

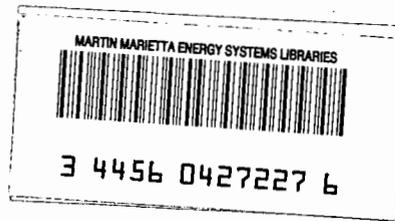
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An Evaluation of Retention and Disposal Options for Tritium in Fuel Reprocessing

W. R. Grimes
D. C. Hampson
D. J. Larkin
J. O. Skolrud
R. W. Benjamin

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Consolidated Fuel Reprocessing Program

**EVALUATION OF RETENTION AND DISPOSAL OPTIONS
FOR TRITIUM IN FUEL REPROCESSING**

W. R. Grimes* and D. C. Hampson†
Oak Ridge National Laboratory

D. J. Larkin and J. O. Skolrud
Exxon Nuclear Company, Inc.

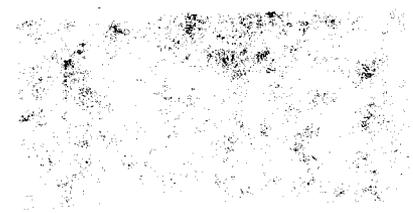
W. Benjamin
Savannah River Laboratory

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*Chemical Technology Division
†Fuel Recycle Division

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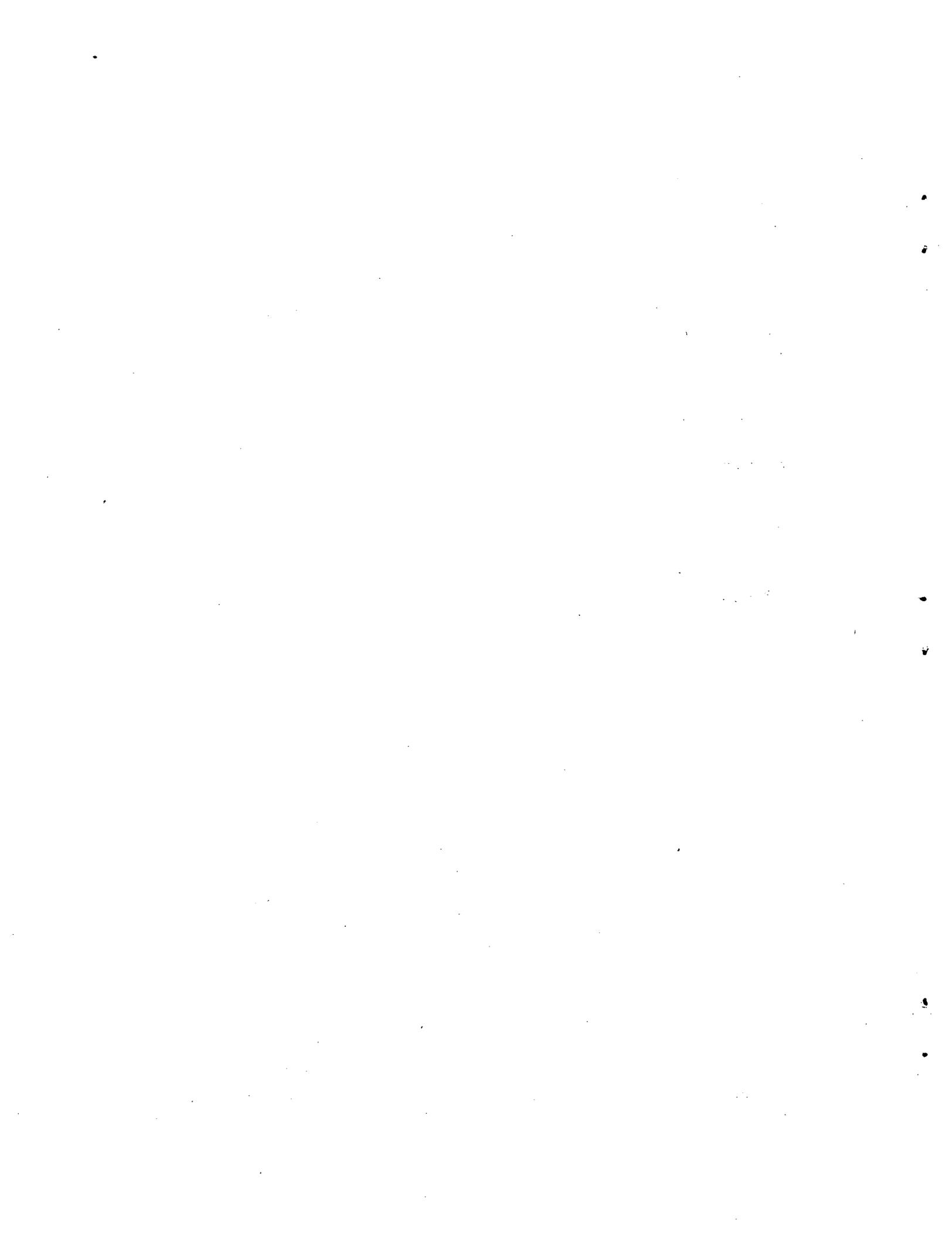
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ABSTRACT

Five options were evaluated as means of retaining tritium released from light-water reactor or fast breeder reactor fuel during the head-end steps of a typical Purex reprocessing scheme. Cost estimates for these options were compared with a base case in which no retention of tritium within the facility was obtained.

Costs were also estimated for a variety of disposal methods of the retained tritium. The disposal costs were combined with the retention costs to yield total costs (capital plus operating) for retention and disposal of tritium under the conditions envisioned.

The above costs were converted to an annual basis and to a "dollars per curie retained" basis. This then was used to estimate the cost in dollars per man-rem saved by retaining the tritium. Only the options that used the least expensive disposal costs could approach the \$1000/man-rem cost used as a guide by the Nuclear Regulatory Commission.



1. EXECUTIVE SUMMARY

The process of voloxidation has been under investigation and development at the Oak Ridge National Laboratory for a number of years. This process was developed as a means of separating tritium from irradiated reactor fuel prior to dissolving the fuel in nitric acid for the reprocessing operations. This document reexamines several aspects of isolation of tritium by commercial reprocessing plants to assist in decisions as to whether tritium isolation is needed and as to what technologies would be appropriate if such a need exists. Consequently, this study was to provide a reexamination of the need for a tritium isolation process for reprocessing and an evaluation of various options, including voloxidation, if this need does exist.

This study assesses in some detail the possible options (including voloxidation) for retention of tritium and the ultimate disposal of this isotope during future reprocessing of irradiated oxide fuels discharged from light-water reactors (LWRs) and liquid-metal fast breeder reactors (LMFBRs). The assessment includes an appraisal of the state of the retention and disposal, an estimate of the dose commitments to the general public, and the potential reduction of the dose commitments resulting from retention and disposal of the tritium.

Separate plants, one to process LWR fuel and one to process LMFBR fuel, were defined for this study. Each of the hypothetical plants was assumed to be constructed on a large site at Oak Ridge, Tennessee, during the 1990's and to operate for a 20-year lifetime beginning in year 2000 at a rate of 1500 metric tons of heavy metal (MTHM) per 300-d year.

Five options for retention of tritium were examined and compared with the base-case (sometimes referred to as reference case) option, which does not provide any capability for retaining tritium. These base-case plants (LWR-I and LMFBR-I) are assumed to recycle process acid but to use minimal recycle of process water and to discharge all releasable tritium as tritiated water (HTO) to the atmosphere by vaporizing the large volume of excess water.

In addition to the base case, other options examined (see Fig. 1.1) were:

- Option II The tritium is concentrated by means of recycling 90% of the water, in addition to the acid; 12% of the water and tritium is vaporized to the stack; and a sidestream from the recycle water stream is disposed of by various options.
- Option III Tritium is concentrated by means of the voloxidation option and is disposed of by various options.
- Option IV Tritium is separated by various means of isotopic separation from the 90% recycle stream and is disposed of by various options.
- Option V Tritium is separated by various means of isotopic separation from the discharge or nonrecycle stream (10%) and disposed of by various options.
- Option VI The aqueous streams in the plants are isolated so that tritium can be confined and concentrated in the head-end portions of the plant. This concentrate can be disposed of by various options.

Simplified flowsheets (presented, along with more detailed descriptions of the retention options, in Sect. 4.1) were prepared for each of the options; water and tritium balances

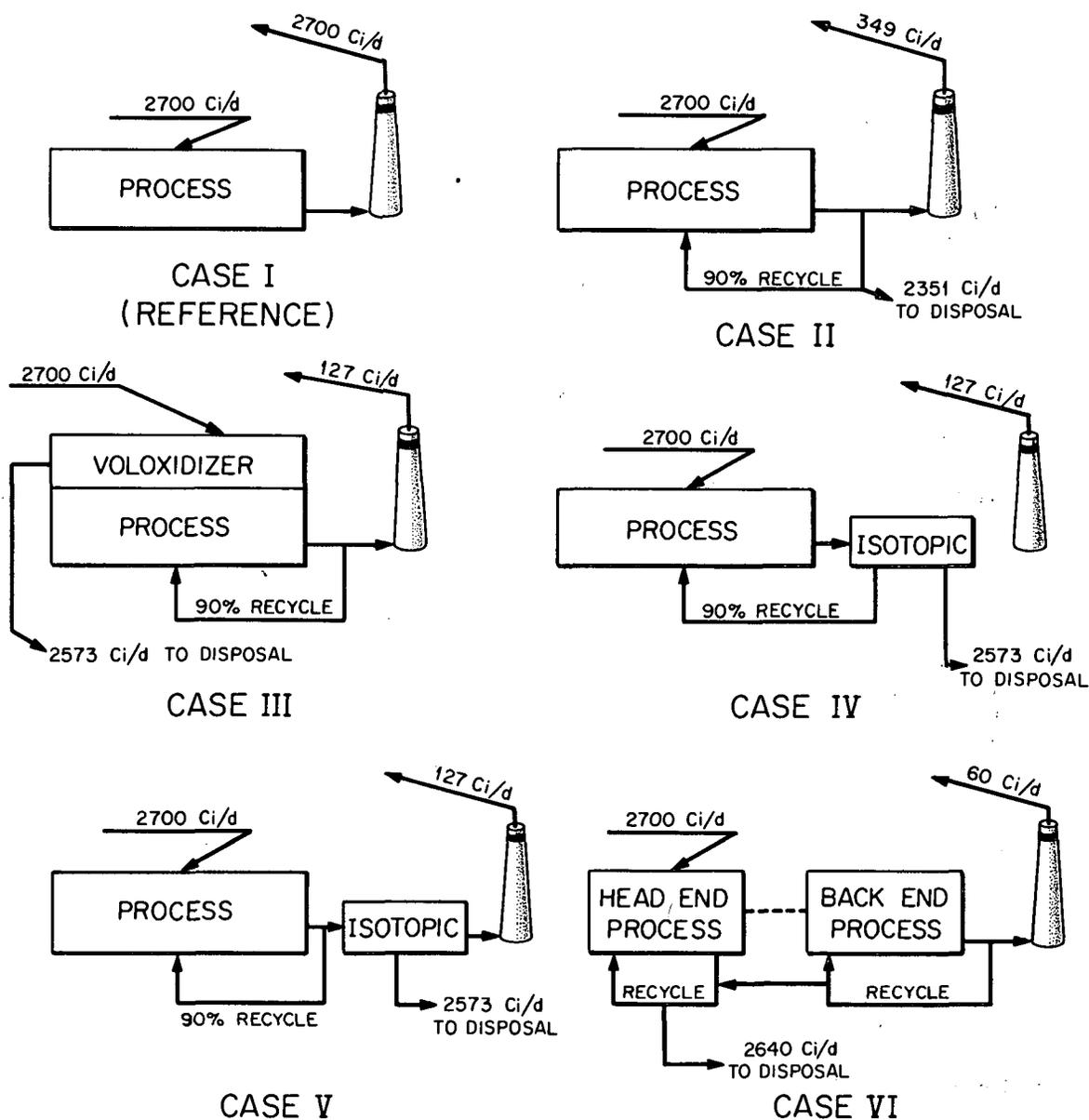


Fig. 1.1. Tritium retention options (LWR).

were determined for each. For the LWR and LMFBR retention options, the water volumes, tritium quantities, and tritium concentrations dispersed to the atmosphere and retained for disposal, as well as tritium concentrations in recycle acid, recycle water, and vessel off-gas, are shown in Table 1.1. All retention options markedly reduce the emission of tritium to the atmosphere relative to the base case. In addition, some, particularly option II (employing voloxidation) and option V (employing isotope separation on the discharge water), provide very small volumes of tritium concentrate for disposal. Some, notably option IV (employing isotope separation on total water recycle) and option II (employing voloxidation), provide recycle water with relatively small concentrations of tritium. In contrast, options II, V, and VI provide recycle water with relatively high tritium concentrations. It can also be observed that the quantity of tritium retained is almost the same for all of the options.

Table 1.1. Summary of water volumes and tritium concentrations for various retention options

Disposition	Retention options					
	I	II	III	IV	V	VI
LWR						
Dispersed through stack						
Tritium, Ci/d	2700	349	127	127	127	60
Retained for disposal						
Water, m ³ /d	0	12.94	0.048	3	0.288	8.86
Tritium, Ci/d	0	2351	2573	2573	2573	2640
Tritium, Ci/m ³	0	182	53 600	858	8930	350
Recycle acid						
Tritium, Ci/m ³	21.2	182	7.30	22.1	182	390
Recycle water						
Tritium, Ci/m ³	0	182	7.38	1.30	182	300
LMFBR						
Dispersed through stack						
Tritium, Ci/d	425	53	28	28	28	17
Retained for disposal						
Water, m ³ /d	0	27.3	0.048	3	0.288	8.78
Water, Ci/d	0	372	397	397	397	408
Tritium, Ci/m ³	0	13.6	8270	132	1378	46.4
Recycle acid						
Tritium, Ci/m ³	1.6	14.0	1.3	1.7	14.0	47.0
Recycle water						
Tritium, Ci/m ³	0	14.0	1.3	0.13	14.0	46.4

The "disposal" options considered in this study are described in detail in Sect. 4.2. The options range from storage for 50 or 100 years as tritiated water in tanks with subsequent dispersal of the undecayed HTO to the atmosphere to isolation (in positions interstitial to solidified high-level waste) in a geologic repository. The 50- or 100-year storage option would result in appreciable decay of the tritium (half-life of 12.33 years). After 50 years the value would be 6% of the original, and after 100 years it would be only 0.4% of the original.

Other disposal options include shallow trench burial for drummed concrete, clay matrix burial of poured or drummed concrete, deep-well disposal of water, shale hydrofracture with concrete grout, and geologic or sea-bed disposal of concrete canisters or drums.

Several of the disposal options require solidification of the tritiated waters. A number of solidification techniques were considered, but none appeared capable of yielding a solid with appreciably smaller volume than did fixation in concrete, and none appeared to be as economical. As a consequence, all disposal options that require solidification of the tritiated waters were assumed to use concrete. Table 1.2 shows the quantities and specific activities of tritiated water and concretes requiring disposal from each of the retention options for

**Table 1.2. Quantities and specific activities
of tritiated water and concretes**

Retention option	Concrete volume (m ³ /year)	Specific activity (Ci/m ³)	Years to qualify for unrestricted burial (0.05 Ci/m ³)
LWR-II	7 500	94	135
LWR-III	27	28 600	236
LWR-IV	1 600	480	160
LWR-V	160	4 830	205
LWR-VI	4 900	162	144
LMFBR-II	15 000	7.65	90
LMFBR-III	500	239	150
LMFBR-IV	1 600	73.3	130
LMFBR-V	160	756	171
LMFBR-VI	4 700	25.8	111

both fuel types. Included are the storage years that would be required to qualify for no further control of the burial area.

Capital and operating costs incremental to the base-case plants were estimated by the same method for each of the retention options. The base-case plants (for which the incremental costs were, of course, zero) were conceptually divided into about 30 functional modules. Those modules of the base-case plants that required additions, deletions, or modifications were identified for each of the retention options, and the capital costs (positive or negative) for design, fabrication, and installation of the required major in-cell equipment items were estimated. The capital cost estimates for in-cell support equipment, shielding walls, windows, wall penetrations, etc.; additions (service galleries, maintenance facilities, etc.) to the "balance of building;" and additions to the "balance of plant" were prorated based on equipment cost estimates. Capital costs for the disposal options were estimated in the same manner and included each of the categories described. All estimates are in first quarter 1980 dollars.

For both the retention and disposal options, the capital costs were converted to an annual capitalized cost by assuming a 20-year plant lifetime with zero salvage value at end-of-life and include the cost of capital. Combining these annual capitalized costs with the operating costs yielded uniform annual costs for each of the options. These costs were used to calculate the incremental cost per curie of tritium for the various retention and disposal options. These costs are additional costs above the base-case costs, and the curie reductions are also referenced to base-case releases. These values are presented in Fig. 1.2 for LWR options and Fig. 1.3 for LMFBR options.

The following can be observed from the figures:

1. Long-term tank storage is always the most expensive disposal option; the longer the storage, the higher the cost.
2. Voloxidizer costs are relatively independent of disposal methods.

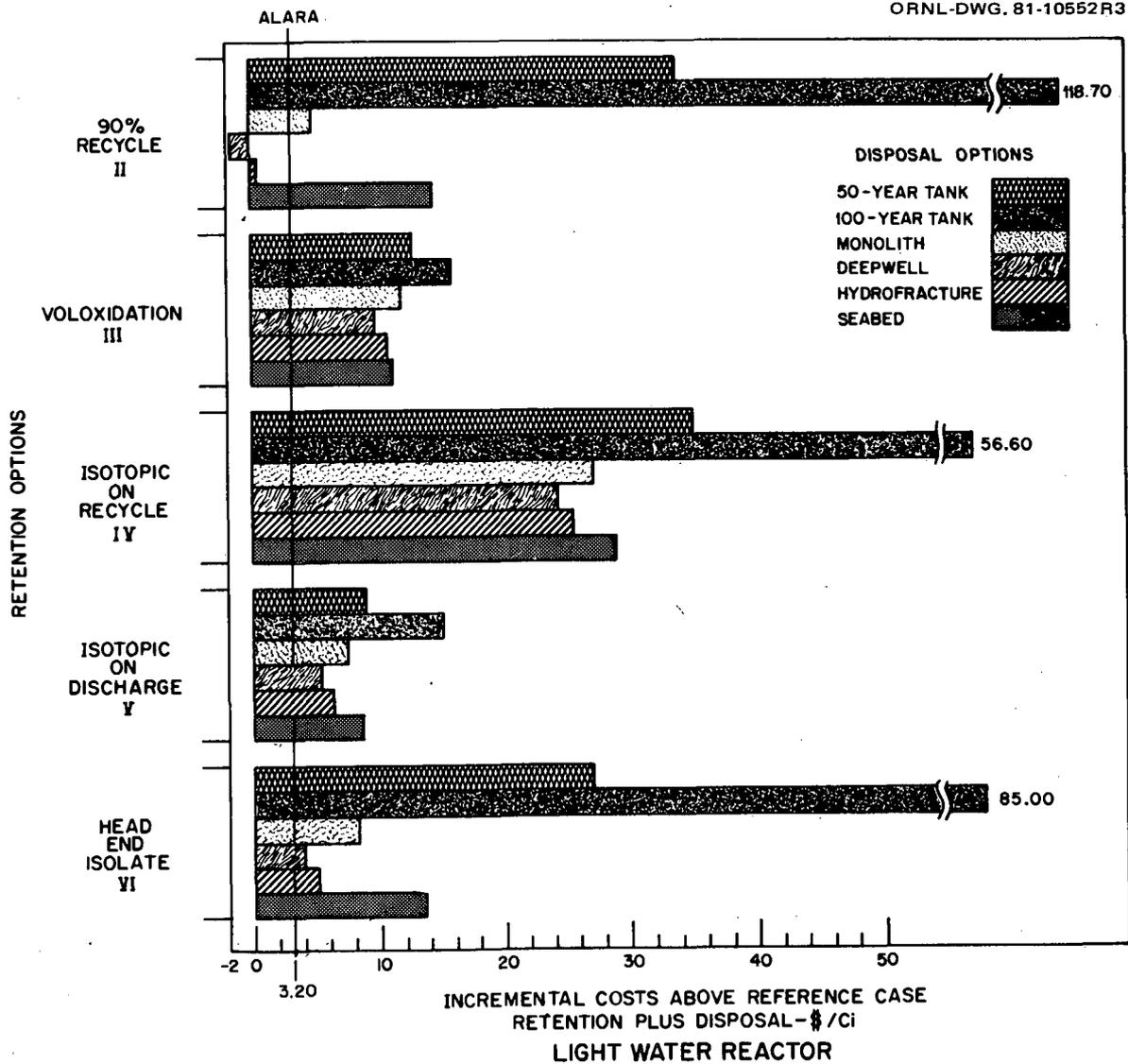


Fig. 1.2. Cost summaries - LWR.

- Monolith, hydrofracture, and deep-well disposal of the tritium concentrate are essentially equal for all cases involving concentration of tritium (options III, IV, and V).
- Sea-bed disposal is 10 to 100% more expensive than any of item 3.
- Deep-well and hydrofracture disposal costs are a small fraction of the retention costs.
- LMFBR dollars per curie are always much higher than LWR dollars per curie for the same options because LMFBR fuel contains markedly less tritium.
- LMFBR costs for the concentration options (III, IV, and V) are essentially independent of disposal options costs; that is, entirely dependent on retention costs in dollars per curie.

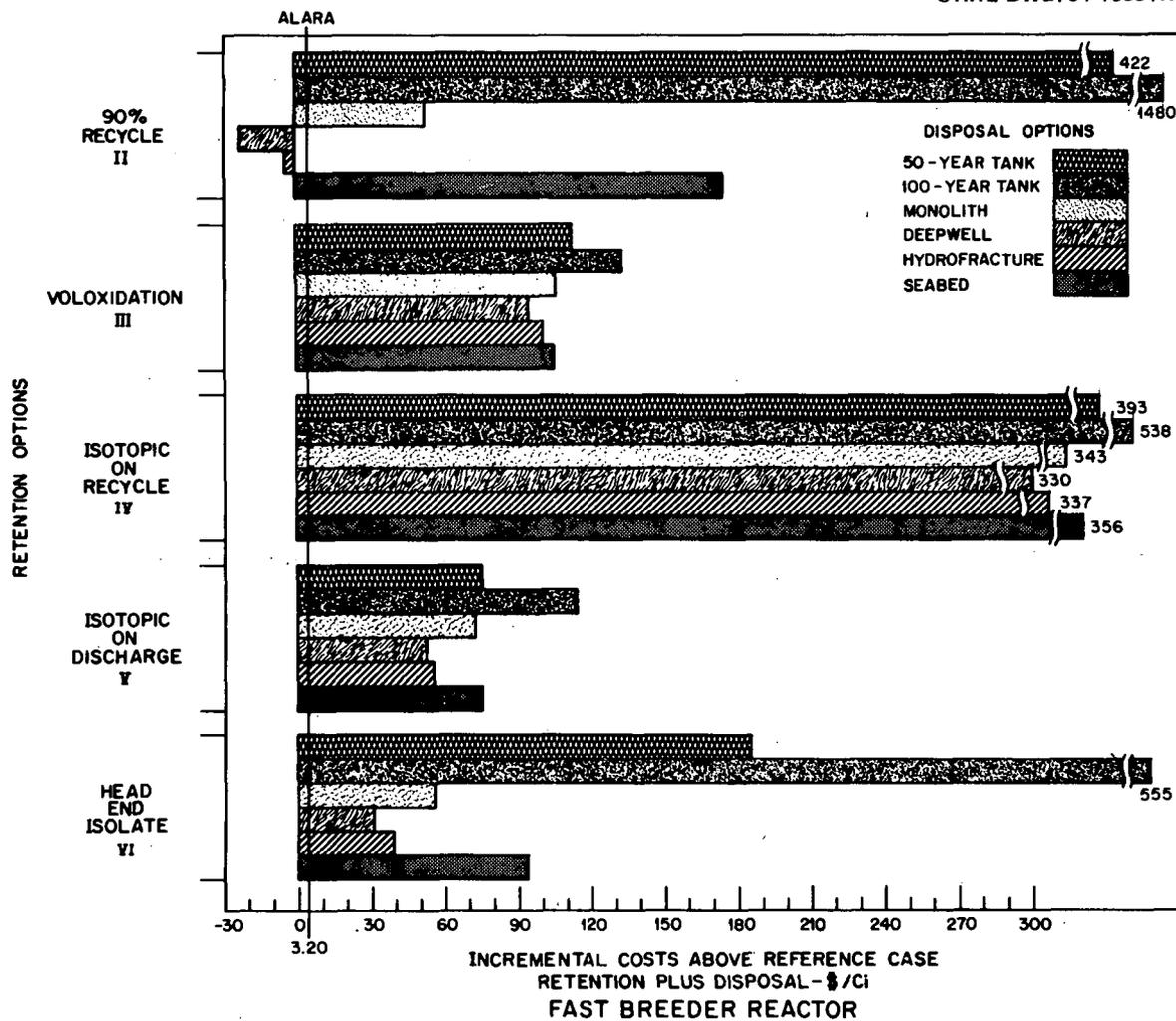


Fig. 1.3. Cost summaries - LMFBR.

Tritiated water vapor released to the atmosphere through the stacks of plants of this study is dispersed in a manner determined first by the local weather and meteorological conditions and subsequently by those conditions of larger areas. Tritiated water is removed from the atmosphere by deposition and (primarily) by precipitation processes. The deposited tritium flows with surface waters and is subject to reevaporation and to run-off to the ocean. The tritiated water mixes with the waters of its hemisphere of origin and remains a part of the hydrological cycle until it decays. The tritium, accordingly, contributes to the radiological dose commitments to individuals, to the population near the plant, and to the population of the much larger area over which the released tritium is dispersed. As a component of the hydrological cycle, it also contributes (at very low dose rates to the average individual) to dose commitments to the population of the world during its decay.

Dose commitments from the tritium released were estimated, using quite conservative assumptions, for four groups of people. These included: (1) individuals who could

theoretically receive the maximum exposure (the individual who lived at the facility fence post), (2) the average "local" individual who lived within 50 miles of the facility, (3) the average "regional" individual who lived in the United States outside the 50-mile range, and (4) global exposures for the rest of the world.

Estimated exposures for the three groups of personnel residing in the United States are summarized in Table 1.3. These doses are expressed as percentages of exposure that an individual would receive from the natural background radiation in the United States. As can be seen, only the LWR and LMFBR base-case options (I) and the LWR recycle option (II) exceed 1% of natural background for the maximum theoretical fence post individual. None of the local or regional doses is an appreciable percentage of natural background.

It is apparent that the LWR base-case plant (with by far the largest tritium release rate) might deliver (with the conservative assumptions used) an annual dose commitment of 20% of natural background or 24 millirem to the maximally exposed hypothetical individual. This maximum dose is below (but not comfortably below) the present Environmental Protection Agency (EPA) requirement that individuals receive less than 25 millirem/year from all portions of the nuclear fuel cycle. It is possible that such dose commitments to individuals may require that not all the tritium be released from the base-case LWR plant or that increasing the stack height and distance to the site boundary would reduce this dose to a lower value. Also site specific values may reduce the calculated dose relative to the conservative assumptions used.

It is, however, apparent that none of the other options appears to offer a significant threat to the maximally exposed individual.

**Table 1.3. Theoretical dose commitments to individuals as a result of tritium releases
(Stated as percent of natural background radiation)**

Tritium retention option	Maximally exposed hypothetical individual (% of background)	Average of local personnel – (50-mile radius; % of background)	Average of regional personnel – (outside 50 miles; % of background)
LWR			
Base	20	0.9	0.02
90% recycle	2.5	0.1	0.002
Voloxidation or isotope separation	1	0.04	0.0008
Head-end isolation	0.3	0.02	0.0004
LMFBR			
Base	3	0.1	0.003
90% recycle	0.3	0.02	0.0003
Voloxidation or isotope separation	0.2	0.01	0.0002
Head-end isolation	0.1	0.006	0.0001

It must be emphasized that even the largest of these population dose commitments are minuscule in comparison with the dose commitments from exposure to the natural background radiation. The local (50-mile) population would receive (in 20 years) a total body dose commitment of 2.9×10^6 person-rem from natural background compared with 2.6×10^4 person-rem from LWR base-case tritium; corresponding figures for the large regional (3.1×10^8) population are 1.35×10^5 person-rem from LWR base-case tritium and 8.0×10^8 person-rem from the natural background.

It is not apparent that a compelling argument can be made for retention of tritium during LWR fuel reprocessing for any personnel other than the "fence post" individual, and then only if the very conservative assumptions are used. Retention of LMFBR tritium will prove more expensive and appears to be even less urgent.

As a consequence of Appendix I to 10 CFR 50,¹ light-water cooled nuclear reactors are required to add equipment for control of radioactive emissions if the resulting decrease in dose commitment from the radionuclides to the population within 50 miles is obtained at an annualized cost of \$1000 or less per total body man-rem and \$1000 or less per man-thyroid rem. Thus the maximum value considered would be \$2000 per man-rem. Release of one curie of tritium from a processing plant at the Oak Ridge site is conservatively estimated to deliver a dose commitment of 1.6×10^{-3} person-rem to the total body and an equal quantity to the thyroid of the 50-mile population. If this criteria is also applied to reprocessing plants, it might be argued that control of tritium could be considered cost effective if it could be achieved for \$3.20 per curie or less. On this basis only the 90% water recycle option (II) combined with deep-well or hydrofracture disposal would be cost effective (see Figs. 1.1 and 1.2).

Two other factors that must be considered in evaluating the need for tritium collection and retention are (1) the effect of release on the world tritium inventory and (2) the effect of recycling high concentrations of tritium in the reprocessing plant.

The total tritium available for release from a 5 t/d LWR reprocessing plant is 8×10^5 Ci/year, which is 20% of the estimated four megacuries that are generated worldwide by natural processes each year. An LMFBR reprocessing facility would have about one-seventh of this quantity available for release; however, as implied in Table 1.3, this quantity could increase the natural background radiation by only very small fractions of a percent on a world-wide basis.

