranium operations, are contaminated by uranium and its daughters. A conversion plant is estimated to release 2.04×10^{-5} metric ton of natural uranium (1.38×10^{-5} Ci) per metric ton of uranium from the stack (ERDA76a). Some possible points of origin are dust collector discharges and vent gases from the hydrofluorinator and fluorinator scrubbers. The gaseous emissions are most likely in the form of insoluble uranium oxide particulates or fluoride compound particulates in the respirable particle range.

Liquid effluents are routed to settling ponds to reduce the amount of suspended solids and then are diluted with process water before release. Occasionally, the liquid effluents are subjected to uranium recovery procedures before dilution and release. In spite of these pre-

cautions, a small amount of uranium remains with the liquid effluent. The release from the conversion step is estimated to be 1.07 μCi of uranium and its daughters per metric ton of uranium [primarily from scrap recovery and scrubber liquids] (ERDA76a).

The majority of the solid waste generated in the hydrofluor process is inert calcium fluoride, which is used to collect the impurities from the U_3O_8 during hydrofluorination and fluorination, In addition, potassium hydroxide, calcium sulfate, and small amounts of nonvolatile metal fluorides contribute to the solid waste stream. Most of these other solid wastes have their origin in scrubber solutions, washings, and in the distillation of UF_6 from the fluorine operations. A conversion plant generates 0.14 m^3 of insoluble waste per metric ton of uranium. This waste contains 9.4×10^{-3} Ci of activity, over 75% of which is attributable to ^{230}Th (ERDA76a). In addition, 6.79 m^3 of miscellaneous solid waste is generated per metric ton of uranium processed.

3.3.2 Recycle Uranium

The conversion of recycle uranium differs from the conversion of fresh uranium in that the feed material is a relatively high-purity uranyl nitrate rather than semi-refined U_3O_8, and that the recycle uranium is contaminated with very small amounts of transuranics and fission products. The change in the chemical makeup of the feed material requires that the uranyl nitrate solution from reprocessing be first evaporated to uranyl nitrate hexahydrate and then calcined to UO_3. The UO_3 may then be converted to UF_6 by the same procedure as is used for fresh yellow cake (AGNS72).

The presence of transuranics and fission products alters the characteristics of the waste streams that are generated when converting fresh uranium. The waste stream are primarily gaseous and solid, with no liquid effluents being released directly to the environment (AGNS72).

Those areas contributing most heavily to the concentration of the gaseous effluents are the uranyl nitrate storage vents, UO_3 wet and dry dust collector discharge, incinerated reductor off-gas, hydrofluorination vent gas scrubber, UF_4 dust collector discharge, fluorinator vent gas scrubber, waste calciner dust collector discharge, ash dust collector discharge, and the scrap recovery dust collector discharge. The activity released in gaseous effluents per metric ton of uranium consists of 3.32×10^{-5} Ci of uranium, 2.45 μCi of actinides and 8.76 μCi of fission products (Ro76).

Solid waste has its origins in the waste calciner discharge, carbonate-leached ash from uranium recovery, and spent fluorinator beds. The waste calciner discharge is mainly composed of calcium fluoride, potassium hydroxide, and sodium carbonate. The total low-level nontransuranic solid waste from these sources consists of 0.29 metric ton (0.20 m^3) per metric ton of uranium containing 292 grams of uranium, 333 μg of plutonium, and 0.57 Ci of fission products (AGNS72). Miscellaneous solid waste is generated at the rate of at least 6.8 m^3 per metric ton of uranium.

3.4 Enrichment Wastes

3.4.1 Fresh Uranium

Isotopic enrichment of ^{235}U in fresh fuel generates very little radioactive waste. Gaseous and liquid effluents are the only waste

streams generated by the diffusion process, and only a nominal amount of contaminated solid waste, consisting mainly of failed equipment and other miscellaneous items, is produced.