The concentration of tritium in the recycle acid and water for options II, V, and VI is more than an order-of-magnitude higher than for the base-case plants. However, this increased tritium concentration would require no additional radiation protection measures beyond those required because of trace fission products in the recycle water and acid.

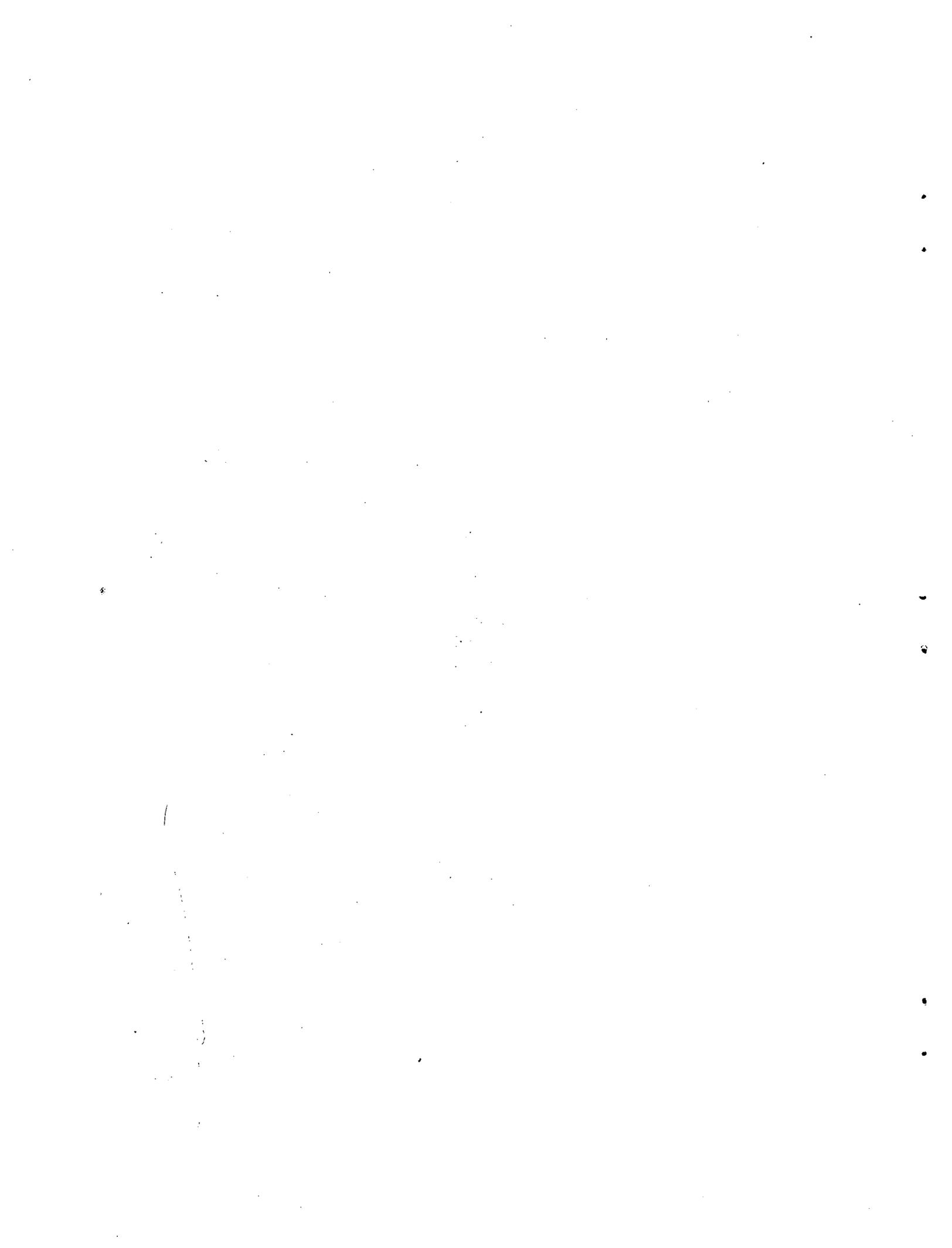
In summary, there appears to be no compelling benefit of tritium retention and disposal for LMFBR reprocessing. The need for tritium retention and disposal for LWR fuel is marginal, except for conservative estimates for the fence post exposure conditions.

If hydrofracture or deep-well disposal is available, this option is recommended for use with water recycle in the facility.

If hydrofracture or deep-well disposal is not recommended, it is recommended that isotope separation on the discharged liquid be further investigated.

REFERENCE

1. U.S. Nuclear Regulatory Commission, *Standards for Protection Against Radiation*, June 15, 1979.



2. INTRODUCTION

2.1 Scope and Objectives of the Study

Several processes that might be used for retention of tritium by plants processing irradiated nuclear fuel from commercial reactors are under study in the United States and elsewhere. As an example, voloxidation has been studied for a number of years at the Oak Ridge National Laboratory as a method for removal of tritium before dissolving the fuel for reprocessing. However, whether tritium retention should be required of fuel reprocessing plants remains an unanswered question at present. This document reexamines tritium retention technologies (including voloxidation) and tritium disposal options to assist in decisions as to the need for tritium isolation and as the choice of appropriate technologies if the need exists.

A detailed assessment of the options for retention of tritium and its ultimate disposal during future reprocessing of oxide fuels from commercial light-water reactors (LWRs) and liquid-metal fast breeder reactors (LMFBRs) was made. This assessment includes an appraisal of the state of the technology, an estimation of the incremental costs (both in capital and operating costs and in increased complexity of plant operation) of the several retention and disposal options, as well as estimation of the dose commitments to the general public and the reduction in those dose commitments resulting from retention and disposal of the tritium.

Two base-case plants, one assumed to process LWR oxide fuel and the other to process LMFBR oxide fuel, were envisioned for this study. These base-case plants (described in more detail in Sect. 3) were each assumed to (1) process 1500 metric tons of heavy metal (MTHM) per 300-d year, (2) discharge no radioactivity in liquid streams, (3) recycle the acid used in the plant but to use minimal recycle of water, and (4) discharge all releasable tritium as tritiated water (HTO) to the atmosphere by vaporization of process water through a single stack 100 m in height. These base-case plants, for which the cost of tritium retention and disposal was assumed to be zero, are each envisioned to be constructed during the 1990's in Oak Ridge, Tennessee, and to operate during the period between the years 2000 and 2020.

Six options have been proposed for retention of major fractions of the releasable tritium by modification of such base-case reprocessing plants. Although many features of the base-case plants remain unchanged, each of these retention options requires some additions, deletions, and modifications of portions of the plants. The retained tritium must obviously be managed or disposed of in a manner that is environmentally acceptable. Several options for such management or disposal have been proposed. A number of these seem feasible, although detailed assessment of their possible impact upon the local environment has seldom been made. The management and disposal options will require additional equipment or facilities to the base-case plants. Incremental costs for the retention and disposal options were evaluated (in first quarter 1980 dollars) by detailed consideration of the required additional equipment, operating personnel, etc., to these base-case plants. Benefits in reduced exposure to the general public are estimated for the hypothetical plants at a specific site at Oak Ridge, Tennessee.

2.2 Applicable Regulations for Reprocessing Plants

The allowable concentration of tritium in water leaving the site has been set at 3 mCi/m³ and of air at 0.2 μ Ci/m³ (10 CFR 20).¹ However, there are no established criteria pertaining specifically to tritium in which to base decisions as to the necessity or the desirability of retention and disposal of this isotope. Moreover, it is obviously not possible at present to divine the regulatory and licensing rules and criteria that may be in effect 15 to 20 years hence.

2.2.1 Limits on Dose to Members of the Public

The Nuclear Regulatory Commission (NRC) subscribes fully to the principle that radiation exposures to the public from nuclear facilities should be kept as low as is reasonably achievable (ALARA). The NRC has given (Appendix I of 10 CFR 20) a quantitative definition of ALARA dose levels for routine emissions from light-water-cooled nuclear reactors, but it has issued no such quantitative guidance for fuel reprocessing plants. Accordingly, the NRC limit for doses to the general public from reprocessing plants is, presumably, that set by 10 CFR 20; by this guide the limit is 500 millirem/year to the total body. These guides and recommendations apply to exposures from all sources other than medical procedures and the natural background. It has been argued that the NRC limit of 500 millirem/year to individuals and of the population at large is too high, but no one denies that the nuclear industry has kept doses to the public at small fractions of this value.

The Environmental Protection Agency (EPA) has adopted a rule-making action (40 CFR 190) that applies to several parts of the uranium fuel cycle associated with light-water reactors. By this rule, exposures to (1) planned discharges of radioactive materials (except radon and its daughters) and (2) direct radiation from uranium fuel cycle operations should not result in annual dose commitments in excess of 25 millirem to the whole body and to any organ except the thyroid or in excess of 75 millirem to the thyroid of any members of the public. These limits would, presumably, be the maximum allowable for the processing plant, assuming that the individuals receiving the maximum exposure were affected by no other nuclear facility. It is presumed that these rules will be extended to apply to the breeder reactor fuel cycle also.

2.2.2 Limits on Doses to Plant Employees

The limits of occupational dose are set (10 CFR 20) at 5 rem/year, with additional restrictions for the allowable dose in any quarter of the year.

2.2.3 Other Regulations that May be Applicable

The limit (set by 10 CFR 20) on tritium concentration in water is 3 mCi/m³ (3 μ Ci/L). Otherwise, and like the situation for tritium releases from nuclear facilities, there are no established guidelines for disposal of wastes containing tritium. However, the currently proposed NRC regulation (to be designated as 10 CFR 61) would permit up to 10⁸ Ci/m³ of tritium to be buried in a stabilized form in near-surface burial sites. The NRC has proposed² a system for classification of radioactive wastes within specific disposal guidelines. Table 2.1 shows the concentrations of tritium (if it were the only radioisotope present) that would fit each of these proposed disposal classes.

Table 2.1. Disposal concentration guide for tritium

Tritium concentration (Ci/m ³)	Waste disposal category	Description
5×10^{-2}	E	No administrative control; worker/reclaimer access (unrestricted release)
94	D	Administrative control for 150 years, followed by reclaimer access (surface burial)
94	C	No administrative control; no reclaimer access except well water (intermediate-depth burial)
430 000	B	Administrative control for 150 years, followed by no reclaimer access except well water (intermediate-depth burial)
2.9×10^9	A	Isolation (geologic repository)

Source: V. C. Rogers, *A Radioactive Waste Disposal Classification System*, NUREG/CR-1005 U.S. Nuclear Regulatory Commission Report, Washington, D.C. (September 1979).

It will become apparent (Sects. 3 and 4) that none of the effluents or concentrates from the plants of this study have tritium concentrations sufficiently low to qualify for category E disposal. In addition, none of the tritiated waters have tritium concentrations sufficiently high to require category A (isolation in a geologic repository). As a consequence, the geologic repository seems not to be a required disposal mode and, presumably, would not be used unless it were less expensive than competing modes.

Finally, it should be noted that these classifications are only *proposed* and may or may not be adopted as applicable to tritiated wastes.

2.3 Origin and Quantities of Tritium to be Considered

2.3.1 Natural Tritium

Tritium is continuously generated by natural processes and occurs naturally in the earth's surface waters. Spontaneous fission of uranium and thorium contributes trivial quantities of this isotope. Interaction of high-energy cosmic rays with oxygen and nitrogen atoms in the upper atmosphere is believed to be responsible for production of about 1.6×10^6 Ci/year; this production rate would lead to a steady-state inventory³ of 28×10^6 Ci. A very similar estimate (26×10^6 Ci) of the steady-state inventory caused by cosmic ray interactions has recently been presented.⁴ Measurements of tritium concentrations in the earth's waters, however, suggest a considerably larger inventory of tritium.⁵ Solar flares^{6,7} may be responsible for much of the earth's total natural tritium. It seems to be generally accepted⁴ that the earth's inventory of natural tritium is near 70×10^6 Ci; this corresponds to an appearance rate of about 4×10^6 Ci/year.

2.3.2 Tritium in Reactor Fuels

Tritium is formed within nuclear reactor fuels (both LWR and LMFBR) by ternary fission and may also be formed there by neutron bombardment of light elements (such as boron or lithium) if they are present within the fuel. Additional tritium is generated in the reactor through interaction of neutrons with some materials (of which lithium and boron are the most important) used for control of excess reactivity and for control of pH. Such tritium, generated outside the fuel element of an LWR or LMFBR, is of some environmental concern for the particular reactor, but it is not delivered to the reprocessing plant and does not concern the present study.

2.3.2.1 Tritium in LWR oxide fuels

It seems clear that little, if any, of the tritium generated within the fuel of the modern LWR is lost via diffusion *through* the zirconium cladding.⁸ Some small quantity (perhaps 0.1 to 1% of that produced) may be lost at such a reactor by leakage through cladding defects.^{9,10} The ORIGEN code,¹¹ often used for estimating isotope generation in long-term fuel cycle projections, predicts that ternary fission will be responsible for the generation of 540 Ci of tritium per MTHM irradiated at an average specific power of 30 MW/MTHM to a burnup of 33 000 MWd/MTHM. An older study¹² assumes that LWR fuel contained some 800 Ci of tritium per metric ton. NUREG-0002¹³ and Finney et al.¹⁴ assume that LWR fuel fed to the reprocessing plant after a 160-d cooling period contained 515 Ci of tritium per metric ton. Finney et al.¹⁴ note that each 0.1 ppm of natural lithium within the fuel would contribute an additional 26 Ci of tritium per metric ton.

Although essentially no tritium escapes through the zircaloy cladding, a portion diffuses into the zircaloy and is immobilized as zirconium tritide. The quantity so immobilized, which is not available for release on dissolution of the oxide fuel, is a function of operating parameters and cannot be said to be surely known. Goode and Vaughen⁸ found 13% of the total tritium within the zirconium cladding of a commercial pressurized water reactor (PWR) fuel irradiated to about 40 000 MWd/t.

For purposes of this study, we have assumed that the LWR oxide fuel delivered to the reprocessing plant will contain 600 Ci of tritium per MTHM. Of this total, 10% (60 Ci/MTHM) is assumed to be retained within the cladding; the remainder (540 Ci/MTHM) is assumed to reside within the fuel oxide and to be available for release.

As a consequence, some 9×10^5 Ci of tritium are delivered each year to the LWR reprocessing plant and, of this, some 8.1×10^5 Ci/year could be released. It is clear that, given total release of the 8.1×10^5 Ci/year, five such plants would release tritium corresponding to the natural appearance rate. These five plants would process fuel from about 250 reactors, each of 1000 MW(e) generating capacity.

2.3.2.2 Tritium in LMFBR oxide fuels

The amount of tritium that will be produced during operation of a large LMFBR is not well known both because the fast fission yields of tritium are not well established and because design details for large LMFBRs are not fully developed. Moreover, a large fraction of the tritium generated in the core fuel is expected to diffuse through the stainless steel fuel cladding and be trapped as NaT in the sodium coolant circuits.¹⁵ Tritium trapped

in this way must be managed by the reactor, but it is not of concern to the reprocessing plant.

Tritium will arise in the LMFBR fuel both by ternary fission and by activation of impurities (boron and lithium) in the core and blanket oxides. The quantities expected to be *produced* have been estimated by several authors. The quantities expected to be delivered to the processing plant are addressed subsequently.

Table 2.2 shows several recent estimates of the quantity expected to be produced by ternary fission in the total core and blanket system. There are almost certainly differences in assumptions about the reactor systems chosen for the various estimates. Moreover, the source documents give the expected yields in different units that have been converted to Ci/MTHM in Table 2.2.

Table 2.2. Tritium production from ternary fission in blended LMFBR core and blanket fuel

Tritium produced (Ci/MTHM)	Source document
1010	<i>Proposed Final Environmental Impact Statement: Liquid Metal Fast Breeder Reactor Program</i> , Vol. II, WASH-1535, U.S. Atomic Energy Commission, Washington, D.C. (December 1974).
545 to 1090	L. E. Trevorrow et al., <i>Tritium and Noble-Gas Fission Products in the Nuclear Fuel Cycle: I. Reactors</i> , ANL-8102, Argonne National Laboratory (October 1974).
490	V. J. Tennery et al., <i>Environmental Assessment of LMFBR Advanced Fuels: A Radiological Analysis of Fuel Reprocessing, Refabrication, and Transportation</i> , ORNL-5320 (November 1976).
815 to 1000	H. Bonka, "Production and Emission of Tritium from Nuclear Facilities, and the Resulting Problems," <i>Proceedings</i> , International Symposium on Behavior of Tritium in the Environment, San Francisco, October 16-20, 1978; International Atomic Energy Agency, Vienna, 1979.
1070	<i>Tritium in the Environment</i> , Report No. 62, National Council on Radiation Protection and Measurements, Washington, D.C. (March 1979).

Trevorrow et al.¹⁶ have estimated that 10 ppm of natural lithium in the core and blanket oxide of a 1000 MW(e) LMFBR would be responsible for about 390, 790, and 290 Ci per 1000 MW(e) year in the core, radial blanket, and axial blanket respectively. Blending of the core and blanket oxides would yield a material containing about 38 Ci/MTHM, with 49% of the lithium-produced tritium in the blanket material. Bonka¹⁷ presents an estimate of tritium resulting from ⁶Li in the oxide materials that seems to be in reasonable agreement.

Trevorrow et al.¹⁶ have also estimated that 10 ppm of boron in the fuel material would yield 370, 200, and 40 Ci/GW(e) within the core, radial blanket, and axial blanket

respectively. If 10 ppm of boron could not be avoided, 16 Ci of tritium would be produced per metric ton of blended material, with 40% of this produced within the blanket.

It seems reasonable to assume that lithium content can be kept to 2 ppm and boron to 1 ppm. If so, the core and the blanket oxides would contribute tritium to the blended material in the quantities shown in Table 2.3.

Table 2.3. Tritium generated from lithium and boron in LMFBR oxide materials^a

Source	Tritium (Ci/MTHM of blend)		Total
	Lithium	Boron	
Core	3.9	0.96	4.9
Blanket	3.7	0.64	4.3
Total	7.6	1.6	9.2

^a Assumes 2 ppm of lithium and 1 ppm of boron in the fuel and blanket oxides.

In addition, it seems conservative to assume that the tritium produced by ternary fission will total 1090 Ci/MTHM for the blended core and blanket oxides (the core material responsible for 1035 Ci/MTHM and the combined radial and axial blankets responsible for 55 Ci/MTHM). Adding the (trivial) contributions from the lithium and boron activation from Table 2.3 suggests 1040 Ci/MTHM generated in the core and 60 Ci/MTHM generated in the combined radial and axial blanket. These estimates would seem to be conservative (perhaps by as much as a factor of 2) unless the recent indications^{18,19} of markedly higher fast fission yields of tritium from ²³⁸U and ²³⁹Pu prove to be correct.

The quantity of interest to this study is, of course, the quantity of tritium that is delivered to the reprocessing plant. Again, there is no certain knowledge of the real situation, and several authors have made different assumptions:

1. Ninety percent of the tritium diffused from the combined fuel and blanket was assumed, and 101 Ci/MTHM of tritium delivered to the processing plant was estimated;¹⁵
2. Tennery et al.^{20,21} (who estimated lower production values) accepted the retention of 10% of the tritium by the combined core and blanket materials;
3. Bonka¹⁷ seems to have assumed that the blended fuel would retain 50% of the generated tritium;
4. No number (or percentage) is given but information⁴ suggests that "only small quantities are expected to be transferred with the fuel to the reprocessing plant.

Nicholson,²² in an evaluation of the voloxidation process, concluded, after a survey of the literature, that some 3% of the tritium generated within the core might be retained by the core fuel, which operates at a high temperature. He also suggests that the axial

blanket might retain a large fraction of its tritium and that the radial blanket (whose contribution is not large, but whose temperature is higher) would probably lose a large fraction of its tritium via diffusion through its stainless steel cladding.

For this study, it has been assumed that the core fuel retains 3% of the tritium generated therein and that the radial and axial blankets each retain 90% of that generated within them. As a consequence, the blend of core and blanket materials will contain 85 Ci of tritium per MTHM. Accordingly, a plant processing 1500 MTHM/year would receive 1.275×10^5 Ci/year of tritium. All of this tritium would be contained in the oxide, and essentially none would reside in the stainless steel cladding. If all of the tritium were released to the atmosphere, some 30 such reprocessing plants (servicing some 1200 LMFBRs) would be required to equal the appearance rate of naturally produced tritium.

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3. CHARACTERISTICS OF BASE-CASE REPROCESSING PLANTS

Two base-case plants, one to process LWR fuel (case LWR-I) and one to process LMFBR fuel (case LMFBR-I), were envisioned for this study. Each of these plants was assumed (as were all other plants of this study) to use variants of the Purex process and to reprocess 1500 metric tons of heavy metal (MTHM) per 300-d year.

The somewhat simplified flowsheets, including process fluid flows and tritium balances, are based on such 300-d operation at 5 MTHM/d. It is important to note, however, that to ensure the capability for processing 1500 MTHM/year, each feature and equipment item of the base-case (and of all other) plants in this study are assumed to be designed and sized to operate at an 8-MTHM/d rate.

All plants were assumed to have an operating lifetime of 20 years and to operate between the years 2000 and 2020. Each plant, including LWR-I and LMFBR-I, was assumed to discharge no radioactivity in aqueous effluents to the environment and to meet applicable regulations (see Sect. 2). All plants were assumed to include facilities for solidifying high-level wastes, and high-activity waste (HAW) solutions were assumed to be evaporated and calcined promptly. All LWR processing plants (cases LWR-I through LWR-VI) were assumed to receive 600 Ci of tritium per MTHM, with 60 Ci/MTHM immobilized within the fuel cladding and 540 Ci/MTHM available for release. All LMFBR plants (cases LMFBR-I through LMFBR-VI) were assumed to receive 85 Ci of tritium per MTHM in the fuel, with all of this available for release (see Sect. 2).

The base-case plants (LWR-I and LMFBR-I) were assumed to recycle plant acid but to use no recycle of process water and to discharge all releasable tritium (as HTO) to the atmosphere by vaporization of process water through a single stack 100 m high. The cost for tritium retention and disposal for these base-case plants was assumed to be zero; costs of the tritium retention and disposal options were evaluated (in first quarter 1980 dollars) for each other case based on equipment, maintenance, and operating costs incremental to those of the base-case plants.

3.1 Site Characteristics

Each of the several hypothetical plants was assumed to be located within a large exclusion area near Oak Ridge, Tennessee. This site, which has been proposed for Exxon Nuclear Company's Nuclear Fuel Recovery and Recycling Center, has been described in detail elsewhere¹.

The site area is roughly rectangular and measures approximately 2 miles north-south by 2.5 miles east-west. It contains about 2500 heavily wooded acres. The approximate center of the process building for each plant lies at latitude 35°55'40"N and longitude 84°21'51"W. This 2500-acre exclusion area (as the term is used in 10 CFR 100) is within the Oak Ridge Reservation of the U.S. Department of Energy and lies in Roane County, Tennessee, with the Clinch River about one mile from the boundary to the south and about one mile from the boundary to the southwest.

The city center of Oak Ridge, Tennessee, is about 8 miles northeast of the site, and the city of Knoxville, Tennessee, is about 25 miles to the east. Details of the population distribution to be expected in the period from year 2000 to 2020 are presented in Appendix D, along with details of the local meteorology.

3.2 Plant Characteristics

3.2.1 For LWR Fuel Processing

The base-case plant for LWR fuels (case LWR-I) contains some process features of the Allied-General Nuclear Services (AGNS) reprocessing plant at Barnwell, South Carolina. Figure 3.1 shows a simplified flowsheet for operating at 5 MTHM/d; water balances are shown in liters per hour and tritium balances (the circled numbers) in curies per day. The flowsheet assumes that all recycle process water is sent to a vaporizer and is discharged to the stack as water vapor. (Details of assumptions relative to the stack are presented in Appendix D.) Daily tritium releases (2700 Ci) represent the total present in the 5 t of fuel processed each day, less that (300 Ci) entrapped in the cladding hulls.

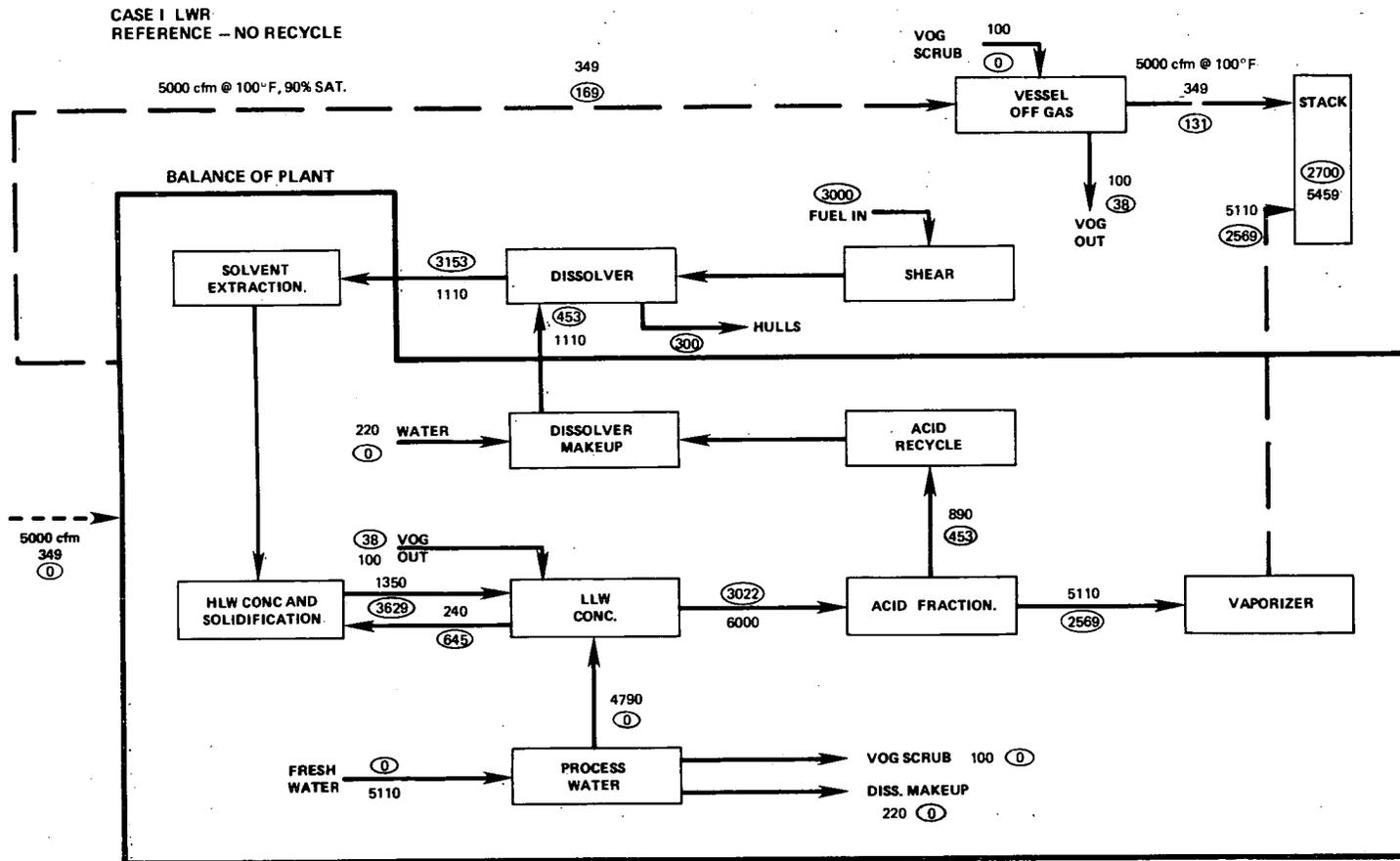
3.2.2 For LMFBR Fuel Processing

The base-case plant for processing LMFBR fuel (LMFBR-I) was adapted from the AGNS-plant block flows and from analysis of Exxon Nuclear Company's study and report on LMFBR fuel reprocessing.² A simplified flowsheet for operating on 5 MTHM/d is shown in Fig. 3.2, with water balances in liters per hour and tritium balances (circled figures) in curies per day. The flowsheet assumes that none of the tritium (425 Ci/d) will be immobilized by the stainless steel fuel cladding, but instead, will be released to the stack as HTO in water vapor. The LMFBR flowsheet shows that twice the amount of water is required for reprocessing LMFBR fuel than for LWR fuel. Aqueous streams in both flowsheets (LMFBR and LWR) will consist of approximately the same fissile quantity (in grams per liter). The LMFBR flowsheet will require larger and more expensive equipment.