The sources of gaseous radioactive wastes in a diffusion plant are the purge cascade and the decontamination and uranium recovery operations. As in all cases of gaseous effluents, the amount released is dependent on the type of trapping system used. Most of the uranium particulates are removed by HEPA filters; gaseous uranium is removed by chemical trapping systems. The gaseous releases per metric ton of uranium are 16 μCi of ^{234}U, o.63 μCi of ^{235}U, and 2.7 μCi of ^{238}U (ERDA76c).

Liquid radioactive discharges originate in the decontamination and uranium recovery facility, and in the laundry. The liquid effluents are routed to a holding pond or a secondary sewage plant before disposal. The releases per metric ton of uranium are 0.6 μCi of ^{234}U, 0.02 μCi of ^{235}U, and 0.05 μCi of ^{238}U and are diluted with 675 liters of liquid before discharge (ERDA76a).

3.4.2 Recycle Uranium

The recycle uranium from either "uranium-only" recycle or mixed uranium-plutonium recycle must be re-enriched before being fabricated into fuel rods. Since it is a recycle material, the enrichment feed is contaminated with trace amounts of transmutation products, transuranics, and fission products. The UF_6 feed is passed through cobaltous fluoride (CoF_2) traps which remove most of the plutonium, neptunium, and fission products prior to entry into the diffusion cascade.

The release rates and sources of ^{234}U, ^{235}U, and ^{238}U do not change

with a switch from fresh uranium to recycle uranium; however, there is a slight increase in the radioactivity of the effluents due to the presence of transuranium element and fission product contamination. The gaseous effluents (per metric ton of uranium) contain 0.5 μCi of additional uranium, 0.2 pCi of ^{239}Pu, 83 pCi of ^{237}Np, and 2.3×10^{-4} Ci of fission products. The liquid effluents are increased by 0.06 μCi of additional uranium, 3.3 pCi of ^{239}Pu, 0.002 μCi of ^{237}Np, and 3.4×10^{-4} Ci of fission products (ERDA76c). The use of cobaltous fluoride traps produces a solid waste, however, if the trap is fully contaminated, it may exceed the long-lived alpha activity limit set by the definition of low-level nontransuranic waste (ERDA76c).

3.5 Fuel Fabrication Wastes

3.5.1 Fresh Uranium

The radioactive waste streams from operating UO_2 fuel fabrication plants consist entirely of uranium and its daughters. The principal chemical operation in fuel fabrication is the ADU process which converts UF_6 to UO_2.

The off-gas is contaminated with entrained particles of UO_2 or entrained droplets of process liquids. The estimated release from a fuel fabrication plant is 5.3 μCi of enriched uranium and its daughters per metric ton of uranium (ERDA76a).

Radioactive liquid effluents are produced in the main process line, recycle and scrap recovery operation, showers, floor drains, laboratory wastes, and laundry water. The liquid effluents are subjected to various treatments (neutralization, filtration, and centrifugation) in an

effort to recover any uranium in the stream and also to reduce the release of radioactivity. After undergoing the treatments, the liquid streams are routed to a holding pond before release from the site.

The approximate volume of the contaminated liquid waste is 5.7 x 10^5 liters per metric ton of uranium. The radioactive release corresponds to approximately 1.2 x 10^{-3} Ci of uranium and daughters per metric ton of uranium (ERDA76a).

The primary source of solid waste is the process waste treatment system which generates large amounts of contaminated calcium fluoride. Other types of contaminated solid waste are rags, clothing, floor sweepings, sump sludges, disposable filters, and filter residues. About 0.28 m^3 of CaF_2 containing 6 x 10^{-3} Ci of uranium is generated per metric ton of uranium processed, and about 4.3 m^3 of other contaminated solid waste (per ton of uranium) is also produced (ERDA76a).

3.5.2 Recycle uranium and plutonium

Fabrication of recycle uranium will not differ from fresh fuel fabrication, and the radioactive releases from the plant will differ only in the isotopic composition of uranium. The recycle uranium waste streams will include ^{236}U, while the fresh uranium waste streams do not. Fission product contamination is not detectable (Pe75), most likely due to the high-purity uranyl nitrate feed to the UF_6 (recycle) conversion plant and the cobaltous fluoride traps at the enrichment plant.