3.2.3 Assumptions for Base-Case Plants

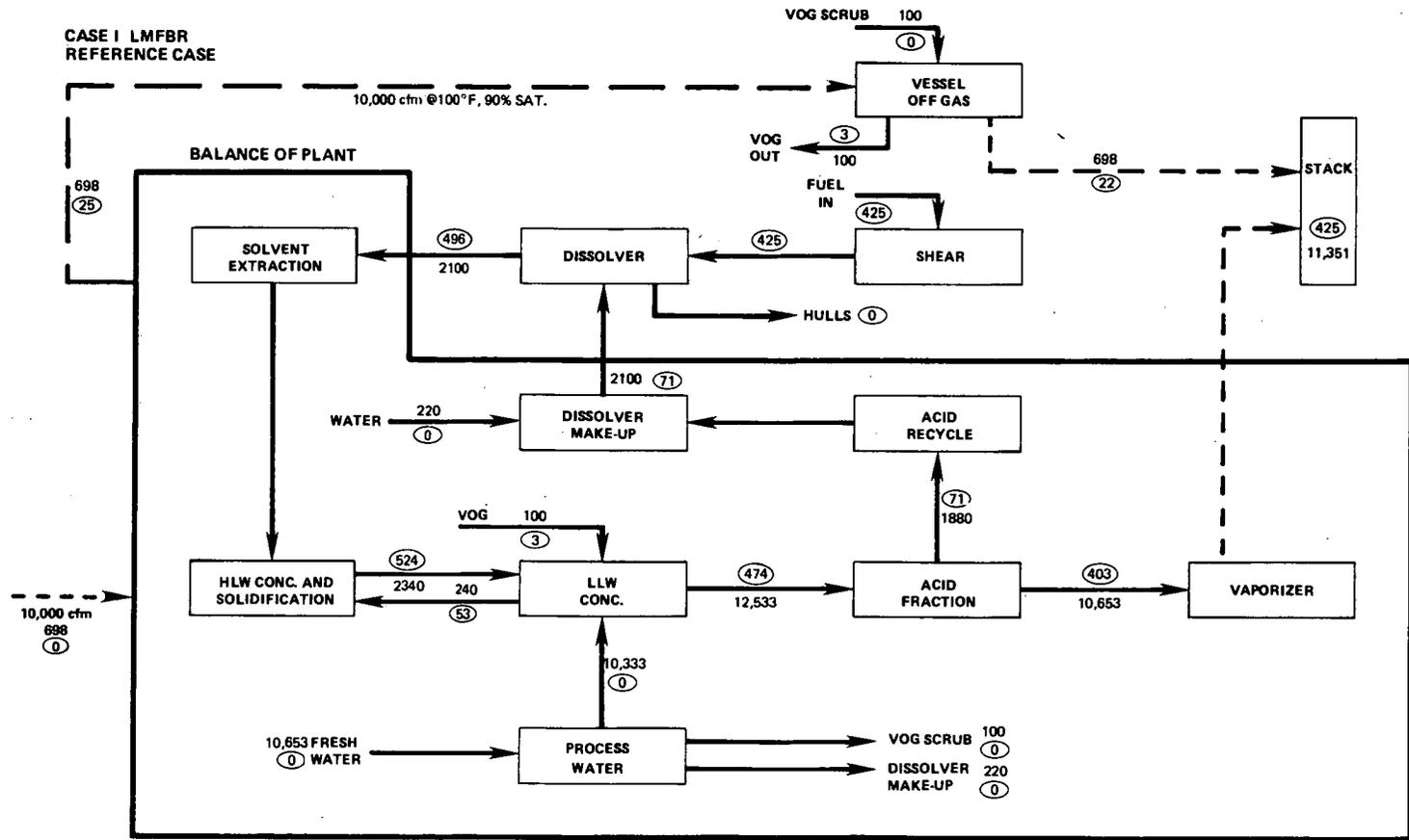
The following assumptions were used for both the LWR and the LMFBR base-case plants (LMFBR values are within parentheses).

1. Dissolver makeup consists of 890 (1880) L of recycle acid and 220 (220) L of hull rinse (process water) per hour.
2. Off-gas system: flow rate taken as 5000 cfm (10 000 cfm) at 90% saturation and 100°F; off-gas water content is 349 (698) L/h.
3. Tritium content in vessel off-gas based on tritium content in process vessels; vapor assumed to be in equilibrium with acid fractionator liquid composition.
4. Vessel off-gas scrubber uses 100 L of clean water per hour; tritium concentration in scrub effluent is in equilibrium with tritium content in outlet air stream.
5. Tritium losses to organic liquids are not shown, but are accounted for in the balance of the plant.
6. Other stream volumes are held constant (all cases).
 - Dissolver product and solvent extraction waste stream, 1100 (2100) L/h;
 - High activity waste concentration and solidification overheads, 1350 (2340) L/h;
 - Low activity waste bottoms, 240 (240) L/h;
 - Low activity waste overheads, 6000 (12 533) L/h;
 - Acid fractionator: bottoms, 890 (1880) L/h; overheads, 5110 (10 653) L/h.



Circled numbers: Curies of tritium per day.
Uncircled numbers: Liters of water per hour.

Fig. 3.1. Case I - LWR reference case - no recycle.



Circled numbers: Curies of tritium per day.
 Uncircled numbers: Liters of water per hour.

Fig. 3.2. Case I - LMFB reference case.

Table 3.1 shows a comparison of water discharge rates and tritium concentrations for the two base cases.

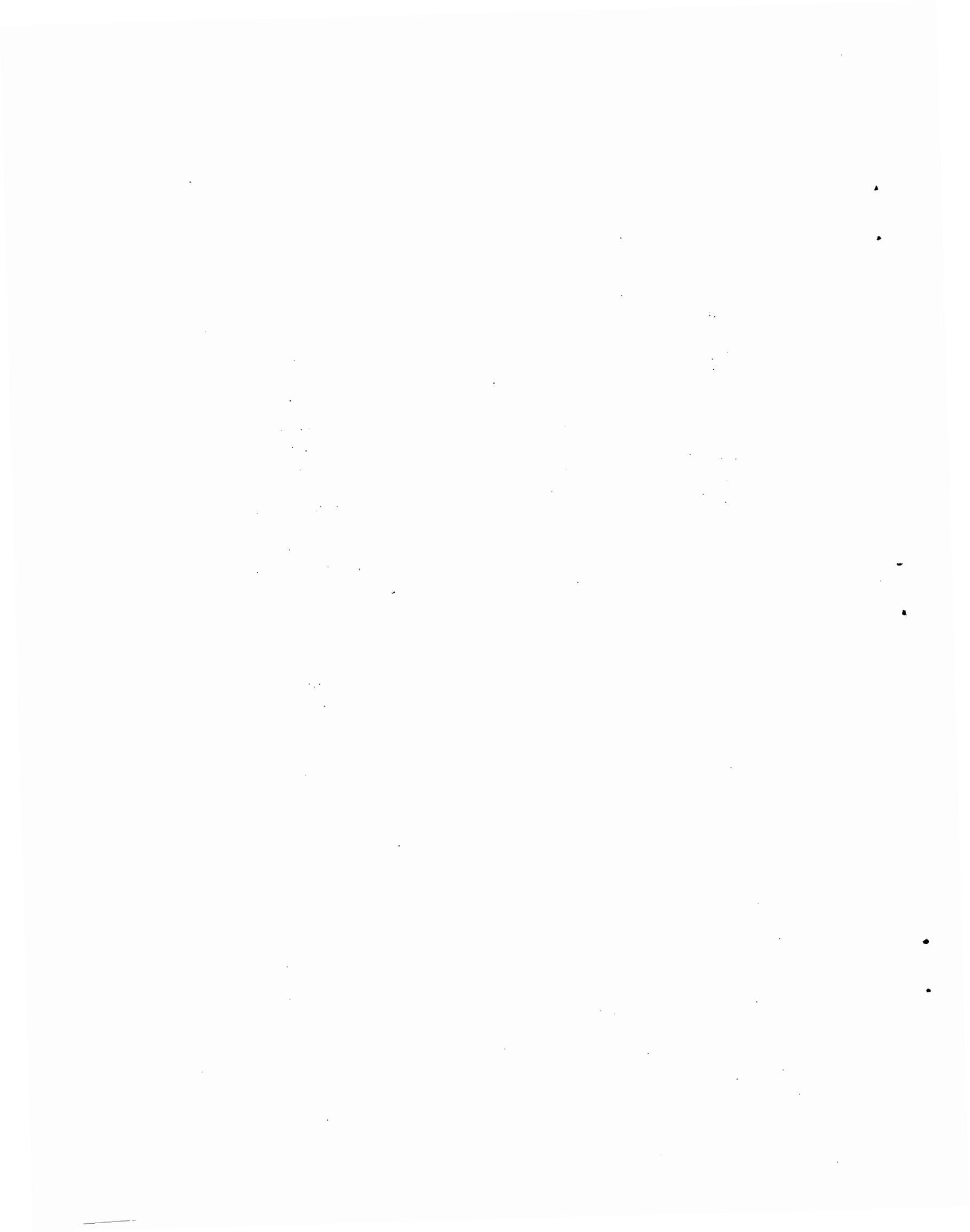
Table 3.1. Water discharge rates and tritium concentrations for base-case plants (no water recycle)

Disposition	Case	
	LWR-I	LMFBR-I
Discharge to stack		
H ₂ O, L/h	5 459	11 350
H ₂ O, m ³ /year	39 300	81 700
³ H, Ci/year	8.1 × 10 ⁵	1.28 × 10 ⁵
³ H, Ci/m ³	20.6	1.57
Retained for disposal		
H ₂ O, L/h	0	0
H ₂ O, m ³ /year	0	0
³ H, Ci/year	0	0
³ H, Ci/m ³	0	0
In recycle acid		
Tritium, Ci/m ³	21.2	1.6
In process water		
Tritium, Ci/m ³	0	0
In vessel off-gas water		
Tritium, Ci/m ³	15.6	1.3

The base-case plants discharge larger quantities of tritium to the environment than do others (see Sect. 4.1) that have provisions for retaining this isotope. The base-case plants are the largest consumers of water, since all other plants in this study include recycle of a large fraction (about 90%) of the plant process water. However, the base-case plants use fresh makeup water and therefore send very little tritium or traces of fission products through the chemical makeup areas. Only the recycle acid will contain tritium or trace fission products. Because of their low-tritium and fission-product content, the base-case plants present the smallest opportunity for exposure of plant personnel.

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4. OPTIONS FOR RETENTION AND DISPOSAL OF TRITIUM

When light-water reactor (LWR) fuel is sent to be reprocessed, a very large fraction of the generated tritium from the reactor will be delivered with the fuel to the reprocessing plant. The liquid-metal fast breeder reactor (LMFBR), whose fuel must be reprocessed if such reactors are to be deployed, will send only a small fraction of its generated tritium to a fuel reprocessing facility (see Sect. 2). These fuel reprocessing plants are the only fuel cycle facilities, except for the reactors themselves, that will handle tritium in appreciable quantities.

Neither retention of tritium nor the management or controlled disposal of this isotope is a present requirement at reactors or fuel reprocessing plants. All reprocessing plants presently in operation disperse the releasable tritium (largely as tritiated water vapor) to the atmosphere. It is possible, however, that retention and disposal of this tritium may be required in the future. This study has examined the state of the art, the probable feasibility, and the approximate incremental cost of several options for retention and for management and disposal of tritium at such fuel reprocessing plants.

Several options have been proposed for retention of tritium in variants of plants using the Purex process for reprocessing oxide fuels from LWRs and LMFBRs (see Sect. 3). In principle at least, an even larger set of options exist for storage or disposal of the tritiated waters that might be retained by the plants. The options for retention of tritium that were considered in this study are examined in detail in Sect. 4.1; options for management and disposal of the retained tritium are subsequently examined in Sect. 4.2.

4.1 Options for Retention of Tritium

Table 4.1 identifies and details some general features of the tritium retention options examined in this study of separate plants processing oxide fuels from LWRs and LMFBRs. It

Table 4.1. Some characteristics of tritium retention options^a

Case	Tritium to dissolver solution (% of total)	Tritium concentration methods		Disposition of tritium		Volume of concentrate (m ³ /year) ^b
		Water recycle (% of total)	Additional method for concentration	In concentrate in solution (% of total)	Dispersed to atmosphere in solution (% of total)	
LWR-I and LMFBR-I	100	0	None	0	100	
LWR-II and LMFBR-II	100	90	None	87.5	12.5	3880
LWR-III and LMFBR-III	4	90	Fuel voloxidation ^c	95.3/93.4	4.7/6.6	14.4
LWR-IVA and LMFBR-IV	100	90	Isotope separation ^d	95.3/93.4	4.7	900
LWR-IVB	100	90	Isotope separation ^d	99.5	0.48	900
LWR-VA and LMFBR-V	100	90	Isotope separation ^e	95.3/93.4	4.7/6.6	86.4
LWR-VB	100	90	Isotope separation ^e	99.5	0.49	86.4
LWR-VI and LMFBR-VI	100	90	First-cycle confinement ^f	97.8	2.2/4.0	2660

^aSee text for more detailed descriptions.

^bVolumes shown are, in all cases, those for plants processing LWR fuel. Those from processing LMFBR fuel differ substantially in a few cases; these values are presented in subsequent portions of this section.

^cA large fraction of the tritium is volatilized and collected during oxidation of fuel material before dissolution.

^dIsotope separation is applied to the entire water recycle stream.

^eIsotope separation is applied only to excess water for discharge to disposal and dispersal systems.

^fTritium is confined to the first (high-activity) cycle, with balance of plant waters containing tritium at low concentrations.

is apparent from Table 4.1 that none of the retention options completely eliminates dispersal of tritium to the atmosphere.

The base-case plants (LWR-I and LMFBR-I, described in Sect. 3) disperse all the dissolved tritium as tritiated water to the atmosphere. It is clear that smaller volumes of tritiated water for disposal are obtained if ~90% of the plant water is recycled and tritium is, as a consequence, permitted to build up to higher concentrations in the plant solutions (cases LWR-II and LMFBR-II). Very small volumes of tritiated water are obtained for disposal if (cases LWR-III and LMFBR-III) a large fraction of the tritium is removed by oxidation of the fuel (voloxidation) prior to its contact with the plant dissolver solutions.

If the tritium is introduced into the dissolver solutions, isotope separations, in addition to water recycle, can be employed to produce relatively small volumes of concentrate for disposal. Such isotope separations can be operated in at least two modes. In the first mode (cases LWR-IVA, LWR-IVB, and LMFBR-IV), the entire water recycle stream is passed through the isotope separator. In the second mode (cases LWR-VA, LWR-VB, and LMFBR-V), the isotope separation is used only on the excess water to be discharged.

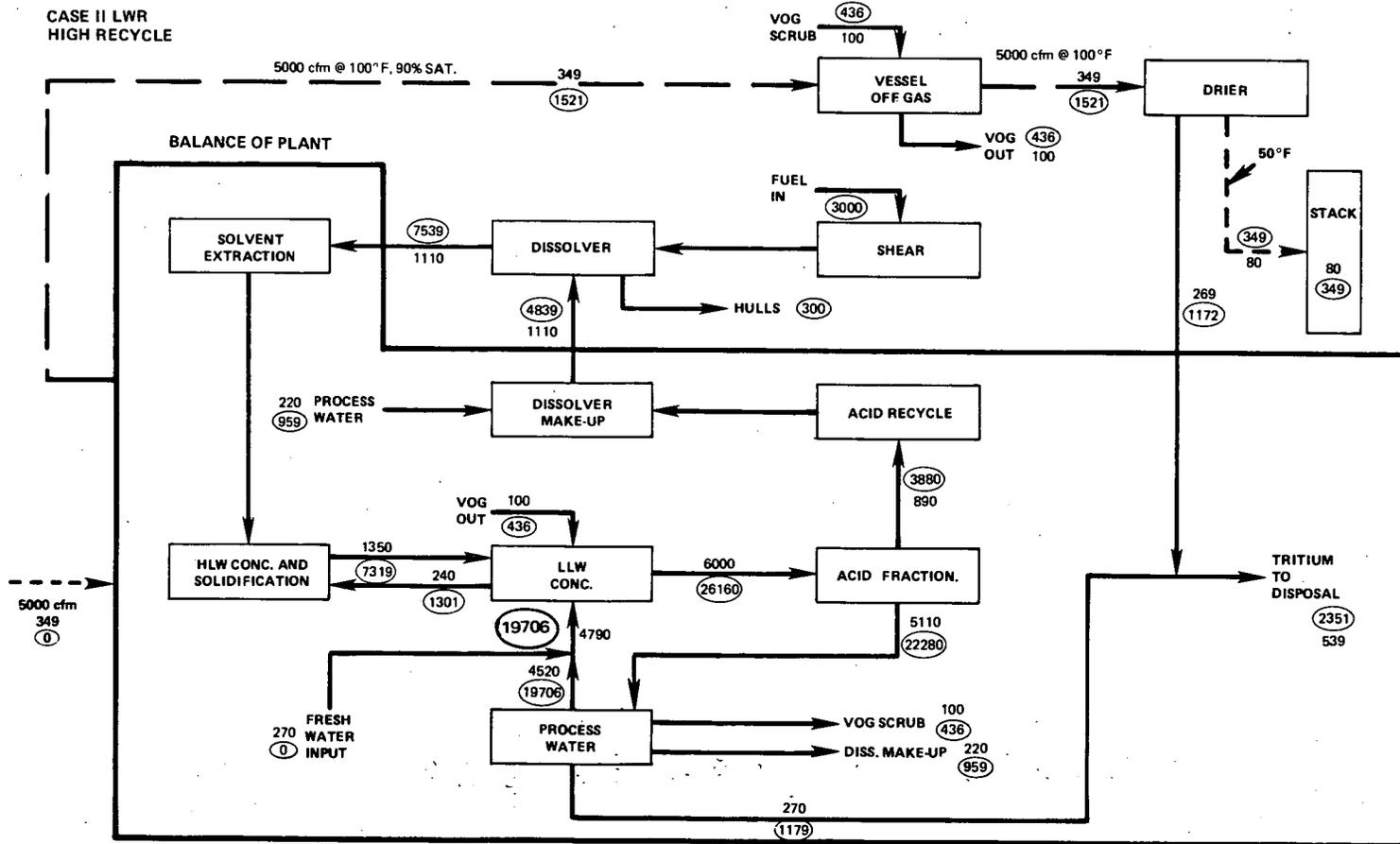
In addition, an attempt can be made (cases LWR-VI and LMFBR-VI) to confine the dissolved tritium to the waters of the first extraction cycle with the rest of the plant waters, which contain relatively little tritium. This alternative can provide a reasonably concentrated tritium solution for disposal from the condensate of the first-cycle waters.

These options, all of which have received some study, are described in the following portions of this section. It must be emphasized that none of these options (other than LWR-I) has been practiced in operating reprocessing plants; none of the several options has been optimized for the particular application; and, given the present state of the art, it is far from certain that such optimization is possible. Each option has been examined objectively and has been evaluated on a comparable basis for its feasibility, complexity, and relative incremental cost.

4.1.1 Cases LWR-II and LMFBR-II: High Water Recycle

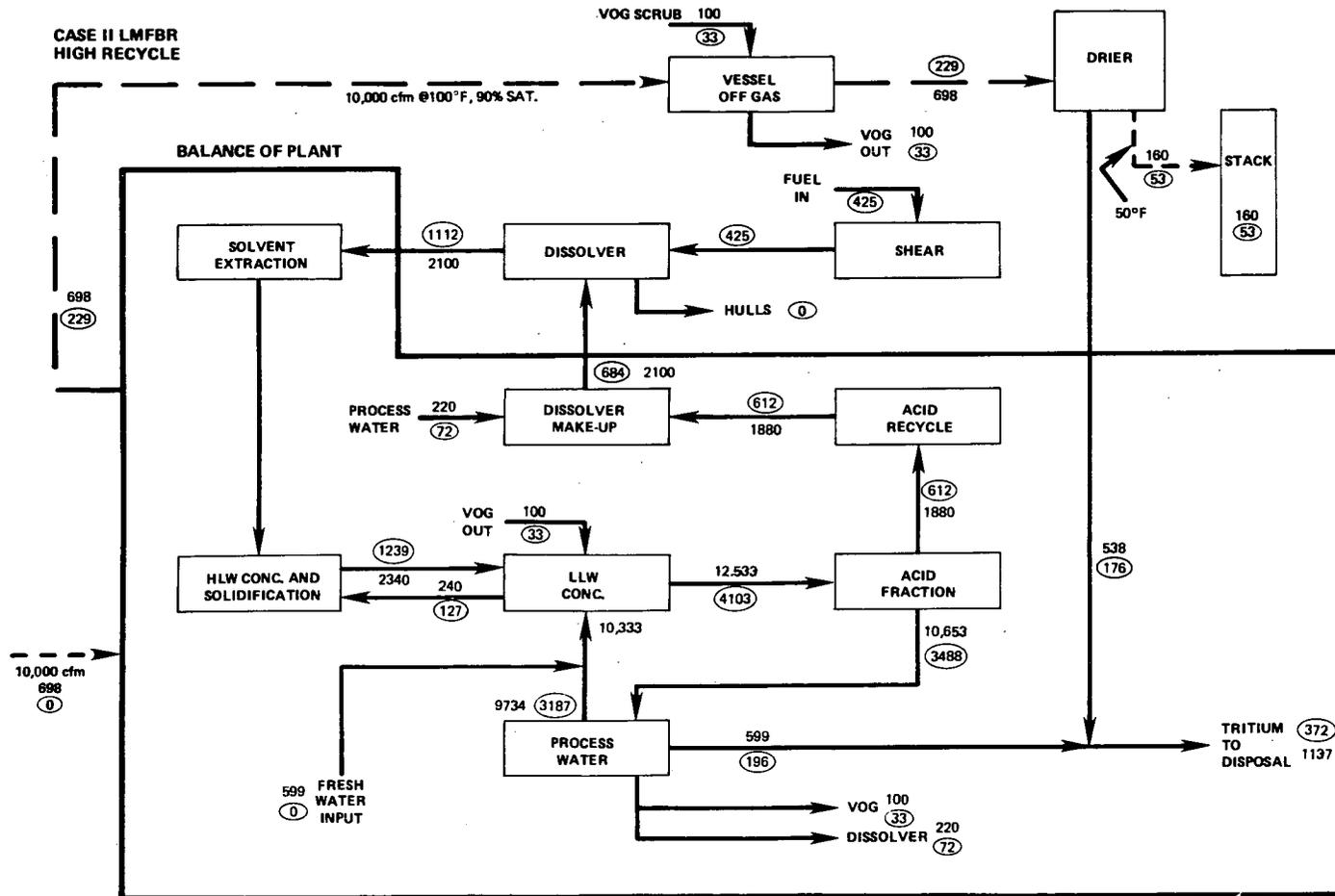
These plants differ from the base-case plants (see Sect. 3) in that process water is recycled in a manner to minimize to a practicable level the fresh water required for plant operations. In both LWR and LMFBR plants, ~90% of the water is recycled, and the excess tritium-bearing process water is routed to storage or disposal (see Sect. 4.2). The simplified flowsheets for these cases are shown in Figs. 4.1 and 4.2. The flowsheets were prepared using the assumptions detailed for the base-case plants (cases LWR-I and LMFBR-I) in Sect. 3. Water discharge rates and tritium concentrations in the stack discharges (in the water retained for disposal), and in the process streams are shown in Table 4.2.

LWR and LMFBR plants employing this manner of recycle discharge only 349 and 53 curies of tritium per day, respectively, to the atmosphere, but they furnish relatively large volumes of tritiated water for management or disposal. They obviously use far less fresh water than do the base-case plants (cases LWR-I and LMFBR-I). However, since they recycle process water along with acid, they send (in common with several other options described below) reasonably concentrated tritium solutions (182 Ci/m^3 from LWR and 13.8 Ci/m^3 from LMFBR fuels) to the chemical make-up areas. Careful design of the plants is vital to ensure protection of the plant personnel. Such safe design and operation would seem feasible, and LWR processing plants employing such recycle have been designed.¹



Circled numbers: Curies of tritium per day.
 Uncircled numbers: Liters of water per hour.

Fig. 4.1. Case II - LWR high recycle.



Circled numbers: Curies of tritium per day.
 Uncircled numbers: Liters of water per hour.

Fig. 4.2. Case II - LMFBR high recycle.

Table 4.2. Water discharge rates and tritium concentrations for plants with water recycle

Discharge conditions	Case	
	LWR-II	LMFBR-II
Discharge to stack		
H ₂ O, L/h	80	160
H ₂ O, m ³ /year	576	1150
Tritium, Ci/year	1.05 × 10 ⁵	1.59 × 10 ⁴
Tritium, Ci/m ³	182	13.8
Retained for disposal		
H ₂ O, L/h	539	1137
H ₂ O, m ³ /year	3880	8190
Tritium, Ci/year	7.05 × 10 ⁵	1.12 × 10 ⁵
Tritium, Ci/m ³	182	13.8
Recycle acid		
Tritium, Ci/m ³	182	13.8
Process water		
Tritium, Ci/m ³	182	13.8
Vessel off-gas water		
Tritium, Ci/m ³	182	13.8

4.1.2 Cases LWR-III and LMFBR-III: Voloxidation with Water Recycle

If tritium can be removed from the oxide fuel before it contacts the dissolver solution, a very small quantity of water at a high tritium concentration can be obtained for disposal. A process by which such removal has been shown to be effective in relatively small-scale operations is known as voloxidation. This process has been under study for years at ORNL and elsewhere.²⁻⁵

Voloxidation releases tritium by heating and oxidizing the oxide pellets of the sheared fuel. When LWR fuel is used, tritium is released with the disintegration of the UO₂ fuel matrix as it expands during its oxidation to U₃O₈. Tritium in voids and along grain boundaries is released immediately, and tritium within the matrix diffuses quickly from the small grains. The exact form of the tritium in the fuel is not known, but most of it is in the oxidized form as it leaves the voloxidizer.

For voloxidation of LWR fuel, holdup time in the voloxidizer, which is assumed to be a specially designed rotary kiln, is ~4 h. Sheared fuel is initially heated to 450°C to start UO₂ oxidation. Cooling is then required because the oxidation reaction is highly exothermic. Voloxidizer temperatures are monitored and maintained at 490 ± 20°C. The voloxidized product is cooled to <150°C before it is discharged to the dissolver.

When LWR fuel (or other fuel containing a sufficiently high concentration of UO₂) is employed, the voloxidation process has been shown in small-scale tests to remove >99%

of the tritium.^{2,3} However, when fuels containing more than ~15% of PuO₂ are voloxidized, the conversion to U₃O₈ does not proceed, the fuel pellets do not disintegrate, and longer times or higher temperatures are necessary to achieve high removal efficiencies. It has been established⁶ that voloxidation of LMFBR fuel will require a voloxidizer kiln with holdup volume four times that used for voloxidation of LWR fuel.

We have chosen, somewhat arbitrarily, to assume that voloxidation of UO₂ fuels (LWR fuels or LMFBR blanket materials) will remove 96% of the contained tritium on a 5 t/d scale. This assumption, which may do somewhat less than justice to the voloxidation process, would result in release of 4% of the tritium in the oxide matrix (108 Ci/d) to the LWR dissolver solution. We have also assumed that voloxidation of LMFBR fuel will be slightly less effective and that 25 Ci/d of tritium (5.8% of the total; see Sect. 2) will be delivered to the dissolver solution with the mixed fuel blanket oxides. The simplified flow-sheets for plants using voloxidation of LWR and LMFBR fuel are shown in Figs. 4.3 and 4.4. Water discharge rates and tritium concentrations in discharges, in concentrates, and in plant solutions are summarized in Table 4.3.

The voloxidation process requires, in addition to the voloxidizer, facilities to:

1. transfer the fuel to a rotary kiln where the fuel is heated, agitated to maintain UO₂ exposure, and oxidized to U₃O₈;
2. collect and filter the off-gas and transfer it to an off-gas treatment facility; and
3. cool the fuel to <150°C and transfer it from the kiln to the dissolver.

In addition to tritium, the off-gas stream contains some of the Kr, I, Xe, Cs, Ru, Br, and ¹⁴C, along with small fuel oxide particles and traces of other nuclides, as well as nitrogen and residual oxygen from the process air. Entrained dust particles are trapped and returned to the process as close to the voloxidizer as possible to prevent spread of radioactive contamination. Ruthenium, which could deposit in process lines, is isolated in traps. The remaining gases from the kiln are transferred to the tritium recovery system. Figure 4.5 is a schematic diagram of this system.

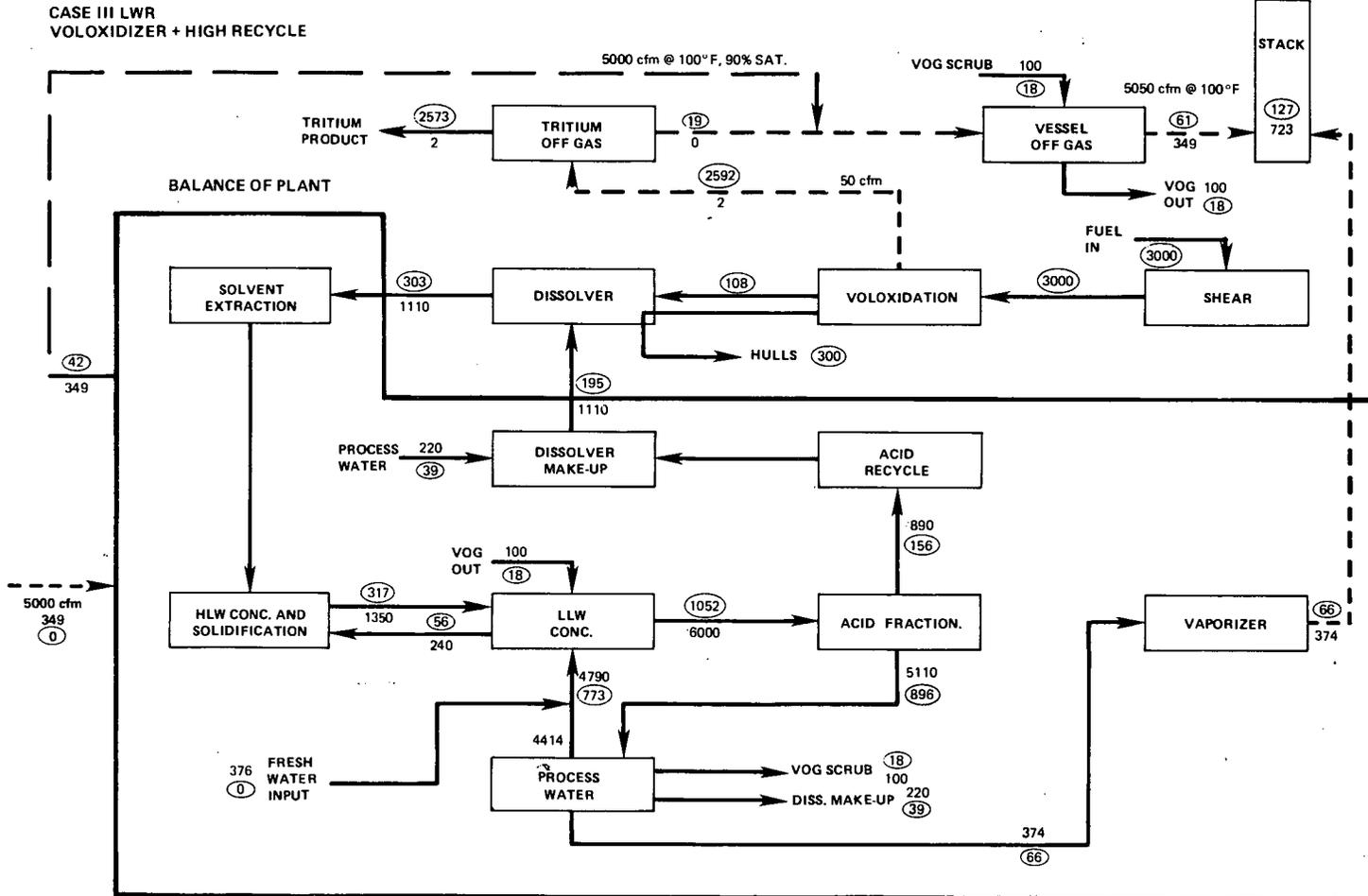
Confinement of the radioactive dust and fission gases to minimize contamination in the cell is a prime consideration in the voloxidation process. Control of process air for the oxidation of UO₂ is necessary, and air leakage must be minimized to keep down the volume of off-gas to be handled in the tritium recovery system.