The waste streams from mixed oxide fuel fabrication plants are mostly transuranic in classification because of the large plutonium throughput.

The gaseous effluent stream consists of air and off-gas from three regions of the plant: 1) chemical process vessel off-gas, 2) the off-gas from alpha enclosures, and 3) ventilation air from personnel areas. Ventilation air is passed through a roughing filter and HEPA filter prior to entry into any area. Air and off-gases to and from alpha enclosures and process equipment are routed through roughing and HEPA filters and/or scrubbers. All gases are then passed through two more banks of HEPA filters before release. Uranium is released at the rate of 5.8 μCi per metric ton of heavy metal, and transuranics are released at the rate of 0.2 μCi per metric ton of heavy metal (Gr75).

The solid waste streams, including solidified liquid effluents, are mainly transuranic. The nontransuranic waste is made up chiefly of miscellaneous solids such as paper, cloth, fiberglass, and metals. Based on ERDA operating experience, a mixed oxide fuel fabrication plant might generate 2265 m^3 of low-level nontransuranic waste (3.5 mCi per m^3) per ton of plutonium processed (Go71).

3.6 Fuel Reprocessing Wastes

The waste streams from fuel reprocessing are diverse in character and classification. Low-level nontransuranic wastes will consist of the fission gases (tritium, iodine, and krypton), ^{14}C, and naturally, miscellaneous solid refuse.

3.6.1 Tritium

The level of fission product tritium in spent reactor fuels varies according to the fissile content of the fuel and its burnup. However,

calculations indicate that about 16 Ci/GWd(th) and 20 Ci/GWd(th) are generated in enriched UO_2 and mixed (U-Pu) oxide fuels, respectively.

In current reprocessing flowsheets, tritium becomes distributed in all liquid process streams and would be eventually discharged in gaseous and liquid effluents unless measures are taken to separate and concentrate it. One possibility is to incorporate a voloxidation step in the head-end of the plant (Goo73). In this event, tritium might be recovered as tritiated water at a concentration of about 200 Ci per liter, immobilized at $Ca(OH)_2$, and disposed of as a solid waste. One liter of the concentrated tritiated water could be immobilized in approximately 4 liters of $Ca(OH)_2$ (Ke76).

3.6.2 Iodine

Iodine is another volatile fission product and is principally made up of the three isotopes ^{127}I, ^{129}I, and ^{131}I. Since ^{127}I is a stable isotope, its impact on the waste stream is mainly as a mass addition. The critical long-term waste management problem arises for ^{129}I, which has a half-life of 1.59×10^7 years.

The generation of fission product iodine for each reactor fuel at the time of its discharge and at a reprocessing time 160 days later is summarized in Table 2.

The release of iodine to the environment ensures that ^{129}I will become a permanent component of the environment and will continue to increase in direct relation with the amount of fuel reprocessed. Therefore, the collection and permanent isolation of iodine is a probability.

Presently, most of the iodine from the fuel appears in the off-gas

Table 2. Iodine content of reference spent fuels[a]

Reactor[b]	Isotope	g/MTHM[c] at discharge	Ci/MTHM at discharge	g/MTHM 160 d later	Ci/MTHM 160 d later
BWR-U	^{127}I	3.98×10^{1}	stable	4.07×10^{1}	stable
	^{129}I	1.63×10^{2}	2.84×10^{-2}	1.64×10^{2}	2.87×10^{-2}
	^{131}I	4.78	5.92×10^{5}	5.05×10^{-6}	6.26×10^{-1}
BWR-Pu	^{127}I	6.16×10^{1}	stable	6.27×10^{1}	stable
	^{129}I	2.20×10^{2}	3.83×10^{-2}	2.22×10^{2}	3.87×10^{-2}
	^{131}I	4.93	6.12×10^{5}	5.21×10^{-6}	6.47×10^{-1}
PWR-U	^{127}I	4.62×10^{1}	stable	4.75×10^{1}	stable
	^{129}I	1.92×10^{2}	3.35×10^{-2}	1.94×10^{2}	3.39×10^{-2}
	^{131}I	6.87	8.52×10^{5}	7.27×10^{-6}	9.02×10^{-1}
PWR-Pu	^{127}I	7.45×10^{1}	stable	7.61×10^{1}	stable
	^{129}I	2.66×10^{2}	4.64×10^{-2}	2.69×10^{2}	4.70×10^{-2}
	^{131}I	7.21	8.94×10^{5}	7.62×10^{-6}	9.45×10^{-1}