The voloxidation equipment is remotely operated and maintained.³ There is a semi-continuous feed from the shear to the voloxidizer, but the voloxidizer operates continuously. The product from the voloxidizer is fed to dissolver vessels.

The tritium recovery system consists of a catalytic converter (to convert any molecular tritium to water) followed by a gas cooler, chiller, and molecular sieve bed (to provide near quantitative recovery of tritiated water).

From small-scale studies, the voloxidation process may be considered to be firmly based, especially for LWR fuels. There are several items that are certain to require additional engineering development before large-scale success of this option can be assured. These include:

1. reliability of the mechanisms for transferring fuel between the shear and voloxidizer and the voloxidizer and the dissolver,



Circled numbers: Curies of tritium per day.
Uncircled numbers: Liters of water per hour.

Fig. 4.3. Case III - LWR voloxidizer plus high recycle.

Table 4.3. Water discharge rates and tritium concentrations for plants with voloxidation and water recycle

Discharge conditions	Case	
	LWR-III	LMFBR-III
Discharge to stack		
H ₂ O, L/h	376	896
H ₂ O, m ³ /year	2 710	6 450
Tritium, Ci/year	3.81×10^4	8.4×10^3
Tritium, Ci/m ³	14.0	1.30
Retained for disposal		
H ₂ O, L/h	2	2
H ₂ O, m ³ /year	14.4	14.4
Tritium, Ci/year	7.72×10^5	1.19×10^5
Tritium, Ci/m ³	53 600	8 300
Recycle acid		
Tritium, Ci/m ³	7.38	1.3
Process water		
Tritium, Ci/m ³	7.38	1.3
Vessel off-gas water		
Tritium, Ci/m ³	7.38	1.3

2. dust confinement within the process train,
3. rotary seal life,
4. removal of decay and reaction heat for process and cell temperature control, and
5. isolation of the voloxidizer process gases from shearing and dissolving equipment.

It should be noted (see Table 4.3) that voloxidation promises to produce a very small volume of water at very high tritium concentrations for management or disposal. The process may offer additional benefits. Conversion by voloxidation of the UO₂ to finely divided U₃O₈ should, in principle, lead to more rapid dissolution of the fuel material; unfortunately in some cases, voloxidation seems to make dissolution of the contained PuO₂ more difficult.² Voloxidation may or may not assist in iodine recovery by volatilizing a portion of this element; evidence is incomplete (and somewhat contradictory). We have given no credit (or demerit) to voloxidation for its effect on this or on other aspects of the processing plants.

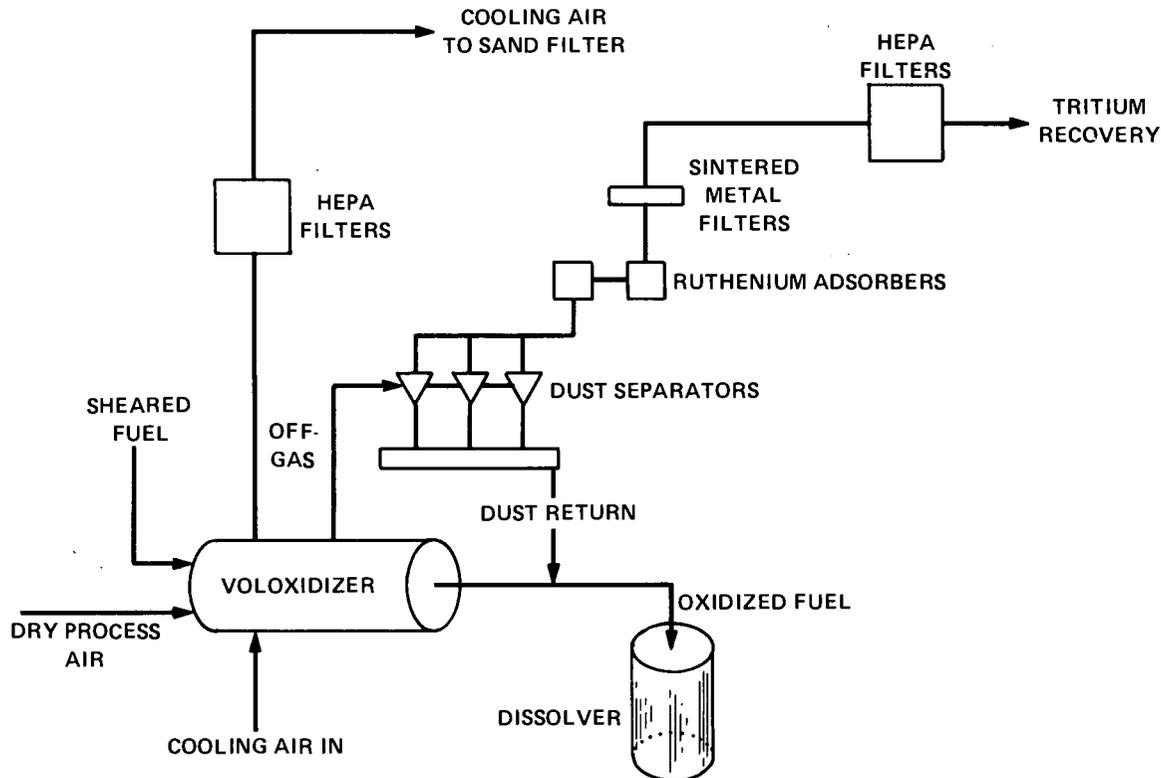


Fig. 4.5. Voloxidation system.

4.1.3 Cases LWR-IVA, LWR-IVB, LWR-VA, LWR-VB, LMFBR-IV, and LMFBR-V: Isotope Separation in Aqueous Streams with Water Recycle

Once the tritium contained in the oxide fuel matrix is diluted by the acidic dissolver solution, some form of isotope separation must be employed in order to obtain a concentrated tritium solution for management or disposal.

Choice of isotope separation techniques. Separation of hydrogen isotopes, primarily for production of deuterium or D_2O from normal water, has been studied for years. Virtually all the studied processes have been considered for their competency in the separation of tritium. Lin,⁷ in his review of the art, considered six methods and concluded that two processes (the dual temperature water- H_2S exchange process and the cryogenic hydrogen distillation process) had the highest potential for economic and technical feasibility. Burger and Trevor⁸ concluded that electrolysis coupled with chemical exchange or molecular excitation by lasers might prove to be viable alternatives to those selected by Lin. These processes are described in more detail in Appendix A.

An attempt was made to establish the preferred isotope separation options for this study from the published values of capital and operating costs of the several systems. In general, the published costs were for systems that differed in throughput, degree of separation,

concentration in product streams, project and process contingency factors, and value of capital money. Many of the published estimates contained little or no information about some of these parameters. In short, the literature did not yield conclusive information about the relative process economics of the several systems.

As a consequence, the several separations methods were compared on an arbitrary (and somewhat subjective) effectiveness basis. The seven criteria considered in this comparison were separation factor, throughput, process readiness, operational simplicity, industrial hazards, maintainability, and energy requirements.

The process options were evaluated on a relative basis for each criterion. Within each criterion, the best process was assigned a maximum of 10 points, whereas poorer processes were assigned lower values. The results of this effectiveness rating are shown in Table 4.4. From this table it was concluded that:

1. The laser-isotope separation process was not selected because this process has only been evaluated on a small scale, and thus, large-scale feasibility has not been established. This process needs to be reexamined in the future, and it may well become a competitor but is not considered further in this study.
2. Cryogenic distillation requires that large volumes of molecular hydrogen be generated, liquefied, stored, and distilled. It was rejected for this study because it seems very difficult to certify that it is sufficiently safe for remote or semi-remote operations.
3. Conventional electrolysis and bipolar electrolysis showed the lowest effectiveness rating of all the processes evaluated. Both of these options were rejected for this study, although further development may make bipolar electrolysis a competitor.

Table 4.4. Isotope separation effectiveness ratings

Criteria	Dual temperature exchange, modified variation	Electrolysis	Bipolar electrolysis	Fractional distillation	Cryogenic distillation	Molecular photo-excitation laser	Combined electrolysis catalytic exchange
Separation factor	4	7	8	0	5	10	6
Throughput	10	0	1	10	10	7	5
Process readiness	8	6	4	10	8	0	7
Operational simplicity	7	4	3	10	6	0	8
Industrial hazards	2	4	4	8	0	10	7
Maintainability	0	2	4	10	6	6	8
Energy requirement	3	4	8	0	4	10	8
Total	34	27	32	48	39	43	49

As a consequence of this evaluation, the two isotope separation processes with the highest ratings were selected for consideration in this study. These processes are the combined electrolysis catalytic exchange and the fractional distillation of H_2O . In addition, the modified or dual-temperature H_2S exchange process was included because it represents an improved version of a proven large-scale process. This modified process (see Appendix A) introduces pure water to the top of the cold towers, and, as a consequence, yields markedly larger volumes of tritium-depleted water than do the other two processes considered.

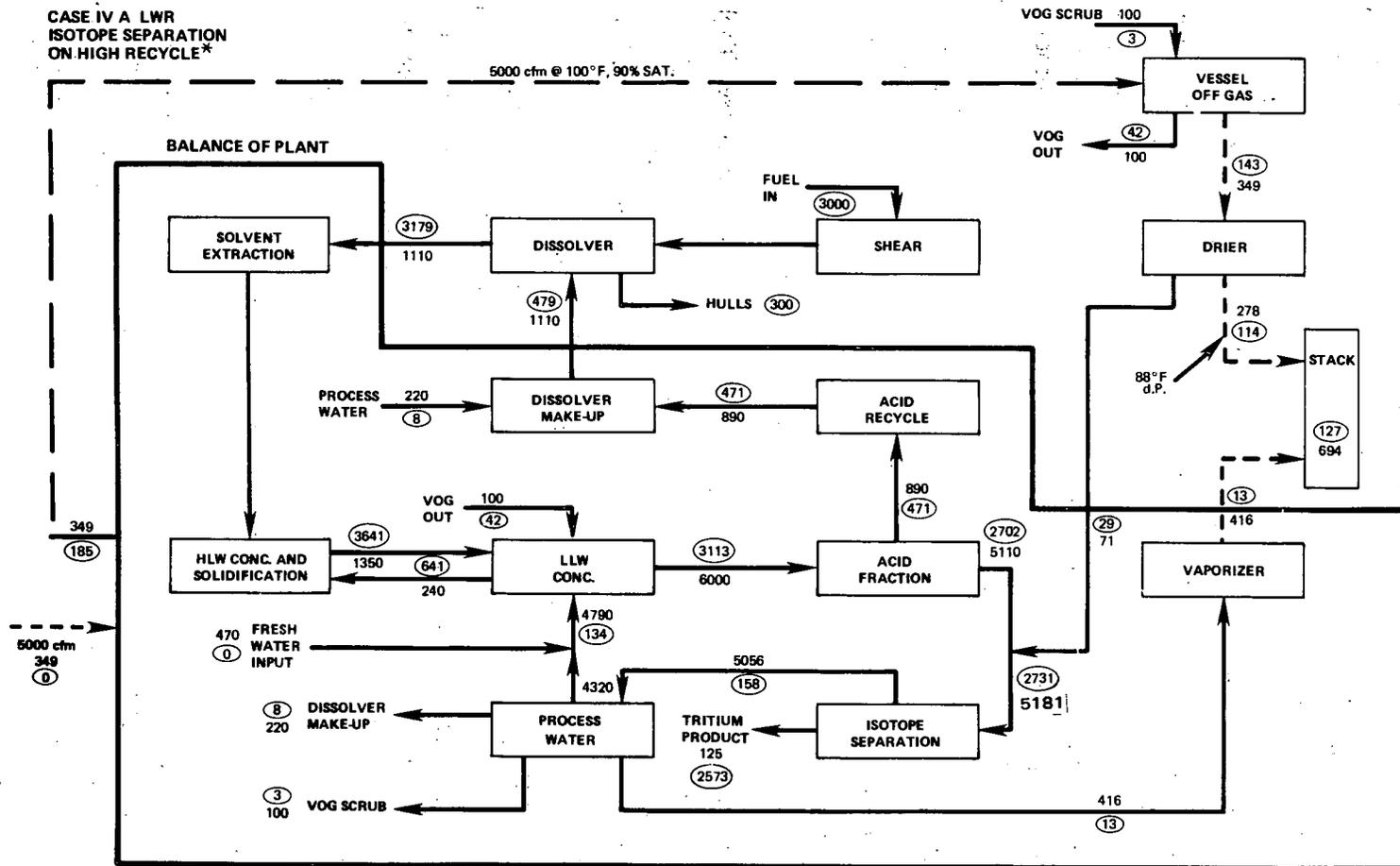
Requirements for the isotope separations process. As described in Sect. 4.1.2, it was assumed that the voloxidation process would remove $\sim 96\%$ of the contained tritium from the oxide contained in the fuel, and as Table 4.2 indicates, would result in the release of 127 Ci/d of tritium to the atmosphere from LWR fuel reprocessing and 28 Ci/d of tritium from reprocessing of LMFBR fuel. The LWR and LMFBR plants employing isotope separation of tritium from the aqueous process solutions were constrained in this study to produce tritium releases to the atmosphere to match those of the plants employing voloxidation (see Table 4.1).

At least two options that use isotope separation processes on aqueous plant solutions are available. These options are detailed as LWR-IVA and LMFBR-IV in Table 4.1 and in the following text. One of these would use the isotope separation system on the entire water recycle system of the reprocessing plants. The other alternative would use the isotope separation process only on the discharge stream to provide (with a considerably smaller volume for treatment) a concentrated tritium solution for management or disposal and a dilute solution of tritiated water for atmospheric dispersal. These alternatives (see Table 4.1) are designated as LWR-VA and LMFBR-V. As a means of testing the sensitivity of removal efficiencies on the overall cost of the facility, two additional sub-cases were examined. In these isotope separations options for LWR fuel, the retention efficiency was increased from 95 to 99.5% of the tritium, and their costs were estimated. These are listed as cases LWR-IVB and LWR-VB.

Where the combined electrolysis catalytic exchange (CECE) process or fractional distillation of water is used as the isotope separation process, the simplified flowsheets for LWR-IVA, LWR-IVB, and LMFBR-IV are shown as Figs. 4.6 through 4.8. For these two isotope separations, the flowsheets for LWR-VA, LWR-VB, and LMFBR-V are Figs. 4.9 through 4.11. For these two isotope separations processes, the water discharge rates and the tritium concentrations in discharges and plant solutions are shown for LWR-IVA, LWR-IVB, and LMFBR-V in Table 4.5. Similar data for LWR-VA, LWR-VB, and LMFBR-V are shown in Table 4.6.

The modified dual-temperature exchange process can furnish concentrates to be retained for disposal and can achieve tritium concentrations in plant solutions essentially equivalent to those shown in Tables 4.5 and 4.6. However, this process adds pure water during the isotope separations process and yields dilute tritiated water in volumes far larger than those shown in the tables. For case LWR-IVA, for example, the tritium (158 Ci/d) in the stripped stream is contained in about 2.83×10^6 L of water. Only a very small fraction of this could be fed to the plant's process water (as shown in Fig. 4.6); therefore, use of the modified dual-temperature exchange system for case LWR-IVA (as also for cases LWR-IVB and LMFBR-IV) requires disposal of very large volumes of dilute tritiated water. Evaporation of these volumes of water via the plant stack might not be feasible, would be

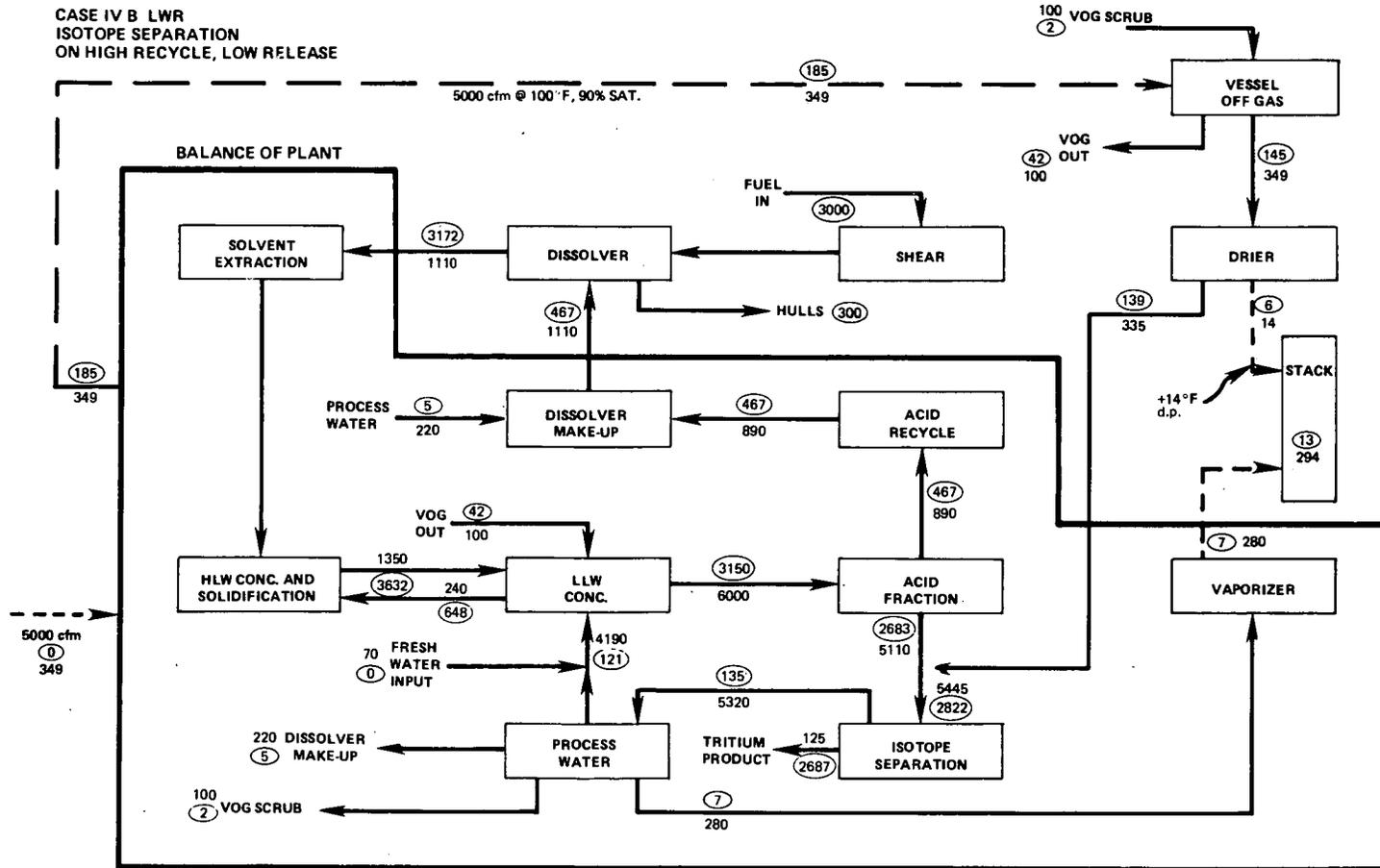
CASE IV A LWR
ISOTOPE SEPARATION
ON HIGH RECYCLE*



*Flow sheet applicable to CECE and fractional distillation processes.
Circled numbers: Curies of tritium per day.
Uncircled numbers: Liters of water per hour.

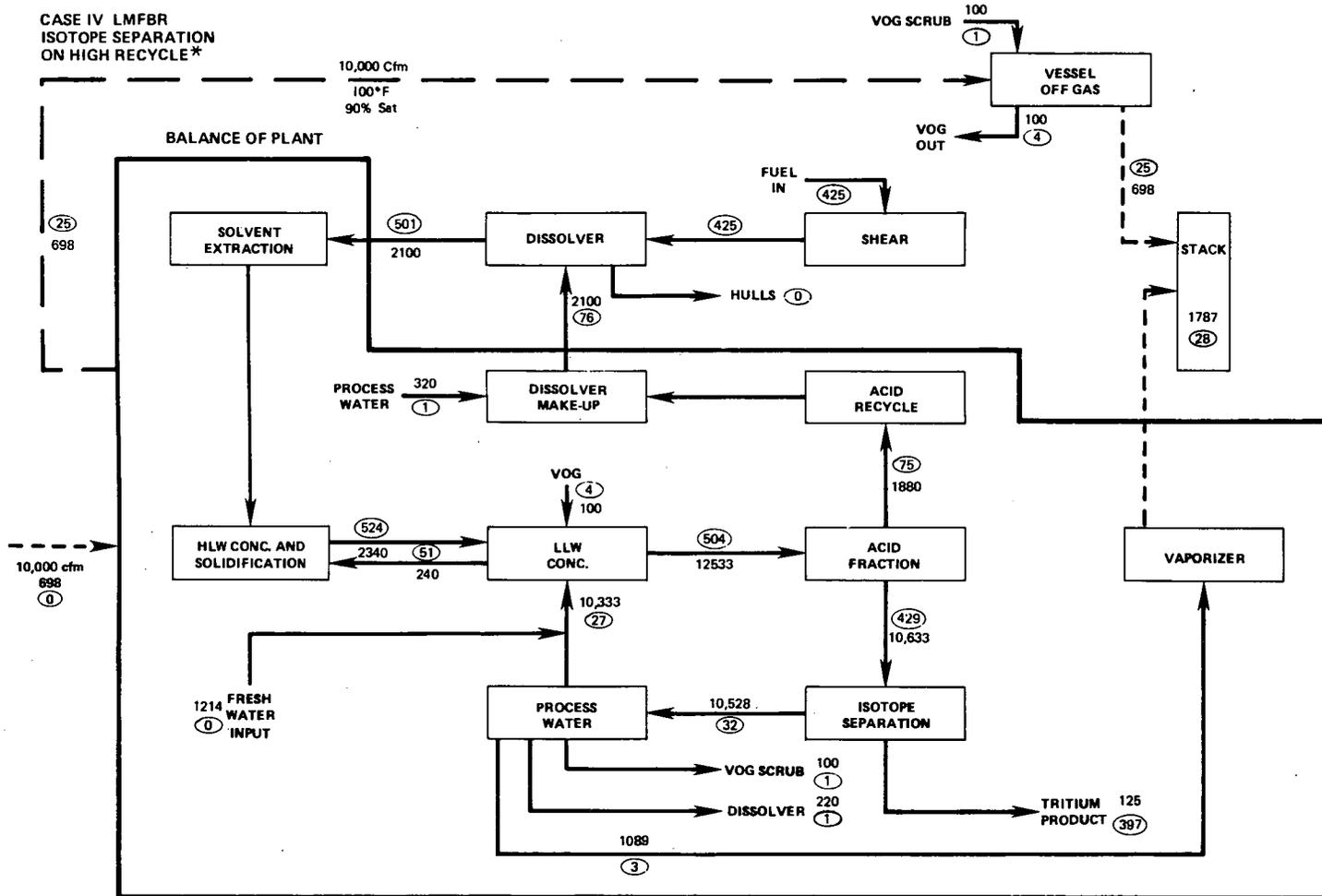
Fig. 4.6. Case IVA - LWR isotope separation on high recycle.

CASE IV B LWR
ISOTOPE SEPARATION
ON HIGH RECYCLE, LOW RELEASE



Circled numbers: Curies of tritium per day.
Uncircled numbers: Liters of water per hour.

Fig. 4.7. Case IVB – LWR isotope separation on high recycle low release.



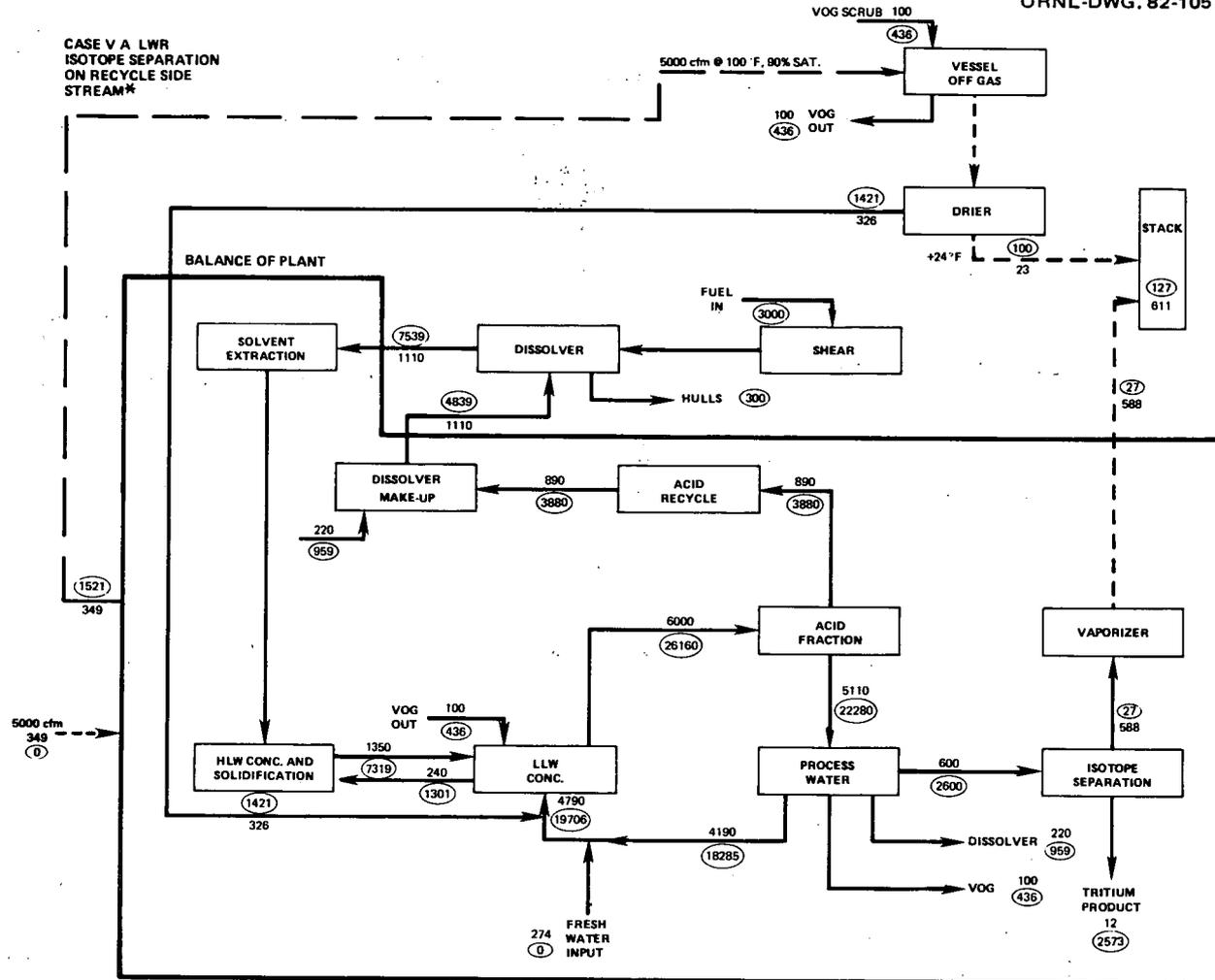
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*Flow sheet applicable to CECE and fractional distillation processes only.

Circled numbers: Curies of tritium per day.

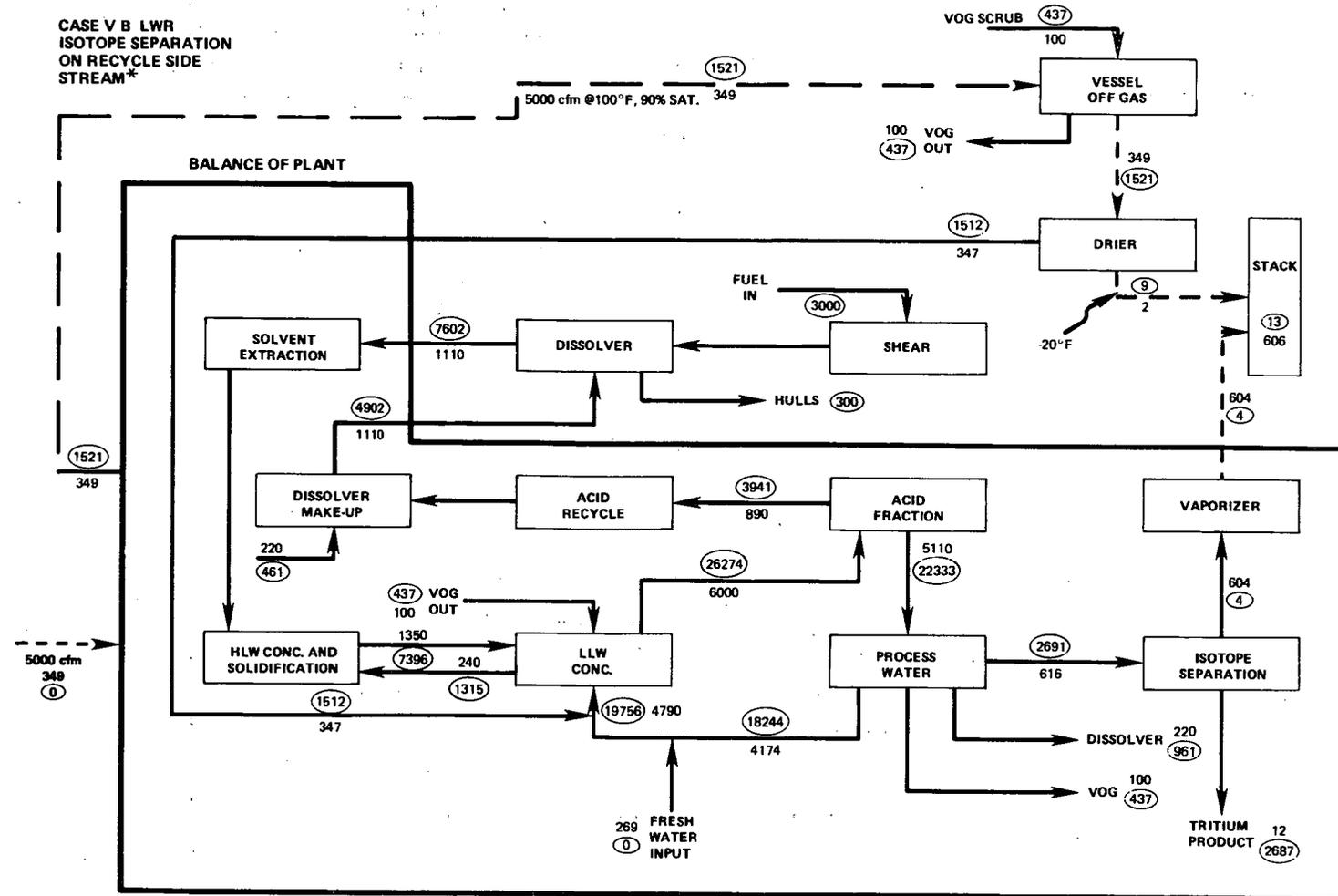
Uncircled numbers: Liters of water per hour.