[a]Based on exposures of 33,000 MWd/MTHM at 30.0 MW/MT for PWRs and 27,500 MWd/MTHM at 20.7 MW/MT for BWRs.
[b]BWR-U and PWR-U represent enriched uranium oxide-fueled reactors, while BWR-Pu and PWR-Pu represent mixed (U-Pu) oxide-fueled reactors.
[c]MTHM designates "metric tons of heavy metal."

from the fuel dissolution step (Th76). To prevent large releases to the atmosphere, it may be removed by mercuric nitrate-nitric acid scrubbers in series (anticipated DF of 100). Other potentially more efficient processes for recovering iodine include voloxidation and scrubbing with concentrated nitric acid (Iodox). In the latter case, the iodine could be converted to barium iodate and mixed with cement, water, and butyl stearate to form a stable solid waste. One cubic foot of the solid product could contain 5.3 kg of iodine (Ke76).

3.6.3 Noble gases

The noble gas fission products are made up of xenon and krypton, both stable and radioactive isotopes. They are released from the fuel during the chop-leach stage, and initial plans were to release the noble gases to the atmosphere. However, prospective buildup of ^{85}Kr in the atomosphere in an expanding nuclear economy makes its retention a future probability.

The ^{85}Kr (10.73-year half-life) in uranium-fueled LWRs accounts for about 0.4 wt % of the total noble gas mixture (Table 3). At the time of discharge of the spent fuel, only aobut 0.1% of the noble gas activity is due to ^{85}Kr. Yet, 160 days later, ^{85}Kr is responsible for 99.98% of the total noble gas activity.

The ^{85}Kr in mixed oxide fuel accounts for about 0.3 wt % of the noble gas mixtures. At the time of discharge only about 0.08% of the total noble gas activity is due to ^{85}Kr, while 160 days later it is responabile for 99.97% of the total noble gas activity.

Table 3. Noble gas content of reference spent fuels[a]

Reactor[b]	Isotope	g/MTHM[c] at discharge	Ci/MTHM at discharge	g/MTHM 160 d later	Ci/MTHM 160 d later
BWR-U	^{85}Kr	1.92×10^{1}	7.54×10^{3}	1.87×10^{1}	7.33×10^{3}
	Kr and Xe	4.73×10^{3}	6.36×10^{6}	4.73×10^{3}	7.33×10^{3}
BWR-Pu	^{85}Kr	1.31×10^{1}	5.14×10^{3}	1.27×10^{1}	5.00×10^{3}
	Kr and Xe	4.55×10^{3}	6.70×10^{6}	4.54×10^{3}	5.00×10^{3}
PWR-U	^{85}Kr	2.39×10^{1}	9.36×10^{3}	2.32×10^{1}	9.10×10^{3}
	Kr and Xe	5.76×10^{3}	1.06×10^{6}	5.76×10^{3}	9.10×10^{3}
PWR-Pu	^{85}Kr	1.58×10^{1}	6.19×10^{3}	1.53×10^{1}	6.02×10^{3}
	Kr and Xe	5.57×10^{3}	9.51×10^{6}	5.57×10^{3}	6.02×10^{3}

[a]Based on exposures of 33,000 MWd/MTHM at 30.0 MW/MT for PWRs and 27,5000 MWd/MTHM at 20.7 MW/MT for BWRs.
[b]BWR-U and PWR-U represent enriched uranium oxide-fueled reactors, while BWR-Pu and PWR-Pu represent mixed (U-Pu) oxide-fueled reactors.
[c]MTHM designates "metric tons of heavy metal."

Several processes are under development for the recovery and retention of the noble gases (ERDA76b) and, if recovered, these gases will probably be either stored under high pressure in gas cylinders, or trapped in zeolites.

3.6.4 Carbon-14

Carbon-14 is formed in the fuel, core hardware, and the cooling water. However, from a reprocessing standpoint, only the ^{14}C formed in the fuel needs to be considered. Carbon-14 is formed during irradiation of oxide fuels primarily by an n,p reaction with ^{14}N and secondarily by an n,α reaction with ^{17}O.

If 25 ppm of nitrogen impurity is assumed (Da77), the ^{14}C contents of the four reference reactor fuels of Tables 2 and 3 are as shown in Table 4.

Table 4. Levels of ^{14}C in reference spent fuels

Reactor	Ci/MTHM
BWR-U	0.539
BWR-Pu	0.293
PWR-U	0.598
PWR-Pu	0.314

It is believed that ^{14}C becomes a gaseous effluent during dissolution of the fuel and, if necessary, can probably be recovered and converted to a solid form suitable for shipment and isolation.

3.6.5 Low-level solid wastes

Low-level nontransuranic solid wastes from reprocessing come from

the general reprocessing operations and are comprised of miscellaneous solids and liquids. Based on ERDA operating experience, a reprocessing plant may generate, in final packaged form, 1586 m^3 (includes plutonium conversion wastes) of low-level nontransuranic waste per ton of plutonium processed. The activity of the waste may average 0.001 Ci of mixed fission products (Ke76).

4. PROJECTIONS

Projections of low-level nontransuranic wastes are presented for both recycle options through the year 2000. The uranium cycle is based on the ERDA Office of Planning and Analysis "Low Case" nuclear power growth, and the recycle of uranium and plutonium is based on the ERDA "Mid Case". The mid case differs from the low case not only in greater nuclear power growth, but also in the use of mixed oxide-fueled LWRs, HTGRs, and LMFBRs.

Several assumptions were used in making the waste-projections. The projections for wastes from conversion of fresh and recycle uranium, fabrication of uranium and mixed oxide fuels, and the solid wastes from reprocessing assume that the entire waste load was generated at the end of the year. This is not an unreasonable assumption in that most of the waste is contaminated with long-lived radioactivity and an average decay time of one-half year (during the year of generation) would not significantly alter the accumulated activity over a thirty seven year period. Decay is assumed in the other wastes because of the influence of shorter-lived isotopes.

The average decay time for the tailings, fission product gases

and ^{14}C is approximately one-half year during the year of generation.

The projections for the tailings, conversion, and fabrication were made only for the nuclear fuel cycle examined in this paper. Wastes from HTGRs and LMFBRs are not considered. The mid case projections (Ke76) for the fission product gases and ^{14}C do include the contirbution from HTGRs and LMFBRs, but their overall impact is assumed to be minimal because of their small installed capacities.

The growth of nuclear power in the United States and the related fuel cycle requirements are shown in Table 5. The projections are presented in Table 6.

5. ACKNOWLEDGEMENT

The authors express their appreciation to H. W. Godbee and A. H. Kibbey of Oak Ridge National Laboratory for their many helpful suggestions and for making available information from their files that was of great use in the preparation of this paper.

Table 5. Projected growth of nuclear power in the United States

Year	Uranium recycle[a]	Uranium and plutonium recycle[b]				
	LWR-U	LWR-U	LWR-Pu	HTGR	LMFBR	Total
		Installed Capacity [GW(e)]				
1980	60.1	67.8		0.3	0.0	68.1
1985	126.8	146.1		0.3	0.4	146.8
1990	194.8	250.2		0.3	1.3	251.8
1995	283.0	367.2		1.3	2.3	370.8
2000	380.2	468.1		10.0	32.0	510.1
		Uranium Mining Requirements (MTHM/year)				
1980	14,606					16,112
1985	26,329					30,536
1990	36,282					41,822
1995	49,747					54,370
2000	61,127					65,358
		Fuel Fabricated (MTHM/year)				
1980	2,158	2,550	0	2.3	0.0	2,552
1985	4,472	4,877	499	2.3	5.0	5,383
1990	6,128	6,902	1171	2.3	32.6	8,108
1995	8,527	9,138	1722	39.1	95.8	10,995
2000	10,611	10,997	1644	129.2	887.2	13,658
		Fuel Reprocessed (MTHM/year)				
1980	0	0		2.2	0.0	2.2
1985	2000	2000		2.1	0.0	2,002
1990	5000	5000		2.1	5.1	5,007
1995	5836	9100		2.0	17.1	9,119
2000	7931	9993		40.0	256.6	10,290