Fig. 4.8. Case IV - LMFBR isotope separation on high recycle.



*Feed to vaporizer shown for CECE and fractional distillation processes. See text.
 Circled numbers: Curies of tritium per day.
 Uncircled numbers: Liters of water per hour.

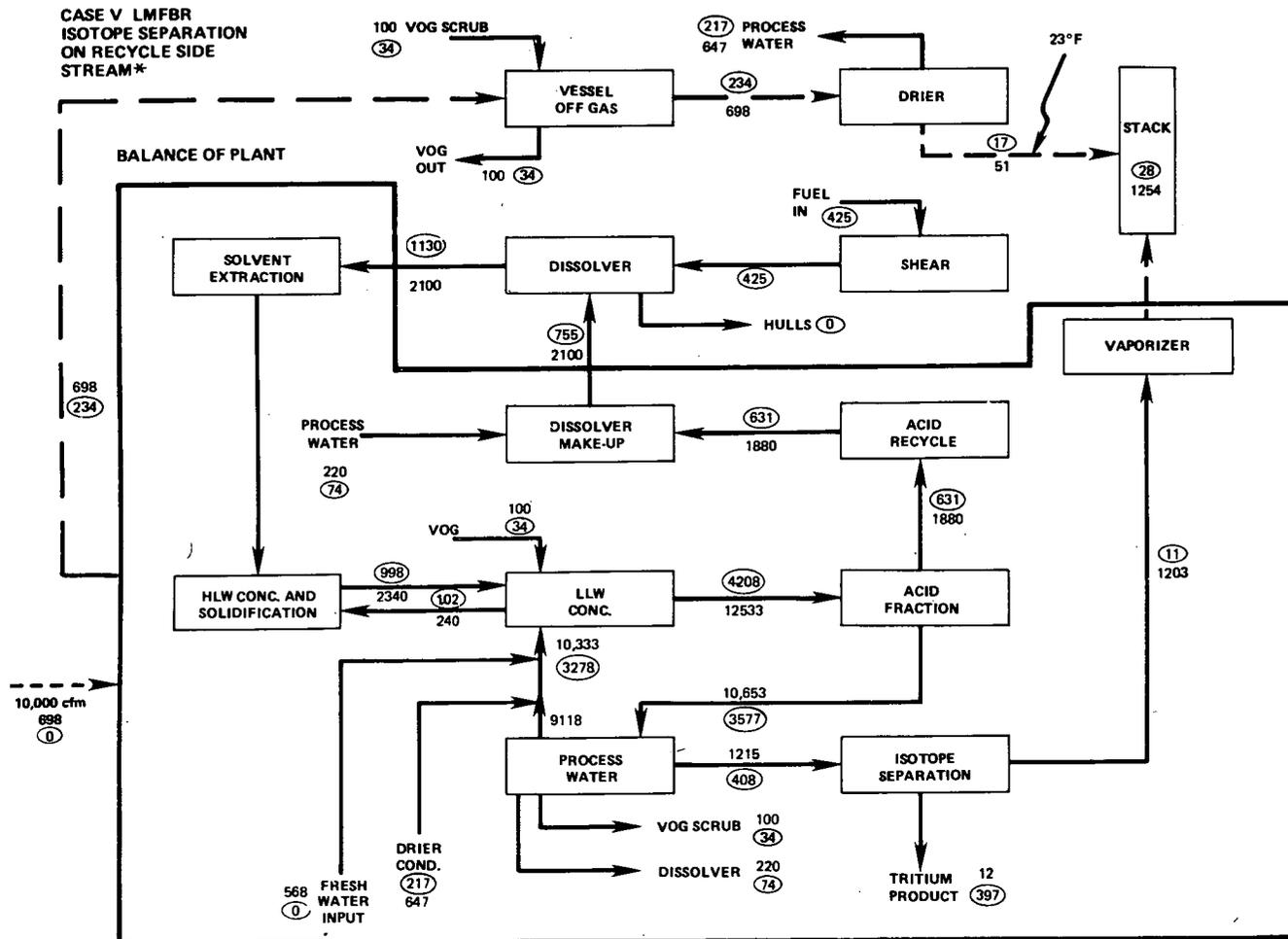
Fig. 4.9. Case VA – LWR isotope separation on recycle side stream.



4-17

*Feed to vaporizer shown for CECE and fractional distillation processes. See text.
 Circled numbers: Curies of tritium per day.
 Uncircled numbers: Liters of water per hour.

Fig. 4.10. Case VB - LWR isotope separation on recycle side stream, low release.



*Feed to vaporizer shown for CECE and fractional distillation processes. See text.

Circled numbers: Curies of tritium per day.

Uncircled numbers: Liters of water per hour.

Fig. 4.11. Case V - LMFBR isotope separation on recycle side stream.

Table 4.5. Water discharge rates and tritium concentrations for plants with the combined electrolysis catalytic exchange or the fractional distillation isotope separation on total recycle water

Discharge conditions	Case		
	LWR-IVA	LWR-IVB	LMFBR-IV
Discharge to stack			
H ₂ O, L/h	694	294	1 787
H ₂ O, m ³ /year	4 997	2 117	12 866
Tritium, Ci/year	3.8×10^4	3.9×10^3	8.4×10^3
Tritium, Ci/m ³	7.62	1.84	0.65
Retained for disposal			
H ₂ O, L/h	125	125	125
H ₂ O, m ³ /year	900	900	900
Tritium, Ci/year	7.7×10^5	8.1×10^5	1.2×10^5
Tritium, Ci/m ³	858	895	133
Recycle acid			
Tritium, Ci/m ³	22.1	21.9	1.7
Process water			
Tritium, Ci/m ³	1.3	1.0	1.7
Vessel off-gas water			
Tritium, Ci/m ³	17.1	17.9	1.5

Table 4.6. Water discharge rates and tritium concentrations for plants with isotope separation on discharge water

Discharge conditions	Case		
	LWR-VA	LWR-VB	LMFBR-V
Discharge to stack			
H ₂ O, L/h	611	606	1254
H ₂ O, m ³ /year	4400	4363	9028
Tritium, Ci/year	3.8×10^4	3.9×10^3	8.4×10^3
Tritium, Ci/m ³	8.67	0.89	0.93
Retained for disposal			
H ₂ O, L/h	12	12	12
H ₂ O, m ³ /year	86.4	86.4	86.4
Tritium, Ci/year	7.7×10^5	8.1×10^5	1.2×10^5
Tritium, Ci/m ³	8935	9329	1377
Recycle acid			
Tritium, Ci/m ³	182	185	14
Process water			
Tritium, Ci/m ³	182	182	14
Vessel off-gas water			
Tritium, Ci/m ³	182	188	14

quite expensive, and would (unless additional separation stages were introduced at considerable added cost) result in larger tritium releases to the atmosphere than those shown in Figs. 4.6 through 4.8. Disposal of such volumes of tritiated water (at 5.6×10^{-5} Ci/L or less) via evaporation from a large holding pond might be feasible in a sufficiently arid region, and disposal by discharge to a sufficiently large surface stream possibly might be permissible. However, discharges of either type would violate the ground rules of this study. Accordingly, the modified dual-temperature exchange process must be considered unacceptable for cases LWR-IVA, LWR-IVB, and LMFBR-IV unless on-site deep-well disposal of very large volumes of dilute tritiated water is feasible and permissible. It should be noted, however, that if such disposal were available, the base-case plants (see Figs. 3.1 and 3.2) could achieve low atmospheric releases without the expense of isotope separation by sending only the vessel off-gas effluent to the stack and by sending the excess water to deep-well disposal instead of to the vaporizer.

Application of the modified dual-temperature exchange process to LWR-VA, LWR-VB, and LMFBR-V also results in far larger volumes of water for release than those shown in Table 4.6. However, the quantities (13 600, 14 000, and 27 500 L/h of water for LWR-VA, LWR-VB, and LMFBR-V respectively) seem feasible (although expensive) to evaporate via the plant stack. The modified dual-temperature exchange process, accordingly, has been included for consideration for these cases when the isotope separation process is applied to the discharge water.

The isotope separation systems operating only on the excess water (LWR-VA, LWR-VB, and LMFBR-V) are smaller in size, and they produce a small volume of concentrate for management or disposal. However, they do not minimize the amount of tritium within the plant's recycle solution; the tritium content of the water to the chemical make-up area is essentially identical to that of cases LWR-II and LMFBR-II, respectively, which operate with ~90% water recycle without special tritium separations.

It should also be noted that, although the isotope separation systems are large facilities, they are designed to operate with tritium as the only significant radionuclide in the water, and, as a consequence, do not require containment within high-cost, heavily shielded cell areas.

4.1.4 Cases LWR-VI and LMFBR-VI (Tritium Confinement to First Cycle Solutions)

A proposed alternative method of isolating the tritium in fuel reprocessing plants is that of confining this isotope, insofar as possible, to the first cycle portion of the plant.⁹⁻¹¹

The hydrogen in Purex solvents exchanges very slowly with tritium; as a consequence, the tritium carried in the organic stream of the first cycle is effectively limited to that in the (~5%) entrained and dissolved nitric acid and water.¹² The tritium/protium ratio in the organic stream is effectively set by the tritium/protium ratio in the scrub stream and is subject to some control. Process parameters can be adjusted to limit tritium passage to the uranium and plutonium decontamination columns to the order of 0.2% of the tritium processed through the first cycle column.

The key to head-end confinement is to reduce the water additions to an absolute minimum through the use of high-level process water (i.e., chemical make-up solutions, high-level acid, steam jets, etc.). The acid must be made using the NO_x produced with a denitrator using recycle acid from the low-level side. The low-level concentrator bottoms

add to the water input to the high level side. Also, a small amount of water from decontamination solutions must be disposed of in this system. In addition, the transfer of water with high tritium concentrations to the low side of the plant must be reduced to a minimum. These precautions will keep the amount of tritiated water for disposal as small as possible.

Simplified flowsheets for cases LWR-VI and LMFBR-VI are shown in Figs. 4.12 and 4.13. Water flow balances and discharge rates are shown along with tritium concentrations in discharge water and process streams in Table 4.7.

It should be noted that these options yield large volumes of tritiated water for disposal and that the concentration of tritium in the process water returned to the chemical make-up area of the plant is the highest of any of the options considered. It also seems likely that plants of this design are more vulnerable to upset conditions than are most of the other options considered.

4.1.5 Estimated Costs of the Retention Options

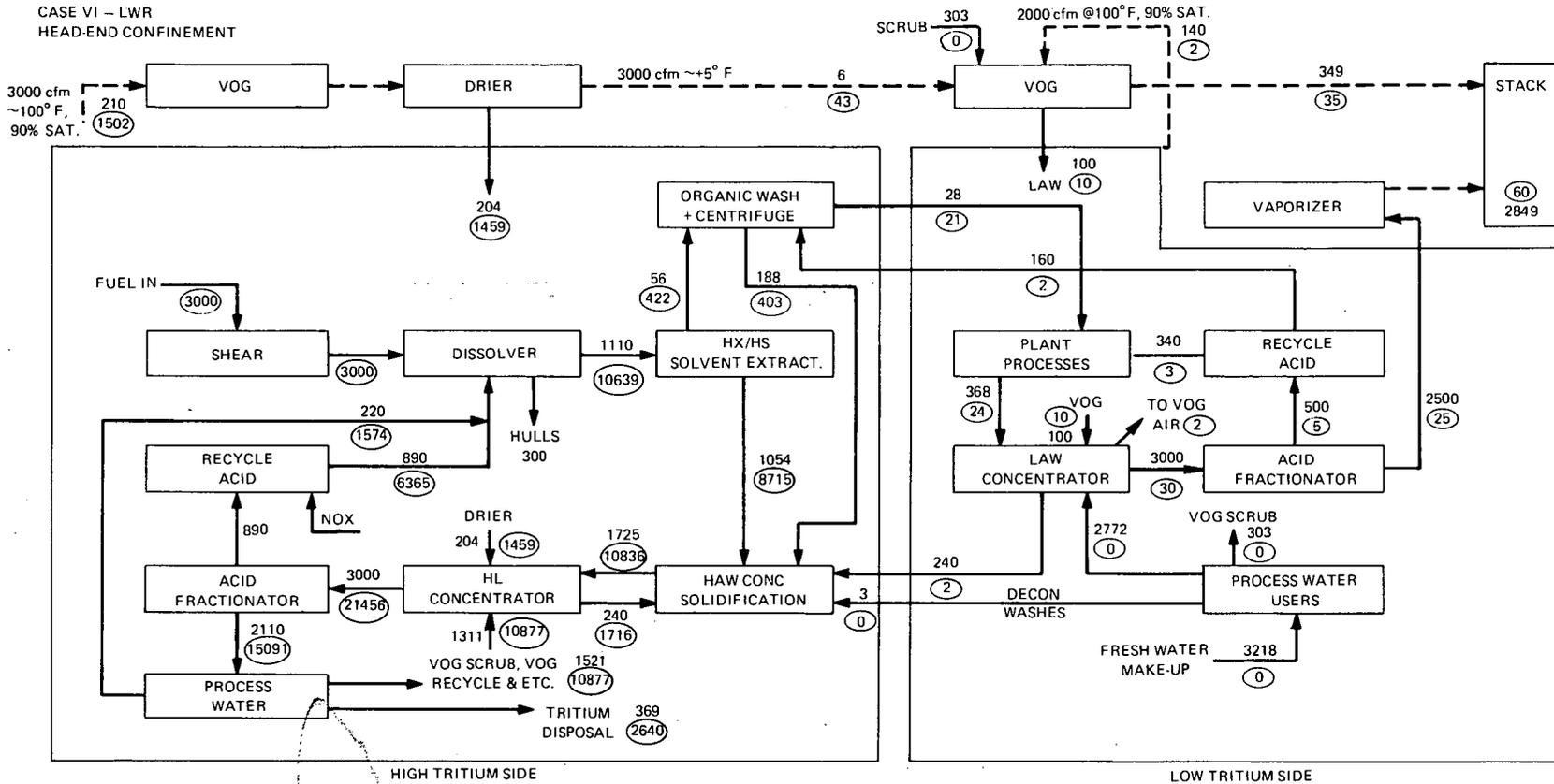
Estimates of incremental capital costs. A previous study¹³ by the Exxon Nuclear Company (ENC) for the Consolidated Fuel Reprocessing Program at the Oak Ridge National Laboratory, along with the detailed studies¹⁴ for the Nuclear Fuel Recovery and Recycling Center proposed by ENC, provided bases for much of the capital cost information developed in the present study. All capital costs presented below have been converted to first quarter 1980 dollars. Details of cost estimation are in Appendix B.

For each case of the methods of tritium isolation, a detailed listing was prepared showing major in-cell process equipment required. In each case, the equipment items were designed and sized to provide the performance indicated in Tables 4.4 through 4.6. The three isotope separations processes [modified dual-temperature Girdler-sulfide (GS), combined electrolysis catalytic exchange, and vacuum distillation] were each sized to provide the separation required for cases LWR-IVA, LWR-IVB, LWR-VA, LWR-VB, LMFBR-IV, and LMFBR-V. Estimated costs of each item were derived from sources judged to be the best available.

Tables 4.8 and 4.9 summarize the incremental costs [above those of the base-case plants (numbers LWR-I and LMFBR-I)] for each case.

Several points, obvious from examination of these tables, include:

1. Incorporation of water recycle (cases LWR-II and LMFBR-II) results in a modest decrease in capital costs.
2. In the isotope separation systems that operate on the entire recycle water (cases LWR-IVA, LWR-IVB, and LMFBR-IV), the modified GS process appears to be substantially less expensive than the CECE process and markedly less expensive than vacuum distillation. However, as previously noted and in Appendix A, application of the modified GS process to these particular cases yields dilute tritiated water in volumes so large as to preclude its dispersal via evaporation through the plant stack; accordingly, no vaporization equipment is included for these cases. Moreover, it should be noted that for these cases all the isotope separations processes appear to be more expensive than voloxidation.
3. For operation on the discharge water only (cases LWR-VA, LWR-VB, and LMFBR-V), the GS process (including equipment for vaporization of relatively large volumes



Circled numbers: Curies of tritium per day.
Uncircled numbers: Liters of water per hour.

Fig. 4.12. Case VI - LWR head-end confinement.

Table 4.7. Water discharge rates and tritium concentrations for plants with first cycle tritium confinement

Discharge conditions	Case	
	LWR-VI	LMFBR-VI
Discharged to stack		
H ₂ O, L/h	2 849	5 698
H ₂ O, m ³ /year	20 500	41 025
Tritium, Ci/year	1.8 × 10 ⁴	5.1 × 10 ³
Tritium, Ci/m ³	0.878	0.125
Retained for disposal		
H ₂ O, L/h	369	366
H ₂ O, m ³ /year	2 660	2 635
Tritium, Ci/year	7.9 × 10 ⁵	1.1 × 10 ⁵
Tritium, Ci/m ³	298	41.7
Recycle acid		
Tritium, Ci/m ³	300	47.0
Process water		
Tritium, Ci/m ³	300	47.0
Vessel off-gas water		
Tritium, Ci/m ³	300	47.0

of dilute tritiated water) appears to be nearly equal in cost to the CECE process; both are markedly less expensive than vacuum distillation and both appear to be less expensive than voloxidation.

4. First cycle containment, although it requires an addition of several equipment items, is relatively inexpensive (cases LWR-VI and LMFBR-VI).

Estimation of operating costs. Operating costs were estimated on a comparable basis for all options for processing LWR and LMFBR fuels. LWR-I and LMFBR-I again served as the base cases, and operating costs for the individual options incremental to those cases were estimated. (The assumptions used and details of the bases employed are included in Appendix B.) Table 4.10 summarizes the incremental annual operating costs for several cases.

It should be noted that the options using water recycle only (cases LWR-II and LMFBR-II) show operating costs below those of the corresponding base-case plants.

When the isotope separation systems are applied to all the recycle water (cases LWR-IVA, LWR-IVB, and LMFBR-IV), the modified GS process shows the lowest operating costs, largely because no costs for evaporation of excess water are included. However, as noted previously, this process could meet the ground rules of this study only if on-site, deep-well disposal of very large volumes of tritiated water were feasible and permissible.

When the isotope separation systems are applied to the discharge water only (cases LWR-VA, LWR-VB, and LMFBR-V), the modified GS process shows the highest operating costs of any of the isotope separations systems. These high operating costs (in large part)

Table 4.8. Capital cost comparisons for options retaining tritium from LWR fuel

Case	Description	Incremental costs (\$1 000)		
		In-cell equipment	Total direct	Total project ^a
I	Reference case – no recycle	0	0	0
II	High recycle	-187	-617	-1 500
III	Voloxidation	4 691	26 270	66 167
IVA	Modified Girdler-sulfide process ^{b,c}	6 874	38 494	96 958
	Vacuum distillation ^b	43 300	242 480	610 747
	Combined electrolysis catalytic exchange ^b	10 978	61 475	154 845
IVB	Modified Girdler-sulfide process – low release ^{b,c}	7 379	41 322	104 081
	Vacuum distillation – low release ^b	45 100	252 560	636 136
	Combined electrolysis catalytic exchange ^b	11 061	61 942	156 015
VA	Modified Girdler-sulfide process ^d	2 011	11 262	28 365
	Vacuum distillation ^d	7 730	43 288	109 032
	Combined electrolysis catalytic exchange ^d	1 947	10 903	27 462
VB	Modified Girdler-sulfide process – low release ^d	2 555	14 308	36 038
	Vacuum distillation – low release ^d	9 830	55 048	138 652
	Combined electrolysis catalytic exchange – low release ^d	2 354	13 182	33 203
VI	First cycle confinement	1 199	5 756	16 912

^aTotal project includes: 30% field distributable costs, 25% home office costs, and 55% contingency; it totals a 2.52 multiplier on total direct costs before rounding.

^bIsotope separation process is applied to entire (~90%) water recycle.

^cThese options, which require very large additions of fresh water to the isotope separation operation, are impracticable unless on-site, deep-well disposal of very large volumes of water is feasible (see text).

^dWater recycle (~90%) employed but isotope separation is applied to discharge (excess) water only.

are because of the high utility costs incurred in vaporization of the very large volumes of tritiated water. These higher operating costs more than offset (for cases LWR-VA, LWR-VB, and LMFBR-V) the relatively small capital cost advantage that the modified GS process has over the CECE process.

Table 4.9. Capital cost comparisons for tritium retention options for LMFBR fuel

Case	Description	Delta costs (\$1 000)		
		In-cell equipment	Total direct	Total project ^d
I	Reference case – no recycle	0	0	0
II	High recycle	-315	-1 040	-2 500
III	Voloxidation	7 255	40 628	102 332
IV	Modified Girdler-sulfide process ^{b,c}	14 427	80 792	203 493
	Vacuum distillation ^b	91 730	513 688	1 293 852
	Combined electrolysis catalytic exchange process ^b	23 315	130 564	328 860
V	Modified Girdler-sulfide process ^d	3 281	18 374	46 279
	Vacuum distillation ^d	10 045	56 252	141 685
	Combined electrolysis catalytic exchange process ^d	3 260	18 256	45 982
VI	First cycle containment	1 299	7 274	18 322

^aTotal project includes: 30% field distributable costs, 25% home office costs, and 55% contingency; it totals a 2.52 multiplier on total direct costs before rounding.

^bIsotope separation process is applied to entire (~90%) water recycle.

^cThis option, which requires very large additions of fresh water to the isotope separation process, is impracticable unless on-site, deep-well disposal of very large volumes of tritiated water is feasible (see text).

^dWater recycle (~90%) employed but isotope separation is applied to discharge (excess) water only.

Summary of estimated capital and operating costs. Tables 4.11 and 4.12 summarize the cost evaluations for LWR and LMFBR reprocessing plant options for retention of tritium with government financing assumed (20-year facility lifetime, zero salvage value, and cost of money at 7.5%) for the capital. Those cases involving isotope separations have assumed that the CECE process would be used.

Capitalized costs for several cases, assuming 20-year lifetime of the facilities, zero value after 20 years, and with the cost of money at 12 and at 15%, are summarized in Tables 4.13 and 4.14.

These tables clearly indicate that for both LWR and LMFBR plants, the cases employing recycle of water as well as acid (LWR-II and LMFBR-II) are, by a considerable margin, the most economical. However, as shown in Tables 4.15 and 4.16, which summarize the water volumes and tritium concentrations for all the cases, these recycle options discharge large volumes of tritiated water for disposal, and they send relatively concentrated solutions of tritium to the chemical make-up areas.

Voloxidation does not appear to be economically attractive for LWR fuel. It has the advantage, however, of providing the smallest volume of concentrate for disposal, and it provides recycle waters containing tritium at low concentrations.

Table 4.10. Incremental annual operating cost summary
(first quarter 1980 dollars)

Case	Description	Incremental cost (\$1 000)
LWR-I		
LWR-II		-1 129
LWR-III		993
LWR-IVA	Modified Girdler-sulfide process ^a	2 014
	Distillation	6 793
	Combined electrolysis catalytic exchange	3 260
LWR-IVB	Modified Girdler-sulfide process ^a	2 166
	Distillation	7 180
	Combined electrolysis catalytic exchange	3 869
LWR-VA	Modified Girdler-sulfide process	3 912
	Distillation	1 753
	Combined electrolysis catalytic exchange	1 169
LWR-VB	Modified Girdler-sulfide process	4 052
	Distillation	2 000
	Combined electrolysis catalytic exchange	1 241
LWR-VI		1 292
LMFBR-I		
LMFBR-II		-2 350
LMFBR-III		1 164
LMFBR-IV	Modified Girdler-sulfide process ^a	3 502
	Distillation	13 635
	Combined electrolysis catalytic exchange	6 903
LMFBR-V	Modified Girdler-sulfide process	7 121
	Distillation	2 309
	Combined electrolysis catalytic exchange	1 623
LMFBR-VI		1 815

^aThese options are impracticable unless on-site, deep-well disposal of very large volumes of tritiated water is feasible (see text). No costs for evaporation of water are included.

Use of the isotope separation process on the total recycle stream (cases LWR-IVA, LWR-IVB, and LMFBR-IV) leads to the most expensive of the facilities and also are the ones that furnish relatively large volumes of tritiated water to storage or disposal (see Tables 4.15 and 4.16). It has the virtue of sending a quite dilute tritiated water as recycle to the plant.

Operation of the isotope separation system on the discharge stream (cases LWR-VA, LWR-VB, and LMFBR-V) produces (see Tables 4.15 and 4.16) a much smaller volume of tritiated water for disposal and, because of the smaller sizes of the separation facility, appears reasonably economical. The recycle process waters, however, are as concentrated

Table 4.11. LWR retention cost summary – government financing
(7.5% interest, 20-year lifetime)

Costs	Case							
	I Reference case	II Water recycle	III Voloxi- dation	IVA Isotope separation ^a	IVB Isotope separation ^b	VA Isotope separation ^c	VB Isotope separation ^d	VI First cycle confinement
Capital costs, million \$	0	-1.5	66.2	154.8	156.0	28.4	36.0	16.9
Capitalized cost, million \$/year	0	-0.147	6.495	15.187	15.305	2.786	3.532	1.658
Operating costs, million \$	0	-1.129	0.933	3.620	3.869	1.169	1.241	1.292
Uniform annual costs, million \$	0	-1.28	7.428	18.81	19.174	3.96	4.60	2.95
Annual tritium reduction to environment, MCI	0	0.705	0.772	0.772	0.806	0.772	0.806	0.792
Annual cost, \$/Ci retained		-1.81	9.62	24.40	23.80	5.15	5.70	3.73

^aCombined electrolysis catalytic exchange process on total recycle stream.

^bCombined electrolysis catalytic exchange process on total recycle stream; very low release to atmosphere.

^cCombined electrolysis catalytic exchange process on discharge stream only.

^dCombined electrolysis catalytic exchange process on discharge stream only; very low release to atmosphere.

Table 4.12. LMFBR retention cost summary – government financing
(7.5% interest, 20-year lifetime)

Costs	Case					
	I Reference	II Water recycle	III Voloxi- dation	IV Isotope separation ^a	V Isotope separation ^b	VI First cycle confinement
Capital costs, million \$	0	-2.5	102.3	328.9	46.0	18.3
Capitalized cost, million \$/year	0	-0.245	10.036	32.268	4.513	1.795
Operating costs, million \$		-2.350	1.164	6.903	1.623	1.815
Uniform annual costs, million \$	0	-2.60	11.20	39.17	6.136	3.61
Annual tritium reduction to environment, MCI		0.112	0.119	0.119	0.119	0.122
Annual costs, \$/Ci retained		-23.20	94.10	329.20	51.60	29.60

^aCombined electrolysis catalytic

^aCombined electrolysis catalytic exchange process on total water recycle.

^bCombined electrolysis catalytic exchange process on excess (discharge) water.

Table 4.13. LWR cost evaluation summary – industrial financing

Case	12% interest, 20-year lifetime		15% interest, 20-year lifetime	
	Capital costs (million dollars)	Capitalized cost (million dollars/year)	Capital costs (million dollars)	Capitalized cost (million dollars/year)
I – Reference	0	0	0	0
II – Recycle	-1.5	-0.201	-1.5	-0.240
III – Voloxidation	66.2	8.86	66.2	10.59
IVA – Isotope separation ^a	154.8	20.73	154.8	24.64
IVB – Isotope separation ^b	156.0	20.88	156.0	24.96
VA – Isotope separation ^c	27.5	3.68	27.5	4.40
VB – Isotope separation ^d	33.2	4.44	33.2	5.31
VI – First cycle confinement	16.9	2.26	16.9	2.70

^aCombined electrolysis catalytic exchange process on total recycle stream.

^bCombined electrolysis catalytic exchange process on total recycle stream; very low release to atmosphere.

^cCombined electrolysis catalytic exchange process on discharge stream only.

^dCombined electrolysis catalytic exchange process on discharge stream only; very low release to atmosphere.

Table 4.14. LMFBR cost evaluation summary – industrial financing

Case	12% interest, 20-year lifetime		15% interest, 20-year lifetime	
	Capital costs (million dollars)	Capitalized cost (million dollars/year)	Capital costs (million dollars)	Capitalized cost (million dollars/year)
I – Reference	0	0	0	0
II – Recycle	-2.5	-0.335	-2.5	-0.399
III – Voloxidation	102.3	13.70	102.3	16.376
IV – Isotope separation ^a	328.9	44.05	328.9	52.62
V – Isotope separation ^b	46.0	6.16	46.0	7.36
VI – First cycle confinement	18.3	2.45	18.3	2.93

^aCombined electrolysis catalytic exchange process on total recycle stream.

^bCombined electrolysis catalytic exchange process on discharge stream only.

Table 4.15. Summary of water volumes and tritium concentrations for LWR fuel reprocessing

Case	Stack discharges			Discharge waste stream			Tritium		
	Water (L/h)	Tritium (Ci/d)	Tritium (Ci/L)	Water (L/h)	Tritium (Ci/d)	Tritium (Ci/L)	Recycle acid (Ci/L)	Process water (Ci/L)	Vessel off-gas (Ci/L)
I	5459	2700	0.0206	0	0	0	0.0212	0	0.0156
II	80	349	0.182	539	2351	0.182	0.182	0.182	0.182
III	376	127	0.0141	2	2573	53.604	0.00730	0.00739	0.00728
IVA	694	127	7.6×10^{-4}	125	2573	0.858	0.0221	0.00130	0.0171
IVB	294	13	1.8×10^{-3}	125	2687	0.86	0.0219	0.00104	0.0179
VA	611	127	8.7×10^{-3}	12	2573	8.93	0.182	0.182	0.181
VB	606	13	8.9×10^{-4}	12	2687	9.3	0.185	0.182	0.188
VI	2849	60	8.8×10^{-4}	369	2640	0.39	0.39	0.30	0.004

Table 4.16. Summary of water volumes and tritium concentrations for LMFBR fuel reprocessing

Case	Stack discharges			Discharge waste stream			Tritium		
	Water (L/h)	Tritium (Ci/d)	Tritium (Ci/L)	Water (L/h)	Tritium (Ci/d)	Tritium (Ci/L)	Recycle acid (Ci/L)	Process water (Ci/L)	Vessel off-gas (Ci/L)
I	11 351	425	0.0016	0	0	0	0.0016	0	0.0013
II	160	53	0.014	1 137	372	0.014	0.014	0.014	0.014
III	896	28	1.3×10^{-3}	2	397	8.3	0.0013	0.0013	0.0013
IV	1 787	28	6×10^{-4}	125	397	0.13	0.0017	0.00013	0.0015
V	1 141	28	1×10^{-3}	12	397	1.38	0.014	0.014	0.014
VI	5 698	17	1.2×10^{-4}	366	408	0.046	0.047	0.046	0.00066

in tritium as those from the options (LWR-II and LMFBR-II) that employ water recycle only.

Confinement of the tritium to the first cycle (cases LWR-VI and LMFBR-VI) appears reasonably economical. However (see Tables 4.15 and 4.16), it produces a large volume of tritiated water for disposal, and it sends very concentrated tritium solutions to the chemical make-up areas.

4.1.6 Protection of Reprocessing Plant Personnel

The several retention options along with the base cases fall into two distinct categories with respect to concentration of tritium in water recycled to the chemical make-up areas of the plant as indicated and emphasized above.

As Table 4.15 shows, case LWR-IVA recycles water with 1.3 Ci/m^3 and case LWR-III recycles water with 7.4 Ci/m^3 . The base-case LWR-I uses fresh water with essentially no contained tritium. Cases LWR-II and LWR-V, on the other hand, recycle water with 182 Ci/m^3 , and case LWR-VI recycles water with 300 Ci/m^3 . The several options for LMFBR reprocessing (see Table 4.16) show a similar pair of categories, but at substantially lower tritium concentrations. The highest concentration in LMFBR recycle water, that of case LMFBR-VI, is 46 Ci/m^3 .

None of the recycle water streams will be completely free of radionuclides other than tritium; as a consequence, all will require precautions for protection of plant personnel. However, tritium will be virtually the only radionuclide capable of vaporization with the recycle water in the event of a leak or a spill. Tritiated water vapor (see Sect. 5) in the atmosphere is taken into the human body both by inhalation and by absorption through the skin. Exposure of plant personnel, as a worst-case example, to a large spill of recycle water at 300 Ci/m^3 of tritium (from LWR-VI) would (at a normal breathing rate of $22 \text{ m}^3/\text{d}$ of air and with skin absorption at 50% that of inhalation) obtain in ~ 12.5 min the maximum permissible body burden of 1 mCi. In contrast, the recycle stream from LWR-III (voloxidation) would give the permissible body burden in ~ 8.5 h, and that from LMFBR-VI, which is the worst of the LMFBR retention cases from this point of view, would require 82 min. If the spill was small (1 ft^2 area), such as would result from a minor valve leak, then the concentration in the air 5 ft above the floor of a normally ventilated room would be less than the 10 CFR 20¹⁵ limit for all concentrations shown except LWR-VI at 300 Ci/m^3 . It is

presumed that leaks larger than this would be repaired and that these smaller leaks would be cleaned up or flushed to the suspect drain system.

It is obvious that for many areas in each of these plants, other radionuclides dominate personnel protection considerations, and in these areas, tritium contributes relatively little hazard to plant employees. Any plant that recycles acid (and water) will, as a minimum, have to treat this material as "suspect" or low activity.¹⁶ In the section on reprocessing, the *Tritium Control Technology* handbook states:

In most cases, the tritium-containing streams also contain some other isotope of greater radiological hazard. Therefore, control of tritium usually is accomplished as a result of controlling something else and is not a direct result of a control measure directed mainly at tritium.

However, for those few areas involving recycle water (where tritium represents the significant hazard), it is equally obvious that special precautions must be taken against leaks and spills of tritiated water. Such precautions obviously include:

1. The tanks receiving or using recycle water should be enclosed by walls that form a ventilation barrier such that air flows from the uncontaminated to the contaminated area.
2. The enclosed tank area should be diked in a manner such that a rupture of the largest tank will be contained. The diked area should be drained back to the process area via the pipe encasement.
3. The air changes should be in the range of 10 to 15 per hour.
4. All operations should be performed from outside the enclosed area by means of solenoid valves and electrical controls for motors.
5. Equipment should be thoroughly flushed out and the enclosed area cleaned before any contact maintenance is performed.
6. All pipes that carry contaminated water through inhabited areas should have welded fittings.
7. The tanks should be vented via a separate system discharging back into the process area.
8. Air monitoring instruments should be installed in the enclosed area to give warning of the presence of airborne tritium.
9. A radiation monitor and alarm should be installed on the incoming recycle water line to warn of any unexpected breakthrough of fission products (other than tritium) from the process.

It is less obvious, without a detailed design of plants using each option, how much such safety provisions would add to the incremental costs since base-case plants must contain safety features to protect personnel. It is even less obvious what the cost differentials for these personnel protection features of the water recycle system should be, for example, for LWR-III (voloxidation) with recycled water at 7.4 Ci/m^3 vs LWR-II or LWR-V with recycled water at 182 Ci/m^3 . The economic evaluations and costs presented above have

assumed that the same features would be required for all options and that the cost differentials among the several retention options would appear trivial. These similar features would be required because the protective features are attributed to the recycle of trace fission products in the recycle streams and not a result of recycle of tritium.

4.2 Options for Storage and Disposal of Tritium

If retention of tritium by the reprocessing plants is practiced, it will be necessary to isolate this isotope from the biosphere for periods sufficient to permit a large fraction of it to decay. Such isolation could be accomplished by relatively long-term storage or, in principle, by any of several methods for permanent disposal. The options that are considered in this study are listed and briefly described in Table 4.17. The preferred isolation method

Table 4.17. Tritium storage and disposal options considered in this study

Number	Disposal option Description	Storage or disposal form	Storage or surveillance time (years)	Applicable to retention option number
A-I	Double-wall tank storage	Liquid H ₂ O	50	All
A-II	Double-wall tank storage	Liquid H ₂ O	100	All
B-I	Shallow trench burial	Drummed concrete	150	LWR-II; LMFBR-II, IV, and VI
C-I	Concrete in clay matrix ^a	Poured concrete	0	LWR-I and II; LMFBR-I, II, IV, and VI
C-II	Concrete in clay matrix ^a	Poured concrete	150	LWR-III, IV, V, and VI; LMFBR-III and V
C-III	Concrete in clay matrix ^a	Drummed concrete ^b	150	LWR-III and V; LMFBR-III and V
D-I	Deep-well disposal	Liquid H ₂ O	0	All
E-I	Shale hydrofracture	Concrete grout	0	All but LWR-I or LMFBR-I
F-I	Geologic disposal ^c	Canisters of concrete		LWR-III and V; LMFBR-III and V
G-I	Seabed disposal ^c	Canisters of concrete	0	All but LWR-I or LMFBR-I

^aBuried at intermediate depth above water table.

^bDrummed concrete emplaced in a massive concrete monolith.

^cCould be used for all retention options but is clearly uneconomical for large volumes.

for each case is a function of many factors. Some of these depend on detailed assessments, beyond the scope of this study, of possible hazards to the public through leakage of tritium from the isolation facility. Key factors considered in this study include the specific activity, the volume of the tritiated water for disposal, and the rate of tritium release from the reprocessing plant stack.

As indicated in Sect. 2.2.4, little, if any, guidance is available from the Nuclear Regulatory Commission or from the Environmental Protection Agency as to what approaches or what limits are acceptable for tritium disposal. It should be noted, however, that if the disposal guide¹⁷ of Table 4.17 was adopted, none of the concentrates obtained from LWR and LMFBR cases II through VI are eligible for waste class E (unrestricted release), and that, in addition, none would become so after storage for 50 years.

Policy decisions are needed to determine whether dispersal, permanent disposal, or engineered storage will be required and, especially, whether the amount of tritium in any controlled disposal will be concentrated or mass limited. The tritium control strategy that can be applied may be dictated by the policy established for storage, dispersal, or disposal.

4.2.1 Options for Interim Storage

Tank storage. Tank storage of high level wastes from military and from some commercial operations has been practiced since the beginning of spent fuel reprocessing in the United States and in Europe. Storage of alkaline high level wastes in first generation, single-walled steel tanks has led to leakage at Hanford and Savannah River and to losses to the subsurface soil at Hanford. However, subsequent experience with carefully constructed double-walled tanks has been excellent. Tanks for high level waste have recently been constructed at both Hanford and Savannah River, with an anticipated tank lifetime of 50 years. Storage of tritiated water in such tanks, with essentially no other radioactivity, should present fewer problems. No cooling is required and no corrosive chemicals are present, although care must be taken (as with the high level waste) to recombine or otherwise control the radiolytic decomposition products of water. These double-walled, below-grade, steel tanks should be capable of storage of tritiated water for at least 50 years.

Storage for 50 years would reduce the tritium concentration to 6.0% of its initial value. For this study, it is assumed that the tritiated water is released to the atmosphere by vaporization from the original facility stack after the 50-year storage period. The added dose commitment to the effected population from this release must be combined with that from the tritium released during the original processing operation.

The number and size of the tanks are obviously functions of the volume of tritiated water to be stored. Moreover, extra tanks must be available at all times in the event that a leak should develop. The choices as to size, number, and extra capacity are somewhat arbitrary as is the choice of construction scheduling. In general, an overcapacity of 20% of the stored volume is adjudged sufficient except for the most concentrated solutions (from cases LWR-III and LMFBR-III) where a 50% overcapacity is assumed. It is generally most economical to construct the tanks in batches of 5 to 10. Table 4.18 shows suggested quantities, sizes, and schedules for the tanks for the 50-year storage options.

Storage of the tritiated water for longer periods is, of course, possible in principle. Storage for 100 years before release would leave <0.4% of the tritium undecayed. It is conceivable that storage of essentially pure tritiated water in tanks such as those described might permit a tank life of that magnitude. It seems more prudent, however, to assume that replacement of the entire complement of tanks would be required after 50 years. It is, in addition, obvious that maintenance and surveillance forces would be required for the tank farm over that long period. It is also unlikely that the required stack and vaporization equipment would be serviceable, without major maintenance, after such a time span. For this study, it has been assumed that two sets of tanks would be required with the second set to be constructed 50 years after the original ones (Table 4.18).

Nearly all the disposal options will require short-term storage of the tritiated water. Such storage might be safely accommodated in somewhat less expensive tanks, but this study assumes that the uncooled, double-walled, below-grade, steel tanks anticipated to be suitable for 50-year storage will be used for whatever short-term storage is required.

Table 4.18. Tritiated water volumes, tank sizes, and construction schedules for 50-year storage

Case	Tritiated water volume (m ³)		Tank size (m ³)	Number of tanks	Construction schedule
	Per year	Total			
LWR-I					
LWR-II	3 900	78 000	4 000	24	6 at year 0, 6 at 5-year intervals
LWR-III	14.4	290	50	9	5 at year 0, 4 at year 12
LWR-IV ^a	900	18 000	3 000	8	5 at year 0, 3 at year 12
LWR-V ^b	86.5	1 730	300	8	5 at year 0, 3 at year 12
LWR-VI	2 700	53 200	4 000	18	6 at year 0, 6 at year 7 6 at year 14
LMFBR-I					
LMFBR-II	8 200	164 000	4 000	50	5 at year 0, 5 at 2-year intervals
LMFBR-III	14.4	290	50	9	5 at year 0, 4 at year 12
LMFBR-IV	900	18 000	3 000	8	5 at year 0, 3 at year 12
LMFBR-V	86.5	1 730	300	8	5 at year 0, 3 at year 12
LMFBR-VI	2 600	52 600	4 000	18	6 at year 0, 6 at year 7 6 at year 14

^aCase numbers LWR-IVA and IVB are essentially identical.

^bCase numbers LWR-VA and VB are essentially identical.

Storage as solid. Tritiated water could be converted to a solid and given interim storage before final disposal. Many options for fixation of tritium that might be suitable for permanent disposal have been described.¹⁸⁻²¹ Incorporation of tritiated water into cement provides one way of preparing tritium for permanent disposal. Additional water retaining solids, such as silica gel, clays, and molecular sieves can be incorporated in the mix. Asphalt coatings and polymer impregnation have been shown to reduce the rate of leaching of tritium from the cement.

Methods of packaging tritiated wastes have been described.²² Rhinehammer and Mershad²³ would introduce tritiated solids into a thick-walled, 103-L (27-gal) polyethylene drum that is sealed and placed in a sealed asphalt-lined, 114-L (30-gal) stainless steel drum, and this in turn is contained in a sealed asphalt-lined, 209-L (55-gal) stainless steel drum. These operations may be carried out in enclosed systems and are therefore suitable for high levels of tritium concentration.

A recent review²⁴ gives the opinion:

For retention or immobilization, incorporation of tritiated water in cement blocks in containers is currently favoured unless the tritium has been captured by a high enrichment process. In this case, immobilization as zirconium hydride is favoured, although many other immobilizing matrices are being investigated.

For some of the final disposal options (deep-sea disposal, for example), interim storage as drummed solid would be necessary. It should be noted, however, that relatively short-term

storage does not promise to simplify the ultimate disposal problem. Incorporation of even the most dilute (14 Ci/m^3 of tritium) concentrate in cement (yielding $\sim 8 \text{ Ci/m}^3$ of cement) would yield a waste that would have to be stored for 90 years to decay to the level ($5 \times 10^{-2} \text{ Ci/m}^3$) recommended in Table 4.17 for unrestricted release.¹⁷

4.2.2 Options for Terminal Disposal

A variety of final disposal options for radioactive wastes have been practiced or are being investigated. Some classes of radioactive wastes are presently disposed of by trench burial, near-shore discharge to the sea, pumping into deep wells, and shale fracturing (hydrofracture). In addition, disposal by incorporation into concrete monoliths at intermediate depth and by deposition in geologic repositories are under active investigation, and disposal as solid waste to the deep oceans has been practiced. Nearly all of these options could, in principle, be used for ultimate disposal of tritiated water. Details of these disposal methods and their economics are included as Appendix C.

4.2.3 Costs for the Disposal Options

Costs of the several disposal options are, as were the costs of the retention options described in Sect. 4.1.3, presented in first quarter 1980 dollars. As in Sect. 4.1.3, the uniform annual cash flow method has been used, and all cash flows associated with the investment have been converted into an equivalent uniform annual amount. Several disposal options effectively deal with the tritiated water produced by the retention system as it is produced. For these options, the disposal plant life is effectively 20 years, and the capital costs have been converted to the equivalent annual cost (at assumed interest rates of 7.5, 12.0, and 15.0% as in Sect. 4.1.3), assuming no salvage value at the end of 20 years. Other disposal options (such as long-term tank storage or disposal by intermediate depth burial) represent investments for a period far greater than the 20 years or entail surveillance and maintenance activities for a long period. For such disposal options, the uniform annual costs are computed (again at the three different interest rates) for the longer period that the disposal or storage system must be used and manned.

Capital costs developed in this study include direct costs of material and labor as well as distributable field costs and home office costs (see Appendix B). They do not include local taxes, insurance, interest during construction, and costs, if any, of facility decommissioning. Wherever possible, the estimated capital and operating costs are based on actual experience, with allowance (at 10% per year) for inflation. If, as is the case for some options, no applicable experience exists, estimates are based (with allowance for inflation) on available published results from other studies.

Costs for long-term tank storage. Capital costs for long-term storage in tanks are based on recent experience.²⁵ The costs expected for 12 double-walled tanks, of 3785-m^3 (10^6 -gal) capacity, placed 2.5 m below ground level, are expected to be $\$978/\text{m}^3$ with a 10% contingency. This cost includes the necessary piping, valves, etc. This cost has been accepted as reasonable for tanks whose service life with tritiated water should exceed 50 years (but not reach 100 years); costs of tanks of other sizes have been scaled using tank volume to the 0.7 power. As indicated in the tank storage section above, extra tanks to the extent of 20% of the inventory are anticipated to be necessary for the more dilute tritiated waters with a 50% excess for the three most concentrated solutions (options LWR-III,

LWR-V, and LMFBR-III). Table 4.18 shows the number, size, and construction schedules assumed for the tanks required for 50-year storage of concentrates from the several retention options both for the LWR and the LMFBR reprocessing plants.

Operating costs are more difficult to obtain for such a facility. The breakdown of talents and personnel required for any of the cases is similar to that required for interim case activities for layaway of a fuel reprocessing plant. Annual costs for these activities were determined by Battelle Pacific Northwest Laboratory for the Nuclear Regulatory Commission in 1975 dollars.²⁶ With adjustments for items not required, conversion to 1980 dollars, and addition of a 50% contingency, operating costs are 1.1×10^6 \$/year in 1980 dollars for the case LWR-II option with 24 tanks in the system. We have assumed that annual operating costs for the tank storage system can be approximately scaled by the 0.5 power of the number of tanks.

The capital cost, annual operating cost, and uniform annual cost (at 7.5% interest) for such storage systems are shown for each of the retention options in Table 4.19, along with numbers and sizes of tanks and the average cost (in \$/Ci) for decay of the tritium over the 50-year period. Table 4.20 shows the effect of higher assumed interest rates on the uniform annual cost and on the cost of tritium decay. The annual operating costs, which will be incurred over the entire 50-year period, would seem to be sufficient to include maintenance and repair of the vaporizer and stack system that are required for ultimate disposal by vaporization of the stored water to the atmosphere. As indicated above, such vaporization after 50 years would result in dispersal of 6% of the tritium originally stored.

Table 4.21 shows similar cost data for tank storage of tritium for 100 years before release. It has been assumed (although this may perhaps be pessimistic) that such storage will entail replacement of each tank and transfer of its contents at the end of 50-year life. Again, it is assumed that the annual operating cost (persisting in this case for 100 years) will suffice to maintain and use the vaporizer-stack disposal system. Storage for 100 years would, of course, result in the ultimate release of some 0.36% of the tritium originally stored.

It is clear from examination of Tables 4.19 through 4.21 that tank storage (given that tanks of this quality are required and that annual surveillance and maintenance costs are at these levels) is a relatively expensive option. However, it must be noted that some previous studies have found this option markedly less expensive. For example, in a study of options for disposal of tritiated water from wells producing natural gas after stimulation with nuclear explosives, Arnold et al.²⁷ found tank storage to be relatively inexpensive. Envisioned were some 240 producing wells, each supplied with a very large 100 000 bbl or 16 000 m³ volume), single-walled, carbon steel tank placed above ground within a retaining earthen dike and with heaters and insulation to prevent freezing of the contents. Their estimate suggested that such tanks could be built for ~\$2.50/barrel (\$15/m³) in 1972 dollars. Inflation might raise this value to \$33/m³ in 1980 dollars, but it remains a factor of 20 below the value we would estimate for a tank of 16 000 m³ and of the quality assumed necessary for this study. The enormous quantities of water that they were to store was assumed to be relatively dilute (0.05 to 0.175 Ci/m³), and these inexpensive tanks may have sufficed for the situation envisioned by Arnold et al.²⁷ However, the least concentrated solution proposed for storage by the present study (that from case LMFBR-II) contains 14 Ci/m³ of tritium and all the LWR cases contain more (and most contain much more) than

Table 4.19. Cost of tank storage for tritium concentrates: 50 years at 7.5% interest

Case	Number of tanks	Tank size (m ³)	Cost per tank (million \$)	Total tank cost (million \$)	Operating cost (million \$/year)	Uniform annual cost (million \$/year)	Average tritium decay (10 ⁵ Ci/year)	Cost of decay (\$/Ci)	Tritium remaining after 50 years (10 ⁵ Ci)
LWR-II	24	4000	4.4	106	1.1	9.34	2.65	35.40	8.5
LWR-III	9	50	0.19	1.71	0.68	0.81	2.90	2.80	9.3
LWR-IV ^a	8	3000	3.6	28.8	0.64	2.86	2.90	9.90	9.3
LWR-V ^b	8	300	0.72	5.76	0.64	1.08	2.90	3.70	9.3
LWR-VI	18	4000	4.4	79.2	0.96	7.06	3.0	23.70	9.5
LMFBR-II	50	4000	4.4	220	1.6	18.56	0.42	442.30	1.33
LMFBR-III	9	50	0.19	1.71	0.68	0.81	0.45	18.10	1.43
LMFBR-IV	8	3000	3.6	28.8	0.64	2.86	0.45	63.60	1.43
LMFBR-V	8	300	0.72	5.76	0.64	1.08	0.45	24.00	1.43
LMFBR-VI	18	4000	4.4	79.2	0.96	7.06	0.46	153.40	1.47

^aLWR case numbers IVA and IVB are essentially identical.

^bLWR case numbers VA and VB are essentially identical.

Table 4.20. Cost of tank storage for tritium concentrates:^a
50 years at 12 and 15% interest

Case	With 12% interest		With 15% interest	
	Uniform annual cost (million \$)	Cost of decay (\$/Ci)	Uniform annual cost (million \$)	Cost of decay (\$/Ci)
LWR-II	13.9	52.30	17.0	64.20
LWR-III	0.89	3.10	0.94	3.25
LWR-IV ^b	4.10	14.15	4.96	17.10
LWR-V ^c	1.33	4.60	1.50	5.20
LWR-VI	10.50	35.00	11.90	39.60
LMFBR-II	28.1	668.80	34.6	824.50
LMFBR-III	0.89	19.80	0.94	20.90
LMFBR-IV	4.10	91.10	4.96	110.20
LMFBR-V	1.33	29.60	1.50	33.30
LMFBR-VI	10.50	228.30	11.90	258.70

^aFor each case: Number of tanks, tank size, tank cost, average tritium decay rate, and amount of stored tritium remaining are those shown in Table 4.19.

^bLWR case numbers IVA and IVB are essentially identical.

^cLWR case numbers VA and VB are essentially identical.

Table 4.21. Cost of tank storage for tritium concentrates:^a 100 years at 7.5% interest

Case	Total tank cost (million \$)	Operating cost (million \$/year)	Uniform annual cost (million \$/year)	Average tritium decay (10 ⁵ Ci/year)	Cost of decay (\$/Ci)	Tritium remaining after 100 years (10 ⁴ Ci)
LWR-II	212	1.1	17.0	1.41	120.50	5.1
LWR-III	3.42	0.68	0.937	1.54	6.10	5.6
LWR-IV ^b	57.6	0.64	4.96	1.54	32.20	5.6
LWR-V ^c	11.52	0.64	1.50	1.54	9.75	5.6
LWR-VI	158.4	0.96	12.85	1.58	81.30	5.7
LMFBR-II	440	1.6	34.6	0.223	1500	0.80
LMFBR-III	3.42	0.68	0.937	0.238	39.40	0.86
LMFBR-IV	57.6	0.64	4.96	0.238	208.40	0.86
LMFBR-V	11.52	0.64	1.50	0.238	63.02	0.86
LMFBR-VI	158.4	0.96	12.85	0.245	525.0	0.89

^aAssume tank replacement (see Table 4.19 for number of "active" tanks) at a 50-year end-of-life.

^bLWR case numbers IVA and IVB are essentially identical.

^cLWR case numbers VA and VB are essentially identical.

180 Ci/m³. We are, accordingly, of the opinion that the concentrates of Table 4.19 through 4.21 would require more expensive tanks than those proposed by Arnold et al.²⁷

A recent study²⁴ by the Organization for Economic Cooperation and Development of the Nuclear Energy Agency (NEA) seems also to find tank storage of tritium somewhat less expensive than does this study. For example, the NEA estimates that tank storage of a solution containing 40 TBq/m³ (1080 Ci/m³) of tritium for 40 years would entail an annual cost of \$25 000 per GW(e) year corresponding to $\sim 1.0 \times 10^6$ \$/year for an LWR fuel reprocessing plant of this study. If we scale our case LWR-IV (whose concentrate carries 860 Ci/m³) to the 1080 Ci/m³ concentration, a total of seven tanks of 3000 m³ capacity would seem to be needed. Capital costs for the tanks would be $\sim 25.2 \times 10^6$, and operating costs would be near 6×10^5 \$/year. If, as we have not done for this study, we use the NEA method²⁴ of determining crude annual costs by dividing the capital cost by the facility lifetime and adding the annual operating cost, we would arrive at an annual cost of 1.23×10^6 \$/year, or some \$31 000 per year per GW(e) year, for tritium storage. This agreement must be considered reasonable. However, similar estimates for a 40-year storage of 1 TBq/m³ (27 Ci/m³) solution in our tank systems (requiring half the storage volume of case LMFBR-II) would entail a capital outlay of $\sim 1.1 \times 10^8$ and annual operating costs of $\sim 1.1 \times 10^6$ \$/year. A crude annual cost would be 3.85×10^6 for the plant storage system, or $\sim \$97$ 000 per GW(e) year. The NEA estimates the much smaller value of \$20 000 per GW(e) year. It is clear that NEA assumes that less expensive tanks will suffice for more dilute tritiated waters.²⁴

Near-surface trench burial. Only a few of the solutions produced by the several retention options may be adjudged suitable for near-surface burial in trenches. Case numbers LWR-II, LMFBR-II, LMFBR-IV and LMFBR-VI yield concentrates producing concretes that qualify for class D burial.

It is obvious that the tritiated waters must be fixed and canned before burial. Concrete is clearly the most economical means for fixation of the very large volumes of water involved. For this study we have assumed that the concrete is cast into 55-gal steel drums in a facility specially designed to provide adequate protection for the disposal crews. Table 4.22 shows the volumes of concrete involved, the capital and operating costs of the concrete fixation facility, and the annual costs of the trench storage facility. Estimates of capital costs include a 35% contingency; steel drums were assumed to cost \$40 each.

Actual surface burial storage experience comes from two sources. The Savannah River Plant (SRP) estimate of surface storage cost is \$6.30/ft³ in 1980 dollars. The Chem-Nuclear Facility at Barnwell currently quotes a surface storage cost of \$6/ft³ plus \$0.75 for the perpetual care fund and \$0.78 for the decommissioning fund — a total of \$7.53/ft³. For the present calculations, the SRP value plus a contingency of 25% was used giving a one-time storage fee of \$7.88/ft³, or \$52 per 55-gal drum.

It is clear from Table 4.22 that such disposal costs are very high for the dilute, large volume options such as LMFBR-I and LWR-I. Uniform annual costs are not alarmingly high for the more concentrated effluents, such as those from option LMFBR-IV, but the costs per curie buried are appreciably higher than those from tank storage (Table 4.21) and for several of the other disposal options. Since surface trench burial appears to be one of the more expensive options for the dilute tritiated waters and might not be permitted for the

Table 4.22. Costs of near-surface trench burial for qualified^a tritiated concretes

Case	Annual concrete volume (m ³ /year)	Annual requirement 50-gal drums (number/year)	Facility capital cost (million \$)	Annual operating costs (million \$/year)			Uniform annual cost ^b (million \$/year)	Dollar per curie buried	
				Cost of drums	Trench storage	Concrete facility			Total
LWR-I	72 000	17 500	110	14	20	19	53	63.8	90.50
LWR-II	7 500	1 800	22	1.45	2.1	2.5	6.05	8.21	11.65
LMFBR-I	150 000	36 000	180	29	42	41	112	130	1165.00
LMFBR-II	15 000	3 600	36	2.9	4.2	5.1	12.2	15.7	140.70
LMFBR-IV	1 600	385	7.5	0.31	0.45	0.86	1.62	2.35	19.70
LMFBR-VI	4 700	1 150	16	0.90	1.35	1.7	3.95	5.52	45.10

^aCases LWR-III, -IVA, -VA, -VB, and -VI and LMFBR-III and -V produce concretes that cannot qualify for near-surface trench burial under the Rogers criteria.

Source: V. C. Rogers, *A Radioactive Waste Disposal Classification System*, NUREG/CR-1005, U.S. Nuclear Regulatory Commission Report, Washington, D.C. (September 1979).

^bAssuming capital costs are amortized over 20 years at 7.5% annual interest rate.

more concentrated ones, we have not examined the effect of varied interest rates upon the costs.

The NEA seems not to have considered shallow burial of tritiated concrete from high-volume, dilute tritiated waters.²⁴ They did include an estimate of $\sim 1 \times 10^6$ \$/year for storage of triply contained concrete blocks in a concrete-lined pit. However, since the volume of concrete seems not to be stated, we are unable to make a cost comparison.

Costs for intermediate depth burial. There is at present no facility in operation for burial of wastes incorporated in concrete monoliths at intermediate depths above the water table. Cost estimates for this emplacement method, which includes a 35% contingency, have been extrapolated and interpolated from an on-going study at Savannah River Laboratory that examines use of such a method for burial of large volumes of modestly contaminated salt.²⁸ In this method it is assumed that the burial area will be excavated and filled with compacted, essentially impermeable clay. Suitable trenches (capable of holding 500 m³ of concrete) will be cut in the compacted clay, and into which the tritiated concrete will be poured and subsequently covered (see Appendix C). Table 4.23 shows details as to concrete volumes and related data, specific activities of the concretes, and estimated capital and operating costs for the disposal facility.