[a]ERDA Office of Assistant Administrator for Planning and Analysis, "Low Case" (August 1976).
[b]ERDA Office of Assistant Administrator for Planning and Analsis, "Mid Case" (August 1976).

Table 6. Accumulated properties of low-level nontransuranic fuel cycle wastes through 2000 A.D.

Calendar year	Uranium recycle Volume (m^3)	Uranium recycle Activity (Ci)	Uranium and plutonium recycle Volume (m^3)	Uranium and plutonium recycle Activity (Ci)
		Uranium Tailings		
1980	2.9×10^7	3.0×10^5	3.1×10^7	3.0×10^5
1990	1.1×10^8	1.3×10^6	1.3×10^8	1.4×10^6
2000	2.7×10^8	3.0×10^6	3.1×10^8	3.4×10^6
		UF_6 Conversion (Fresh Uranium)		
1980	6.5×10^5	2.0×10^3	7.0×10^5	2.1×10^3
1990	2.5×10^6	7.8×10^3	2.9×10^6	9.0×10^3
2000	6.0×10^6	1.9×10^4	6.7×10^6	2.1×10^4
		UF_6 Conversion (Recycle Uranium)		
1980	0.0	0.0	0.0	0.0
1990	1.8×10^5	4.8×10^3	1.8×10^5	4.8×10^3
2000	6.5×10^5	1.0×10^4	8.2×10^5	1.3×10^4
		Fuel Fabrication (Fresh Uranium)		
1980	6.7×10^4	2.0×10^2	7.2×10^4	2.1×10^2
1990	2.7×10^5	7.8×10^2	3.0×10^5	9.2×10^2
2000	6.7×10^5	2.0×10^3	7.2×10^5	2.3×10^3
		Fuel Fabrication (Mixed Oxide)		
1980			0.0	0.0
1990			4.7×10^5	4.4×10^4
2000			2.3×10^6	2.4×10^5
		Fuel Reprocessing		
		Tritium		
1980	0.0	0.0	0.0	0.0
1990	1.6×10^2	7.4×10^6	1.7×10^2	7.5×10^6
2000	7.0×10^2	2.7×10^7	9.1×10^2	3.6×10^7

Table 6 (Continued)

Calendar year	Uranium recycle Volume (m^3)	Uranium recycle Activity (Ci)	Uranium and plutonium recycle Volume (m^3)	Uranium and plutonium recycle Activity (Ci)
		Iodine		
1980	0.0	0.0	0.0	0.0
1990	2.3×10^1	5.8×10^2	2.3×10^1	6.0×10^2
2000	9.8×10^1	2.5×10^3	1.3×10^2	3.3×10^3
		^{85}Kr		
1980	0.0[a]	0.0	0.0[a]	0.0
1990	1094	1.2×10^8	1094	1.2×10^8
2000	4722	4.4×10^8	5829	5.5×10^8
		^{14}C		
1980		0.0		0.0
1990		1.3×10^4		1.3×10^4
2000		5.0×10^4		5.8×10^4
		Miscellaneous Solids		
1980	0.0	0.0	0.0	0.0
1990	3.9×10^5	4.4×10^3	3.9×10^5	4.4×10^3
2000	1.4×10^6	9.7×10^3	1.7×10^6	1.3×10^4

[a]All volumes for ^{85}Kr are reported as numbers of cylinders of ^{85}Kr (50 liters, 220 psig).

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