It should be noted that six of the options considered (LWR-I, LWR-II, LMFBR-I, LMFBR-II, LMFBR-IV, and LMFBR-VI) produce concretes that can qualify for class C burial at intermediate depth so that no long-term surveillance is required. It should also be noted that the tritiated waters from the four options producing the most concentrated solutions (LWR-III, LWR-V, LMFBR-III, and LMFBR-V) are assumed to be diluted sufficiently to produce a single 500-m³ monolith each year, and that 2.5×10^5 \$/year has been added to the operating costs to provide for the modest cost of long-term surveillance of the site where this is required. It is clear from Table 4.23 that application of this procedure to the more concentrated solutions leads to estimated costs well below those of tank storage. Assuming, as is not guaranteed, that such concentrated tritiated concretes can be safely emplaced in such a facility, this concept would seem to warrant further study.

It seems certain that a facility can be designed for safe production of concretes prepared with highly tritiated waters and for safe emplacement of these concretes in drums. Accordingly, it should be possible to prepare highly tritiated concrete, pour it into subsequently sealed drums, emplace the drums on concrete pads within the clay trench, and pour normal concrete to seal the drums within large monoliths. Table 4.24 shows the volume of drummed concrete, the volume of monolith, and the capital and operating costs associated with emplacement of the four most concentrated tritiated waters. It should be noted that all of these cases would require surveillance for 150 years;¹⁷ the operating cost estimates contain \$250 000/year for this surveillance.

The uniform annual costs and the price per curie for disposal are, as anticipated, higher for the drummed concrete than for directly cast concretes. However, as suggested above, such direct casting of highly tritiated concretes might not be feasible, and the higher costs of this alternative may be necessary. Although considerable study and testing of such disposal would be necessary, it seems likely that the contained tritium would be safely immobilized by such a procedure.

Table 4.23. Costs of intermediate depth burial of tritiated waters as 500 m³ concrete monoliths in clay matrix

Case	Annual concrete volume (m ³ /year)	Concrete specific activity (Ci/m ³)	Trenches required per year	Total land commitment (acres)	Facility capital cost (million \$)	Annual operating cost ^a (million \$/year)	Uniform annual cost ^b (million \$/year)	Dollar per curie buried
LWR-II	7 500	94 ^c	15	17	22	2.5	4.66	6.60
LWR-III	500 ^d	1 540	1	1.2	3.3	0.86	1.17	1.52
LWR-IV	1 600	480	3+	4	7.5	1.11	1.83	2.40
LWR-V	500	1 540	1	1.2	3.3	0.86	1.17	1.52
LWR-VI	4 900	160	10-	11	16	1.95	3.52	4.45
LMFBR-II	15 000	8 ^c	30	34	36	5.1	8.63	77.30
LMFBR-III	500 ^d	240	1	1.2	3.3	0.86	1.17	9.80
LMFBR-IV	1 600	74 ^c	3+	4	7.5	0.86	1.60	13.40
LMFBR-V	500 ^d	240	1	1.2	3.3	0.86	1.17	9.80
LMFBR-VI	4 700	24 ^c	10-	11	16	1.7	3.27	26.70

^aIncludes \$250 000/year to pay for 150-year surveillance for those cases required.

^bAssuming capital amortized over 20 years at 7.5% interest.

^cThese cases qualify for class C burial; no surveillance required.

^dThese concentrated solutions are assumed to be diluted with sufficient tritium-free water to yield these volumes of concrete.

Table 4.24. Costs for burial of concentrated tritiated waters as dunned concrete emplaced in concrete monoliths at intermediate depth in clay matrix

Case	Drummed concrete volume (m ³ /year)	Monolith volume (m ³ /year)	Trenches required per year	Total land commitment (acres)	Facility cost (million \$)		Operating costs (million \$/year)			Uniform annual cost (million \$/year)	Disposal cost (\$/Ci)
					Burial	Drumming	Drumming facility	Burial facility	Total		
LWR-III	25 ^a	150	0.3	0.5	1.9	2.5	0.505 ^b	0.75 ^c	1.205	1.69	2.20
LWR-V	160 ^d	1000	2	2	4.6	3.3	0.73 ^e	1.29 ^d	2.02	2.79	3.62
LMFBR-III	25 ^a	150	0.3	0.5	1.9	2.5	0.70 ^b	0.75 ^d	1.70	1.69	12.93
LMFBR-V	160 ^d	1000	2	2	4.6	3.3	0.73 ^e	1.29 ^d	2.02	2.79	21.34

^aRequires one hundred twenty 50-gal drums per year.

^bIncludes \$31 000/year for drums.

^cRequires seven hundred seventy 50-gal drums per year.

^dIncludes \$250 000 per year to pay for 150-year surveillance.

^eIncludes \$5 000/year for drums.

Costs for discharge to deep wells. The Argonne National Laboratory examined on-site deep-well disposal of tritiated waters for the reprocessing facility (never operated) at Morris, Illinois.²⁹ This installation was designed for a disposal rate of 35 gal/min (57 000 m³ per 300-d year), which is sufficient for all case options considered here. The well depth was 6000 ft (1900 m), and injection was to be accomplished at 3000 psi. The cost estimate included well construction, piping and pumping equipment, and a pretreatment area that included necessary surge capacity; the estimate did not include cost of appreciable storage capacity. Given that the disposal stratum can accommodate high injection rates and that no appreciable storage capacity is required, the capital cost of the facility should be essentially independent of disposal rate. Capital costs, updated for inflation and with a 50% contingency included, are estimated to be $\$5 \times 10^5$. On that same basis, operating costs appear to be near $\$70\,000/\text{year}$.

Table 4.25 presents the costs of deep-well injection, the disposal cost (in dollars per curie) for all the retention options, and the three assumed rates of interest. It is clear that

Table 4.25. Cost of deep-well injection for disposal of tritiated waters

All retention cases		Uniform annual cost (million \$/year)		
Capital cost disposal facility (million \$)	Operating cost (million \$/year)	7.5% ^a	12% ^a	15% ^a
0.50	0.070	0.120	0.137	0.150

Case	Disposal cost (\$/Ci)		
	7.5% ^a	12% ^a	15% ^a
LWR – all	0.170	0.194	0.212
LMFBR – all	1.08	1.74	1.90

^a Assumed interest rates.

such disposal, if a suitable well can be demonstrated *on-site*, is by far the least expensive of the options. Indeed, given that the well is capable of the required injection rates, it is clear that only options LWR-II and LMFBR-II would need to be considered.

It must be noted, however, that use of this procedure as described requires that the injection system function all the time that the plant is in operation. If some interim storage capacity must be added to avoid plant downtime, the costs would increase substantially. Addition, for example, of a single 4000 m³ tank (such as those of Table 4.19) to the system would provide safe interim storage for a month of injection well downtime and would raise the uniform annual cost to $\$0.48 \times 10^6$ (at 7.5%) and the cost of injection to $\$0.68/\text{Ci}$. This would still be the most attractive disposal option, but it would be cheaper (and probably sufficient) to duplicate the injection system (maximum capital outlay $\$500\,000$) instead.

The NEA estimates (by procedures described above) the annual cost of on-site injection of a 1 TBq/m³ (27 Ci/m³) corresponding to about 0.5 times the volume of our retention case LMFBR-II to be 0.18×10^6 \$/year with some \$10 000/year (presumably a capital cost of \$300 000 for their 30-year lifetime plant) for interim storage tanks.²⁴ This would seem to be slightly higher than the present estimate, but the agreement seems reasonable. A much smaller value of \$20 000/year (with \$15 000/year of this amount for tank costs) is estimated for injection of effluent at 40 TBq/m³ (1080 Ci/m³). Although such tanks (whose use might rarely be required) might suffice in place of the more expensive ones assumed for this study, it seems highly unlikely that the costs of the injection facility (and of its operation) scale linearly with the injection rate over this fortyfold range.

It should be noted that ultimate approval of deep-well injection of tritium in these quantities in the United States must be considered to be uncertain. There has been much objection³⁰ from the environmentalists to deep-well injection of quite dilute (5×10^{-4} Ci/m³) tritiated waters at the Idaho National Engineering Laboratory.

Cost of disposal by hydrofracture. Since medium-level waste has been disposed of for years by hydrofracture at the ORNL, the costs of such operation with tritiated waters can be reasonably estimated. The costs described here are based on that experience and on the capital costs estimated³¹ for construction of a new hydrofracture facility at that site. Including a 25% contingency, capital costs are estimated to be $\$6.5 \times 10^6$ and (since all injections are assumed to include 300 m³ of tritiated water) are essentially independent of total volume to be injected. This capital cost does not include the cost of the necessary storage tanks. Storage tanks, as indicated in Table 4.26, are assumed to be of the quality and cost of those described above. In accordance with the ORNL experience³² and with a 25% contingency, operating costs (including grouting materials as well as maintenance and operations) are estimated to be \$37 500 per injection. Table 4.26 shows the costs associated

Table 4.26. Cost of shale fracturing (hydrofracture) for disposal of tritiated waters^a

Case	Annual concentrate volume (m ³ /year)	Number of injections per year	Required storage volume (m ³)	Tank size (m ³)	Tank capital cost (million \$)	Operating cost ^b (million \$/year)	Uniform annual cost ^c (million \$/year)	Disposal cost (\$/Ci)
LWR-II	3880	13	2000	1000	3.4	0.49	1.46	2.07
LWR-III	14	0.2 ^d	100	50	0.38	0.0075	0.684	0.89
LWR-IV	900	3	600	300	1.4	0.11	0.885	1.15
LWR-V	86	0.33 ^d	300	150	0.88	0.013	0.739	0.96
LWR-VI	2660	9	1500	750	2.7	0.34	1.24	1.57
LMFBR-II	8200	28	4000	2000	5.4	1.1	2.28	20.40
LMFBR-III	14	0.2 ^d	100	50	0.38	0.0075	0.684	5.74
LMFBR-IV	900	3	600	300	1.4	0.11	0.885	7.43
LMFBR-V	86	0.33 ^d	300	150	0.88	0.013	0.739	6.20
LMFBR-VI	2600	9	1500	750	2.7	0.34	1.242	10.14

^aHydrofracture facility assumed to cost 6.5 million dollars for each case.

^bTotal cost of a 300-m³ injection assumed (25% contingency) to be \$37 500.

^cFacility life - 20 years at 7.5% interest.

^dAssumed to be diluted to 300 m³ at time of injection.

with this operation. It should be noted that since the capital costs of the facility and tanks contribute appreciably to the hydrofracture costs, the cost of disposal (Table 4.27) varies considerably with the assumed cost of money.

Table 4.27. Cost of shale fracturing (hydrofracture) for disposal of tritiated waters as a function of interest rate^a

Case	Interest at 12%		Interest at 15%	
	Uniform annual cost (million \$/year)	Disposal cost (\$/Ci)	Uniform annual cost (million \$/year)	Disposal cost (\$/Ci)
LWR-II	1.82	2.60	2.07	2.95
LWR-III	0.931	1.20	0.111	1.45
LWR-IV	1.17	1.50	1.37	1.80
LWR-V	1.00	1.30	1.20	1.55
LWR-VI	1.54	2.00	1.81	2.30
LMFBR-II	2.70	24.25	3.00	27.05
LMFBR-III	0.931	7.80	1.11	9.32
LMFBR-IV	1.17	9.80	1.37	11.50
LMFBR-V	1.00	8.45	1.20	10.05
LMFBR-VI	1.57	12.85	1.81	14.80

^aAll other data are those shown in Table 4.26.

It is clear that, given a suitable shale formation at the reprocessing plant site, the uniform annual costs and the disposal costs (in \$/Ci) are lower for hydrofracture than for any other disposal option other than deep-well injection. The tritiated waters from LWR-III, LWR-V, LMFBR-III, and LMFBR-V are assumed to be diluted to a 300-m³ volume during grout manufacture at the time of injection. It should also be noted that, as discussed in Appendix C, the hydrofracture technique (with its several attractive features) may not be practicable at all reprocessing plant sites.

Costs of geologic disposal. It seems obvious that disposal of relatively short-lived isotopes, such as tritium, could not justify construction (or even enlargement) of a geologic repository. Geologic disposal should be considered only for the most concentrated tritiated waters and, even for these, only if such disposal can be accomplished without augmentation of the repository. Accordingly, we have assumed that only the concentrates from options LWR-III, LWR-V, LMFBR-III, and LMFBR-V need to be considered for geologic disposal and that the solidified and contained waste can be emplaced in repository positions interstitial to the canisters of high-level wastes.

Geologic disposal obviously requires solidification of the tritiated waters. It is clear that solidification in concrete is the least expensive and almost equally clear that no other method (preparation of zirconium hydride, for example) produces smaller volumes of solid for disposal. It is obvious, in addition, that the solidified waste must be adequately contained

for the transportation and emplacement steps. We have assumed that the tritiated concrete will be contained within 55-gal drums and that the drums will be contained within a relatively thin overpack of stainless steel sealed by welding. Transportation costs (from Oak Ridge, Tennessee, to Carlsbad, New Mexico) are assumed, with a 35% contingency, to cost \$6000 per trip involving 36 canisters in a Super Tiger Overpack (Certification of Compliance 6400). Costs of introduction into the repository are assumed to be \$9000 per canister.³³

Table 4.28 shows the costs of geologic disposal estimated on these bases for the four retention options considered. It is obvious that cost of disposal in this manner is a very strong function of waste volume and that the costs, even for the very low volume cases, are relatively high. Geologic disposal seems unlikely to prove attractive, and the operating costs (especially the repository handling costs) dominate uniform annual costs; therefore, we have not evaluated the relatively small effect of the cost of capital money.

NEA (1980) has estimated the annual cost of geologic disposal as concrete blocks prepared from the concentrate from voloxidation of the fuel (which must correspond at least roughly to our case LWR-III) to be 0.64×10^6 \$/year with 0.6×10^6 of this allocated to preparation of the blocks. Our estimate would seem to be in reasonable agreement with the cost of concrete preparation, although we would add the canisters (Table 4.28); the \$40 000 per year allotted to repository disposal by NEA²⁴ seems likely to prove quite inadequate.

Cost of sea-bed disposal. Deep-ocean (at a depth of ~5000 m) disposal of tritiated wastes seems feasible, as indicated in Appendix C. However, it involves a considerable number of operations including both land and ocean transport. For estimation of the costs of sea-bed disposal, we have again assumed that the tritiated wastes are solidified as concrete in 55-gal drums. The drums of the more concentrated wastes (those from cases LWR-III, LWR-V, LMFBR-III, and LMFBR-V) are further assumed to be contained in a relatively thin overpack of stainless steel sealed by welding. Transportation from Oak Ridge, Tennessee, to the port (assumed to be Charleston, South Carolina) is estimated to cost, with a 35% contingency, \$1800 per load of 36 drums or canisters in a Super Tiger Overpack (Certification of Compliance 6400). Such sea-bed disposal obviously requires a special receiving and loading facility at the port as well as at the on-site facility for preparation of the drummed concrete. The capital cost estimates for these facilities, for which a 35% contingency has been assumed, are shown with other data in Table 4.29.

The cost of the sea-bed disposal operation conducted by NEA in 1976 was (given that the port facility exists) \$30/t of wastes.³³ Allowance for inflation in the 1976–1980 period, and an inclusion of a 15% contingency suggests a cost of \$55/t in 1980 dollars. Using these bases and assumptions, the costs shown in Table 4.29 are estimated. It is apparent that the costs of sea-bed disposal, especially for the low volume cases, are reasonably low and that they compare favorably with those of 50-year tank storage, for example.

It should be noted, however, that the very low costs of the sea-bed disposal operation itself may be illusory, especially for the retention options producing small volumes for disposal. The \$55/t for sea-bed disposal presumably is based on a reasonable load for the disposal ship, and several years accumulation of tritiated wastes from a number of reprocessing plants (or adoption of sea-bed disposal for other classes of low-level wastes) probably would be required to achieve the low disposal costs of Table 4.30.

Costs shown for sea-bed disposal of the wastes from the high-volume options seem likely to be realizable, but they are considerably higher than those for shallow land burial

Table 4.28. Cost of geologic repository disposal of drummed concretes prepared from tritiated waters

Case	Annual concrete volume (m ³ /year)	Number of drums per year	Concrete facility capital costs (million \$)	Annual operating costs (million \$/year)			Uniform annual cost ^d (million \$/year)	Disposal cost (\$/Ci)	
				Canister costs ^a	Concrete facility operation	Transport costs ^b			Repository handling costs ^c
LWR-III	25	120	2.5	0.185	0.5	0.024	1.1	2.05	2.65
LWR-V	160	770	3.3	1.20	0.7	0.13	6.95	9.30	12.05
LMFBR-III	25	120	2.5	0.185	0.5	0.024	1.1	2.05	17.20
LMFBR-V	160	770	3.3	1.20	0.7	0.13	6.95	9.30	78.10

^a Assumes concrete contained in sealed drum and sealed stainless steel overpack at cost of \$1500 each.

^b Assumes transportation of 36 canisters at \$6000 per trip.

^c Assumes repository handling costs at \$9000 per canister.

^d With a 20-year lifetime of processing plant facilities at 7.5% interest.

Table 4.29. Cost of sea-bed disposal of drummed concrete prepared from tritiated waters

Case	Annual concrete volume (m ³ /year)	Drums required per year	Capital costs (million \$)		Annual operating costs (\$10 ⁶ /year)					Uniform annual cost ^c (million \$/year)	Disposal cost (\$/Ci)
			Concrete facility	Port facility	Concrete facility	Canisters	Port facility	Land transport ^a	Sea-bed ^b disposal		
LWR-II	7 500	36 000	22	4.4	2.5	1.45	1.8	1.8	1.25	11.4	16.20
LWR-III	25	120	2.5	0.5	0.5	0.19 ^d	0.4	0.0060	0.0041	1.39	1.80
LWR-IV	1 600	7 700	7.5	1.5	0.9	0.31	0.7	0.39	0.055	3.23	4.20
LWR-V	160	770	3.3	0.7	0.7	1.2 ^d	0.5	0.039	0.0055	2.84	3.70
LWR-VI	4 800	24 000	16	3.2	1.7	0.95	1.3	1.2	0.8	7.83	9.90
LMFBR-II	15 000	72 000	36	7.2	5.1	2.9	3.8	3.6	2.45	22.1	198.00
LMFBR-III	25	120	2.5	0.5	0.5	0.19 ^d	0.4	0.006	0.0041	1.39	11.70
LMFBR-IV	1 600	7 700	7.5	1.5	0.9	0.31	0.7	0.39	0.055	3.23	27.10
LMFBR-V	160	770	3.3	0.7	0.7	1.2 ^d	0.5	0.039	0.0055	2.84	23.80
LMFBR-VI	4 840	24 000	16	3.2	1.7	0.95	1.3	1.2	0.8	7.83	64.00

^aTransportation from Oak Ridge, Tennessee, to Charleston, South Carolina, at \$1800 per load of 36 canisters (including 35% contingency).

^bSea-bed disposal at \$55/t (15% contingency).

^cFacility lifetime – 20 years at 7.5% interest rate.

^dMore concentrated wastes assumed to be drummed and emplaced in sealed stainless steel overpack; cost \$1500 each.

Table 4.30. Summary of costs for disposal options for tritium from LWR fuel reprocessing with water recycle only (retention case LWR-II)

Disposal option Number	Description	Capital cost (million \$)	Capitalized cost ^a (million \$/year)	Operating cost (million \$/year)	Uniform annual cost (million \$/year)	Disposal cost (\$/Ci)
A-I	50-year storage	106	8.2	1.1	9.3	35.40
A-II	100-year storage	212	15.9	1.1	17.0	120.50
B-I	Trench burial	22	2.16	6.05	8.2	11.70
C-I	Monolith ^b	22	2.16	2.5	4.7	6.60
D-I	Deep-well disposal	0.5	0.049	0.070	0.120	0.17
E-I	Hydrofracture	9.9	0.97	0.49	1.46	2.07
G-I	Sea-bed disposal	26.4	2.59	7.0	11.4	16.20

^aCapitalized costs assume 7.5% interest rate in all cases.

^bMonolith at intermediate depth in clay matrix; no long-term surveillance assumed.

or for intermediate-depth burial in concrete monoliths (if these methods are applicable) and are markedly higher than those for deep-well disposal or for disposal by hydrofracture.

It appears that NEA estimated the cost of sea-bed disposal of concrete blocks prepared from the tritiated waters from voloxidation of the fuel (which should correspond crudely to our case LWR-III) to be 1×10^6 \$/year with $\$0.6 \times 10^6$ of this allotted to preparation of the blocks, and, presumably, $\$0.4 \times 10^6$ allocated to all other steps. Although not presented in detail sufficient to show the costs of the various steps, these estimates agree reasonably with the value 1.35×10^6 \$/year obtained from the data for LWR-III (Table 4.27) by dividing the capital costs by 30 (the NEA 1980 facility lifetime) and by adding the operating costs.

4.2.4 Comparisons of costs of applicable disposal options for selected retention options

Table 4.30 presents a summary of the capital and operating costs of the several disposal options that might be considered for the large volumes of tritiated water resulting from retention option LWR-II employing recycle of ~90% of the plant water. As expected, it is clear that tank storage of these large volumes for 100, or even for 50 years, is expensive. Capital costs of the tanks for 50-year storage would amount to ~7% of the cost of the entire reprocessing facility, and longer term storage would be much more expensive. None of the other disposal options would seem to entail capital costs of as much as 2% of the total facility, or to add more than ~\$7.50/kg to the cost of reprocessing heavy metal. It is clear, however, that the only disposal options that are relatively inexpensive for such large volumes of tritiated water are deep-well disposal or hydrofracture.

Tables 4.31 and 4.32 show similar cost data for cases LWR-III (voloxidation) and LWR-V (isotope separation on discharge water), both of which produce small volumes of tritiated water for disposal. Tank storage for 100 years would add approximately \$3.15/kg of heavy metal to the total reprocessing cost. The least expensive options, deep-well disposal and hydrofracture, would contribute about \$0.11 and \$0.45/kg to the cost of reprocessing heavy metal. For the intermediate (although still low) volume option LWR-V only, geologic disposal would add as much as \$6/kg of heavy metal to the reprocessing cost, although deep-well disposal and hydrofracture are the least expensive options.

Table 4.31. Summary of costs for disposal options for tritium from LWR fuel reprocessing with tritium retention by voloxidation (retention option LWR-III)

Disposal option		Capital cost (million \$)	Capitalized cost ^a (million \$/year)	Operating cost (million \$/year)	Uniform annual cost (million \$/year)	Disposal cost (\$/Ci)
Number	Description					
A-I	50-year storage	1.71	0.132	0.68	0.81	2.80
A-II	100-year storage	3.42	0.257	0.68	0.94	6.10
C-II	Monolith ^b	3.3	0.32	0.86	1.18	1.52
C-III	Monolith ^c	4.4		1.21	1.64	2.20
D-I	Deep-well disposal	0.50	0.049	0.070	0.120	0.17
E-I	Hydrofracture	6.88	0.67	0.0075	0.68	0.89
F-I	Geologic disposal	2.5	0.24	1.81	2.05	2.65
G-I	Sea-bed disposal	3.0	0.29	1.10	1.39	1.80

^aCapitalized costs assume 7.5% interest rate in all cases.

^bConcrete monolith poured in clay matrix; 150-year surveillance assumed.

^cDrummed concrete encased in monolith (ordinary concrete) poured in clay matrix; 150-year surveillance assumed.

Table 4.32. Summary of costs for disposal options for tritium from LWR fuel reprocessing with isotope separation on discharge water (retention case LWR-VA)^a

Disposal option		Capital cost (million \$)	Capitalized cost ^a (million \$/year)	Operating cost (million \$/year)	Uniform annual cost (million \$/year)	Disposal cost (\$/Ci)
Number	Description					
A-I	50-year storage	5.62	0.44	0.64	1.08	3.70
A-II	100-year storage	11.52	0.86	0.64	1.50	9.75
C-II	Monolith ^b	3.3	0.32	0.86	1.18	1.52
C-III	Monolith ^c	7.9	0.77	2.02	2.79	3.62
D-I	Deep-well disposal	0.50	0.049	0.070	0.12	0.17
E-I	Hydrofracture	7.38	0.72	0.013	0.73	0.96
F-I	Geologic disposal	3.3	0.32	8.98	9.30	12.10
G-I	Sea-bed disposal	4.0	0.39	2.44	2.83	3.70

^aCapitalized costs assume 7.5% interest rate in all cases.

^bConcrete monolith poured in clay matrix; 150-year surveillance assumed.

^cDrummed concrete encased in monolith (ordinary concrete) poured in clay matrix; 150-year surveillance assumed.

In general, the costs of the disposal options for tritium from LMFBR fuel are similar fractions of the cost of the overall plant. However, primarily because far less tritium is delivered to the plant with the fuel, the costs per unit of tritium disposal or tritium stored are much higher than those from LWR fuel reprocessing.

4.3 Summary of Costs of Combined Retention and Disposal Cases

Tables 4.33 and 4.34 summarize the uniform annual costs and the annual cost in dollars per curie of confined tritium for the several combinations of retention and storage or disposal options for plants processing LWR and LMFBR fuels respectively. These values represent syntheses of costs from Tables 4.11 and 4.12 with those from Tables 4.19 through 4.29.

Table 4.33. Costs of combined retention and disposal options for LWR fuel reprocessing^a

Retention case	Disposal option								
	A-I 50-year storage	A-II 100-year storage	B-I Trench burial	C-I or C-II Monolith (concrete)	C-III Monolith (drums)	D-I Deep-well disposal	E-I Hydro- fracture	F-I Geologic disposal	G-I Sea-bed disposal
LWR-II water recycle									
Uniform annual cost (million \$/year)	8.06	15.72	6.93	3.38 ^b	—	-1.16	0.18	—	10.12
Cost (\$/Ci)	33.60	118.70	9.80	4.80 ^b	—	-1.60	0.30	—	14.40
LWR-III voloxidation									
Uniform annual cost (million \$/year)	8.23	8.40	—	8.60 ^c	9.12 ^d	7.55	8.11	9.48	8.82
Cost (\$/Ci)	12.40	15.70	—	11.20 ^c	11.80 ^d	9.80	10.50	12.30	11.40
LWR-IV-A^e									
Uniform annual cost (million \$/year)	21.67	23.77	—	20.64 ^c	—	18.93	19.70	—	22.04
Cost (\$/Ci)	34.30	56.60	—	26.80 ^c	—	24.60	25.50	—	28.60
LWR-IV-B^f									
Uniform annual cost (million \$/year)	22.03	24.10	—	21.00 ^c	—	19.29	20.06	—	22.40
Cost (\$/Ci)	33.70	56.00	—	26.20 ^c	—	24.00	25.00	—	28.00
LWR-VA^g									
Uniform annual cost (million \$/year)	5.04	5.46	—	5.13 ^c	6.75 ^d	4.08	4.70	13.26	6.80
Cost (\$/Ci)	8.90	14.90	—	6.70 ^c	8.80 ^d	5.30	6.10	17.20	8.85
LWR-VB^h									
Uniform annual cost (million \$/year)	5.68	6.10	—	5.77 ^c	7.39 ^d	4.72	5.34	13.90	7.44
Cost (\$/Ci)	9.40	15.40	—	7.20 ^c	9.30 ^d	5.90	6.70	17.80	9.40
LWR-VI first cycle confinement									
Uniform annual cost (million \$/year)	10.01	15.80	—	6.47 ^c	—	3.07	4.19	—	10.78
Cost (\$/Ci)	27.40	85.00	—	8.20 ^c	—	3.90	5.30	—	13.60

^a7.5% interest on capital money in all cases.

^bOption C-I – poured concrete monolith in clay matrix; no surveillance assumed.

^cOption C-II - poured concrete monolith in clay matrix; 150-year surveillance assumed.

^dOption C-III – drummed concrete emplaced in concrete monolith in clay matrix; 150-year surveillance assumed.

^eCombined electrolysis catalytic exchange process on total water recycle.

^fSame as (e) but with very small atmospheric release.

^gCombined electrolysis catalytic exchange process on excess (discharge) water only.

^hSame as (g) but with very small atmospheric release.

Table 4.34. Costs of combined retention and disposal options for LMFBR fuel reprocessing^a

Retention case	Disposal option								
	A-I 50-year storage	A-II 100-year storage	B-I Trench burial	C-I or C-II Monolith (concrete)	C-III Monolith (drums)	D-I Deep-well disposal	E-I Hydro- fracture	F-I Geologic disposal	G-I Sea-bed disposal
LMFBR-II water recycle									
Uniform annual cost (million \$/year)	15.96	32.0	13.10	6.03 ^b	—	-2.48	-0.32	—	19.50
Cost (\$/Ci)	422	1480	118	54.10 ^b	—	-22.10	-2.80	—	175
LMFBR-III voloxidation									
Uniform annual cost (million \$/year)	12.01	12.14	—	12.37 ^c	12.89 ^d	11.32	11.88	13.25	12.59
Cost (\$/Ci)	112	134	—	104	107	95.20	99.80	111	106
LMFBR-IV^e									
Uniform annual cost (million \$/year)	42.03	44.13	41.52	40.77 ^b	—	39.29	40.06	—	42.40
Cost (\$/Ci)	393	538	349	343 ^b	—	330	337	—	356
LMFBR-V^f									
Uniform annual cost (million \$/year)	7.22	7.64	—	7.31 ^c	8.93 ^d	6.26	6.98	15.44	8.98
Cost (\$/Ci)	75.60	115	—	61.40 ^c	72.90 ^d	52.70	57.80	130	75.40
LMFBR-VI first cycle confinement									
Uniform annual cost (million \$/year)	10.67	16.46	9.13	6.88 ^b	—	3.73	4.85	—	11.46
Cost (\$/Ci)	185	555	74.70	56.30 ^b	—	30.70	39.70	—	93.60

^a7.5% interest on capital money in all cases.

^bOption C-I — poured concrete monolith in clay matrix; no surveillance assumed.

^cOption C-II — poured concrete monolith in clay matrix; 150-year surveillance assumed.

^dOption C-III — drummed concrete emplaced in concrete monolith in clay matrix; 150-year surveillance assumed.

^eCombined electrolysis catalytic exchange process on total water recycle.

^fCombined electrolysis catalytic exchange process on excess (discharge) water only.

Examination of Table 4.33 indicates that the retention cases (LWR-IVA, LWR-IVB, and LWR-III) that provide the lowest concentrations of tritium in the plant recycle waters fare poorly in these cost comparisons. Cases LWR-IVA and LWR-IVB are the most expensive of the lot, and they remain so in all combinations with the disposal options other than long-term storage. Voloxidation (LWR-III), which also provides recycle water containing relatively little tritium, is also among the more expensive. Retention cases LWR-VA and LWR-VB, which provide recycle water at 182 Ci/m^3 , are by comparison less expensive. However, the least expensive of these (LWR-VA) can retain and immobilize tritium for less than \$4/Ci only by use of deep-well disposal and for \$5 or less per curie only by use of hydrofracture disposal, or (if feasible) by disposal as poured concrete monoliths.

First cycle confinement (case LWR-VI), which provides the most concentrated tritiated water as recycle, is also reasonably economical. However, the volume of tritiated water for disposal is relatively large, and its disposal in deep wells or by hydrofracture is required if the combined cost is to be below \$4/Ci or \$5/Ci respectively.

It is clear from consideration of Table 4.33 that if disposal to deep wells (perhaps unlikely) or by hydrofracture (likely) can be employed, the water recycle (case LWR-II) is the most economical case and that it remains so (but with little margin) should poured concrete monoliths in a clay matrix be employed for disposal. With its large volume of tritiated water for disposal, the water recycle retention cases (LWR-II and LMFBR-II) could not compete if expensive disposal options (tank storage, geologic disposal, or sea-bed disposal) were mandated. Use of the water recycle case and of case LWR-VA or LWR-VB requires design to safely accommodate recycle water with 182 Ci/m^3 of tritium.

Examination of Table 4.34 reveals the same general pattern, but with the important exception that the costs in dollars per curie of tritium confined are quite (and probably prohibitively) high. Indeed, only the water recycle (case LMFBR-II), when combined with deep-well disposal or hydrofracture, seems likely to result in costs below about \$25/Ci retained.

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5. ENVIRONMENTAL IMPACT OF RELEASED TRITIUM

5.1 Tritium Dosimetry

Tritium decays with a half-life of 12.33 years by emission of an electron¹ (maximum energy 18 keV and average energy 5.88 keV) to form helium. External doses to man are inconsequential because of the low electron energy. However, tritium in the atmosphere and in surface waters equilibrates rapidly with the biosphere and, as a consequence, can affect man by several pathways.

Tritium inhaled as water vapor is essentially completely retained, and tritiated water vapor in air is readily absorbed through the skin. Tritium is ingested in drinking water, as the water content of foods, and as organically bound tritium in foods. Tritium taken into the body seems to be nearly uniformly distributed throughout the body water.

Tritium is eliminated rapidly from the human body. A recent publication² assumes a three-compartment model of the human body for which the effective biological half-time for tritium elimination is 9.7 d. Several publications^{1,3-5} assume a biological half-time of 10 d; that value is used in the dose estimates presented here.

A key factor in estimating a radiation dose is the quantity of radiation energy absorbed in the tissue. The basic unit is the rad, which corresponds to absorption of 100 ergs/g of tissue. Many radionuclides concentrate in particular tissues (such as the liver), and different radionuclide distribution factors (RDFs) must be assigned to evaluate the dose to individual organs from a unit intake of such a nuclide into the body. However, tritium distributes essentially uniformly in body tissues, and its distribution factor is assumed to be in unity. The relatively small decay energy (average 5.83 keV per disintegration) is completely absorbed very near its point of origin.

Consequently, a single uptake of 1 μ Ci of tritium uniformly distributed (at 1.923×10^{-11} Ci/g) within the 52 kg of wet tissues of a 70-kg man would deliver an initial dose rate of 6.69×10^{-11} rad/s. If this tritium were eliminated with a biological half-life of 10 d, the integral dose delivered by the 1 μ Ci would be 8.3×10^{-5} rad.

The International Commission on Radiological Protection and the International Commission on Radiation Units and Measurements⁶ introduced the concept of dose equivalent in order to define a uniform scale of damage from exposure to radiation of different types and energies.⁵ The basic unit of dose equivalent is the rem, which is calculated from:

$$D_e = D \times Q,$$

where D_e is the dose equivalent in rem, D is the absorbed dose in rad, and Q is a dimensionless quality factor that relates the damage from radiation of the nuclide under examination to that from a standard radiation source such as 25-kV x rays or ⁶⁰Co gamma rays. (For many radioisotopes, $D \times Q$ in the equation above would be further multiplied by the appropriate RDFs; for tritium these are unity for all body tissues.⁷)

The quality factor (Q) for tritium beta rays was at one time assigned⁶ a value of 1.7. After several reviews during the 1960's, it was suggested that Q could not be shown to differ from unity; therefore, a value of 1.0 was recommended,³ and that recommendation is still in effect,² although some authorities believe a higher value is justified. Bonka⁸ has used a Q of 1.7; others^{1,4,5,9} assume the official value of 1.0. A quality factor of 1.0 has been used in this study.

With Q and applicable RDFs all equal to unity, it is obvious that the dose equivalent in rem is numerically equal to the absorbed dose in rad. The value 8.3×10^{-5} rem/Ci ingested as the dose conversion factor was adopted by Killough et al.⁴; this value has been accepted by Moore et al.⁵ and Till et al.⁷ and is used in the computations (with the AIRDOS-EPA⁵ computer model) for this study.

5.2 Radiological Impact of Released Tritium

All tritium released from reprocessing plants in this study is assumed to be released as tritiated water (HTO) in water vapor from the stack. This tritiated water is dispersed in a manner determined first by the local weather and meteorological conditions and subsequently by those of larger areas. Tritiated water is removed from the atmosphere by deposition and (primarily) by precipitation processes. The deposited tritium contaminates surface waters and is subject to reevaporation and to run-off to the ocean. The tritiated water mixes with waters of its hemisphere of origin and remains a part of the hydrological cycle until it decays.

The tritium, accordingly, is responsible for radiological dose commitments to the population near the plant and to the population of the much larger area over which the released tritium is deposited. As a component of the hydrological cycle, it is also responsible – although at much lower dose rates to the average individual – for dose commitments to the population of the world during its decay. These several dose commitments are described briefly in the following subsections.

5.2.1 Methodology for Estimation of Local Doses

The dose to the maximum individual is clearly an important consideration for the reprocessing plants in this study. In addition, doses to the population reasonably expected to be within a 50-mile radius of the plant are traditionally estimated for the nuclear facilities of the United States. The computer program AIRDOS-EPA has been used to estimate these doses for released tritium from hypothetical plants in this study.

AIRDOS-EPA⁵ is a methodology designed for use on IBM-360 computers to estimate (for radionuclides released to the atmosphere) concentrations in air, rates of deposition on ground surfaces, concentrations on ground surfaces, and intake rates via inhalation of air and ingestion of meat, milk, and fresh vegetables. Both horizontal and vertical dispersion can be estimated for as many as 36 radionuclides released from one to six stacks. Radionuclide concentrations in meat, milk, and fresh produce consumed by man are estimated by coupling the output of the atmospheric transport models with terrestrial food chain models. Dose conversion factors are input to the computer program, and doses to man at each specified distance and direction are estimated for total body, red marrow, lungs, endosteal cells, stomach wall, lower large intestine wall, thyroid, liver, kidneys, testes, and ovaries. Five exposure modes are included: (1) immersion in air containing radionuclides, (2) exposure to ground surfaces contaminated by deposited radionuclides, (3) immersion in contaminated water, (4) inhalation of radionuclides in air, and (5) ingestion of water and food produced in the area or elsewhere. If (as is the case for this study) tritium is the only isotope to be considered, the first three exposure modes listed above make no contribution to the dose.

Subroutine CONCEN of AIRDOS-EPA includes models used for plume rise above the top of the 100-m stack, the atmospheric dispersion model for dilution of the radionuclides (only tritium in this case) in an airborne plume as it is blown downwind, and models describing

deposition processes. The input parameters to CONCEN that were used in these estimates, along with details as to diets and sources of food and drink, are detailed in Appendix D.

5.2.2 Dose Commitments to Maximum Individuals and to Local Population

It is clear that the maximum actual individual resides at a point 4023 m (2.5 miles) southwest (sector 7) of the plant where the diffusion coefficient (χ/Q value) is 0.976×10^{-7} s/m³ (Appendix D, Table D.7). It is possible, but highly unlikely, that some maximum hypothetical individual could reside within that sector at a distance of 2414 m (1.5 miles) from the plant where χ/Q is 0.131×10^{-6} s/m³. For a release rate of 1 Ci/year,* the annual total body doses to these individuals would be those shown in Table 5.1. Annual organ doses for these individuals, with tritium as the only radionuclide released, would have very similar values.

Table 5.1. Estimated dose rates to total body of maximum individuals for release rate of 1 Ci/year of tritium

Dose rate via	Dose rate (millirem/year)	
	Maximum actual individual	Maximum hypothetical individual
Inhalation	3.11×10^{-6}	4.17×10^{-6}
Food ingestion	1.91×10^{-5}	2.57×10^{-5}
Water ingestion	1.8×10^{-7}	2.4×10^{-7}
Total	2.24×10^{-5}	3.01×10^{-5}

The annual total body dose to the population (nearly 1 155 000 persons) from this release rate (1 Ci/year of tritium) is estimated to be (in person-rem) 2.23×10^{-4} from inhalation, 1.374×10^{-3} from ingestion of food, and 1.2×10^{-5} from ingestion of drinking water, for a total of 1.61×10^{-3} person-rem. The average member of the population would receive some 1.394×10^{-9} person-rem for each year of such release. Organ doses would, again, have very similar values.

5.2.3 Dose Commitments to Large Regional Population

Released tritium is removed from the atmosphere by deposition and precipitation processes. As a consequence, the tritium contaminates surface waters; it is subject to reevaporation and reprecipitation, as well as to run-off to the ocean; and it is, accordingly, responsible for radiological dose commitments to the population in the large area over which its deposition becomes essentially complete.

Evidence⁷⁻¹⁰ suggests that these first-pass dose commitments to the population considerably exceed those to the population within 50 miles (above) and those caused by long-term exposure (below) of the world population to tritium after "equilibration" in the surface waters of the hemisphere. Unfortunately, however, no completely satisfactory

*This trivial release rate obviously gives negligible dose rates. The rates are presented for subsequent comparison with others estimated on the same basis. As will be apparent, these local doses are not always trivial when scaled to some of the possible real release rates.

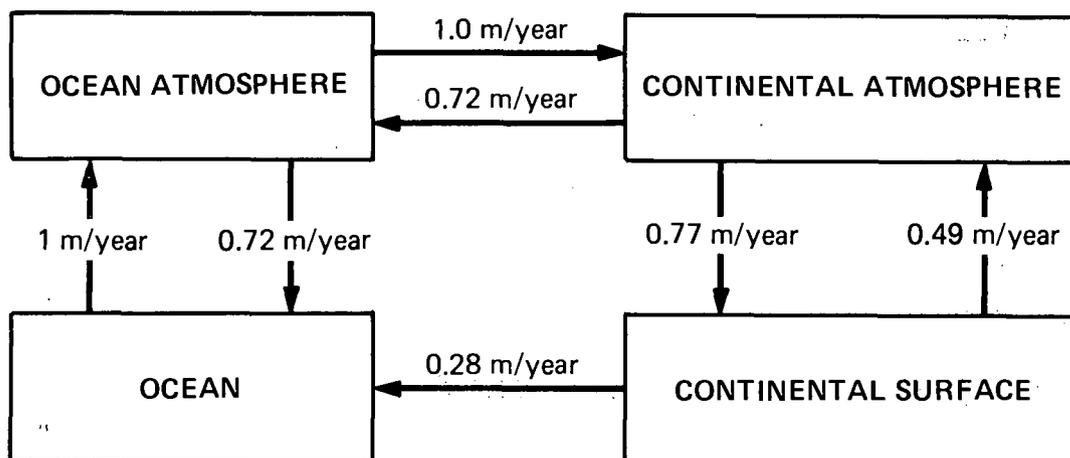
model has been described for estimating these large regional first-pass doses from tritium released within the United States.⁷

Published estimates for large regional populations. A simple model has been used¹¹ for estimating the dose from a U.S. reprocessing plant to the population of the United States. That estimate assumed rainout of one-half the HTO over the eastern United States (3.7×10^6 km² containing 80% of the U.S. population), dilution of that tritium by 100 cm of annual rainfall, and exposure of the population along with their animals and crops to tritium at that concentration. By this means, USEPA¹¹ estimated the annual dose to the U.S. population from a 1 Ci/year release of tritium to be about 3.1×10^{-3} person-rem. Soldat and Baker¹² show 2.1×10^{-3} person-rem/year to the U.S. population (number unspecified) east of the Rocky Mountains from release of 1 Ci/year from a source at Morris, Illinois. Both of these estimates used a quality factor of 1 for tritium irradiation; Soldat and Baker indicate that drinking water contributes negligibly to the dose commitment.

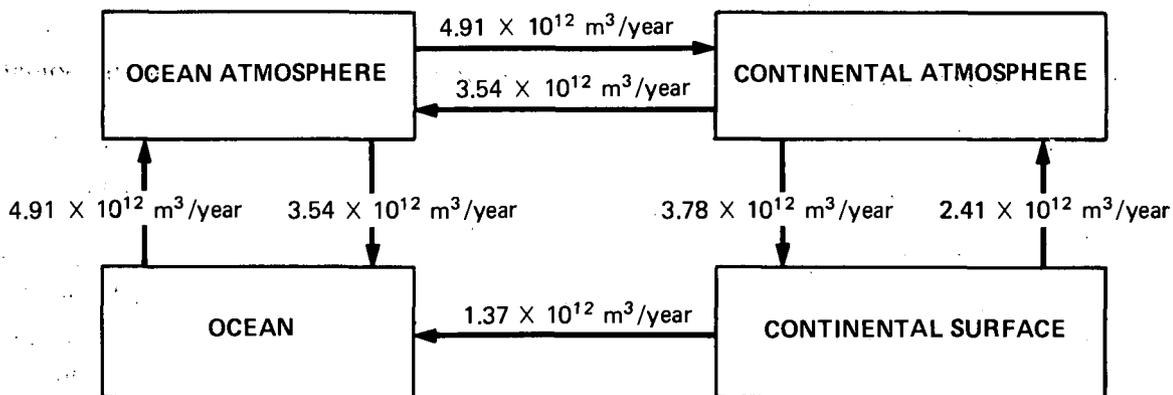
Bonka⁸ and NEA⁹ present estimates of the impact on the population of Europe from tritium release from a reprocessing plant near Hanover in Germany. Bonka uses 1.7 as the quality factor and estimates a curie per year release of tritium to be 17×10^3 person-rem/year. NEA, which seems otherwise to have adopted the Bonka model, uses a quality factor of 1.0 and obtains 1×10^{-2} person-rem/year.

Large regional impact by a simple model. Development of a model for estimating first-pass, large regional doses from tritium releases at the Oak Ridge site was not among the objectives of this study. Nevertheless, as a crude approximation the following very simple model is suggestive.

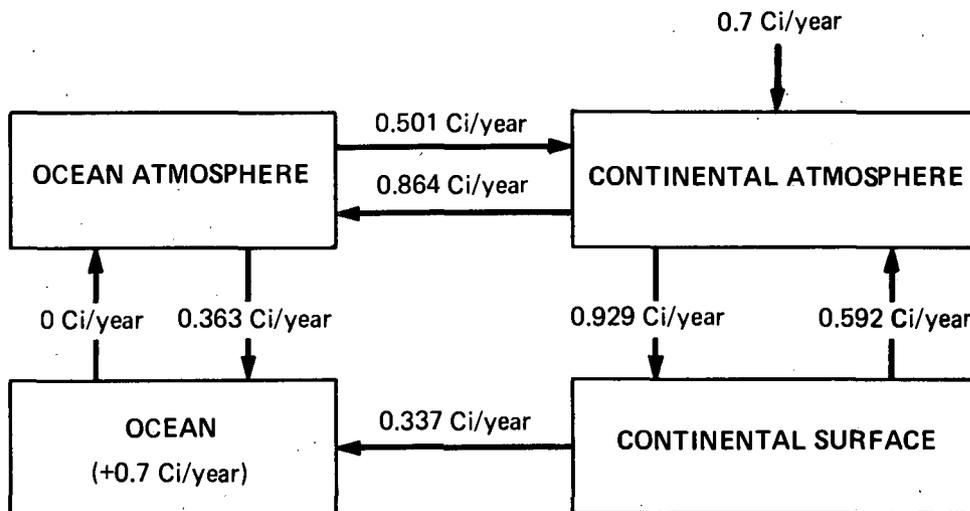
Begemann and Libby¹³ presented a water model for the Mississippi Valley in their analysis of behavior of cosmic ray and bomb tritium. A somewhat oversimplified version of their model is the following: for each unit of land area, the equivalent of 1 m/year of water comes in as atmospheric moisture from the ocean. On the continent, this moisture is joined by 0.49 m/year of moisture evaporated from the continental surface. Of this total (1.49 m/year), 0.77 m falls as rain or snow on the continental surface, and 0.72 falls as precipitation in the ocean. The difference between the 0.77 m/year that falls as rain or snow the 0.49 m that evaporates from the continental surface constitutes annual run-off to the ocean (0.28 m/year). On this basis, the simple block diagram of the system is:



As a crude approximation, it is assumed that 70% of the released tritium is deposited within a circular area (1250-km radius) during its transport by winds that average 4 m/s in velocity and that blow equally to all points of the compass. We assume that the model of Begemann and Libby¹³ is applicable over the entire $4.91 \times 10^6 \text{ m}^2$ area.* If so, the volumes of water involved per year are those in the block diagram below.



If the uniform release rate is 1 Ci/year, then, by these assumptions, 0.7 Ci/year of tritium becomes involved in this hydrological cycle. We assume that tritium delivered as precipitation (and as run-off) to the ocean is immediately diluted to negligible concentrations. We further assume that equilibrium is achieved within the boxes representing ocean atmosphere, continental atmosphere, and continental surface. If so, the tritium fluxes (with 0.7 Ci deposited each year) are:



*With the circular area centered on Oak Ridge, Tennessee, nearly 25% of the area consists of portions of the Atlantic Ocean and the Gulf of Mexico. This ocean area begins, on the average, some 450 miles from the site and would receive directly only about 7.5% of the tritium released. These facts are ignored and the entire area ($4.91 \times 10^6 \text{ m}^2$) is assumed to be continental surface.

If this model is assumed, the release of 1 Ci/year of tritium would lead, on the average, to tritium concentrations of 2.46×10^{-13} Ci/m³ (2.46×10^{-7} pCi/g) in the continental surface waters of the circular area. If the average absolute humidity over this area were 8 g/m³, the air concentration would be $\sim 1.97 \times 10^{-6}$ pCi/m³. If exposed to air at that tritium concentration, the average individual would (with his food in equilibrium with that air, and with his drinking water at 1% of the specific activity of atmospheric moisture) receive 1.43×10^{-8} millirem/year. If his drinking water were assumed to be at the same specific activity as atmospheric moisture (as is very conservative, but perhaps is less so than a similar assumption for the local population), the dose rate becomes 2.54×10^{-8} millirem/year.

This circular area, centered on Oak Ridge, Tennessee, includes considerable ocean area, but it includes somewhat more than the eastern half of the United States, along with a portion of the more populous area of southern Canada. Its population in year 2020 might be 3.1×10^8 persons. If so, each curie released from our hypothetical processing plant would result in a total body dose of between 4.5×10^{-3} and 7.9×10^{-3} person-rem to this large region.

This model has, of course, ignored some 30% of the release. Impact of this 30%, although not negligible, is small since it (1) would be deposited over a very much larger area (a substantial portion of which is ocean), (2) would be diluted with markedly larger quantities of water, and (3) would generally affect areas with considerably lower population densities since little would survive the Atlantic crossing. The conservatism included in the higher value seem certainly sufficient to include the impact of the ignored 30%. It seems quite unlikely that release of 1 Ci of tritium in Oak Ridge, Tennessee, could be responsible for more than 8×10^{-3} person-rem during the process of its incorporation into the world's surface waters. Indeed, it may well be that 5×10^{-3} person-rem/Ci released is more reasonable.

Summary of doses to large regional populations. It is clear from the foregoing that the impact of tritium on the large regional population is not well established and that better models – particularly for release within the United States – are needed. For purposes of this study we will use the value of 8×10^{-3} person-rem/Ci released for the impact of tritium from a hypothetical plant in Oak Ridge, Tennessee. This value, which includes (in principle) the dose to the smaller population within 50 miles of the plant, suggests that the average individual within this large population will receive about 2.3×10^{-11} rem/Ci released.

5.2.4 Ultimate Dose Commitment to World Population

Once the released tritium becomes part of the hydrological cycle, it will continue to give very small dose commitments to the global population until its decay. Although some authorities^{9,14} present dose commitments to the entire world population, most (and perhaps all) authorities agree that little of the tritium released to the troposphere leaves the hemisphere in which it originates. NCRP,² for example, finds that releases of tritium within the 30 to 50° north latitude band (which includes Oak Ridge, Tennessee) delivers most of its dose commitment to the population of that band and essentially none to populations outside the 10 to 60° north latitude band. Bergman et al.¹⁴ show relatively small dose commitments to the population outside the hemisphere of release.

Estimates of global dose commitments from specific tritium releases assume that the tritium reaches a semiequilibrium state in the earth's (hemisphere's) waters and that it

follows the hydrological cycle without discrimination. A variety of models have been used to estimate the tritium concentrations in the environmental pools that contribute to exposure of the population. Till et al.⁷ have presented a discussion of several of the recent models.

The simplest model¹¹ treats the entire circulating surface water of the northern hemisphere as a single compartment. Bonka⁸ has used, and NEA⁹ has adopted, a model that considers the mixed layer and the deep layer of each hemisphere's ocean as separate compartments with transfer of tritium within the compartments of each hemisphere and between the hemispheres. These simple models have very large compartments in which the tritium concentrations are assumed to be uniform.

More complex models have been introduced in recent years. A three-compartment model (atmospheric water, circulating ocean surface water, and land surface water), along with a simple one-compartment model and a seven-compartment model, very similar to that proposed by Easterly and Jacobs,¹⁵ have been described by NCRP²; a model with eight compartments was used by Bergman et al.¹⁴ The complex models must estimate transfer coefficients between compartments and, via a computer, simulate the total system until it reaches a pseudo-steady state.

Estimates of ultimate dose from releases of tritium at some future time must make use of some assessment of the population to be affected by the tritium in the earth's waters. The United Nations¹⁶ estimated as its "medium" variant, the world population to be (in billions) 6.406, 9.065, 11.163, and 12.210 in years 2000, 2025, 2050, and 2075 respectively. In reasonable conformity with this, we have assumed that the population increases linearly in the 2000 to 2025, the 2025 to 2050, and the 2050 to 2075 year periods. We have further assumed that the world population stabilizes and remains constant at 12.21×10^9 persons after year 2075, that after year 2020 the population of the northern hemisphere remains at 80% of the world population, and that the population of the 30 to 50° north latitude band remains at 26% of the world population. With these assumptions, the populations after year 2020 are those of Table 5.2.

Table 5.2. Values assumed for future populations

Location	Population (billions) in —			
	2020	2025	2050	2075 and forever forward
World total	8.54	9.06	11.16	12.21
Northern hemisphere	6.83	7.25	8.93	9.77
30 to 50°N latitude	2.22	2.35	2.90	3.17

Virtually all who have presented "global" models have estimated dose commitments to the affected populations. They have, in general, used different and (usually but not invariably) constant populations for the world or for portions of it. Some have presented values for the ultimate dose commitment to the average individual from a single-unit release, and most have presented data from which values for such a commitment can be reasonably well inferred. Table 5.3 lists such values from several recent sources.

Table 5.3. Values presented or inferred from estimates presented by several sources for dose commitments to global populations from a single release of 1 Ci of tritium during 1 year to troposphere of northern hemisphere

Source	Model used	Global commitment (person-rem)	Affected population		Dose commitment to average affected individual (rem)
			Location	Number (billions)	
USEPA ^a	Single compartment	4.2×10^{-4}	Northern Hemisphere	4.4	1×10^{-13}
NCRP ^b	One compartment		30 to 50°N latitude	1.7	5.9×10^{-13}
NCRP ^b	Seven compartments		30 to 50°N latitude	1.7	1.1×10^{-12}
Bergman et al. ^c	Eight compartments ^d		World		1.8×10^{-13}
NEA ^e	Separate	1.7×10^{-3}	World	10	1.7×10^{-13}
Till ^f		1.4×10^{-3}	Northern Hemisphere	3.2	4.4×10^{-13}

^aU.S. Environmental Protection Agency Environmental Analysis of the Uranium Fuel Cycle, Part III. Nuclear Fuel Reprocessing, PB-235806, EPA-520/9-73-003D, Office of Radiation Programs, U.S. Environmental Protection Agency, Washington, D.C. (1973).

^bTritium in the Environment, Report No. 62, National Council on Radiation Protection and Measurements, Washington, D.C. (March 1979).

^cR. Bergman, U. Bergstrom, and S. Evans, "Environmental Transport and Long-Term Exposure for Tritium Released in the Biosphere," Proceedings, International Symposium on Behavior of Tritium in the Environment, San Francisco, October 16–20, 1978; International Atomic Energy Agency, Vienna, 1979.

^dCommitment to average individual after normalization to dosimetry used in this study.

^eRadiological Significance and Management of Tritium, Carbon-14, Krypton-85, and Iodine-129 Arising from the Nuclear Fuel Cycle, ISBN 92-64-12083-1, Report by an NEA Group of Experts, Nuclear Energy Agency, Paris, France (April 1980).

^fJ. E. Till (Oak Ridge National Laboratory), personal communication to D. C. Hampson, May 1980.

Given such a value for the ultimate dose commitment (in rem) to the average individual in the affected population from a unit release of tritium (D_{ai}), it is clear that the dose to the expanding affected population is the product of D_{ai} by the summation of the affected population in any year (P_t) times the fraction (f_t) of the unit of tritium that decays during that year. That portion of the ultimate dose delivered after the population (P_c) times that fraction of the unit of tritium decaying after the population becomes constant. Thus the ultimate dose to the population (D_p^∞) in our scenario (for release of 1 Ci of tritium in year 2020) becomes

$$D_p^\infty = D_{ai} \sum_0^{55} P_t \times f_t + D_{ai} P_c \int_{55}^{\infty} \lambda e^{-\lambda t} dt$$

Estimates of the ultimate dose commitment to the affected population from 1 Ci of tritium released during year 2020 prepared in this way from the data of Table 5.3 are shown in Table 5.4. The values range from about 9.3×10^{-4} person-rem to 3.2×10^{-3}

Table 5.4. Ultimate global dose commitment per curie of tritium released in year 2020 estimated for population scenario assumed and data from several sources

Source	Model	Ultimate dose commitment (person-rem)
USEPA ^a	Single compartment	9.3×10^{-4}
NCRP ^b	One compartment	1.4×10^{-3}
	Seven compartments	2.6×10^{-3}
NEA ^c	Separate compartment	1.6×10^{-3}
Bergman et al. ^d	Eight compartments	1.7×10^{-3}
Till ^e		3.2×10^{-3}

^aU.S. Environmental Protection Agency Environmental Analysis of the Uranium Fuel Cycle, Part III. Nuclear Fuel Reprocessing, PB-235806, EPA-520/9-73-003D, Office of Radiation Programs, U.S. Environmental Protection Agency, Washington, D.C. (1973).

^bTritium in the Environment, Report No. 62, National Council on Radiation Protection and Measurements, Washington, D.C. (March 1979).

^cRadiological Significance and Management of Tritium, Carbon-14, Krypton-85, and Iodine-129 Arising from the Nuclear Fuel Cycle, ISBN 92-64-12083-1, Report by an NEA Group of Experts, Nuclear Energy Agency, Paris, France (April 1980).

^dR. Bergman, U. Bergstrom, and S. Evans, "Environmental Transport and Long-Term Exposure for Tritium Released in the Biosphere," *Proceedings*, International Symposium on Behavior of Tritium in the Environment, San Francisco, October 16-20, 1978; International Atomic Energy Agency, Vienna, 1979.

^eJ. E. Till (Oak Ridge National Laboratory), personal communication to D. C. Hampson

person-rem for this unit release. For purposes of this document, we will adopt the value of 2.0×10^{-3} person-rem committed to the global population for each Ci released from the reprocessing plants in this study.

5.2.5 Summary of Dose Commitments from Reprocessing Plant Operation

From the dose commitments from unit releases of tritium to the atmosphere and from the source terms (described in Sects. 3 and 4 and presented in Table 5.5), the dose commitment to individuals and to the several population groups from a year's operation of the hypothetical facilities are obtained. The estimates for releases during year 2020 are shown in Table 5.5.

It is apparent that the base-case light-water reactor (LWR) plant (case LWR-I) would be responsible for dose commitments (largely delivered within year 2020) of 1300 person-rem and 6500 person-rem to the local (50 miles) and to the large regional population respectively. The year 2020 releases would also ultimately be responsible for an additional 1620 person-rem delivered to the global population over a long period (with 99.6% delivered within 100 years). In principle, the commitment to the large regional population includes that to the local population. If that is ignored and the three values are totaled, year 2020 operation might ultimately be responsible for 9420 person-rem to the combined population groups. Releases in earlier years would yield lower commitments since the several populations would be smaller. However, if this is ignored, the 20 years of LWR processing plant operation would ultimately be responsible for 1.9×10^5 person-rem committed to the combined population groups. Similar considerations indicate that the year 2020 releases from the base-case liquid-metal fast breeder reactor (LMFBR) plant would be responsible for dose commitments of 1520 person-rem to the combined populations; its 20 years of operation would ultimately deliver 3.0×10^4 person-rem to those population groups. It is, in addition, obvious that all other plants are responsible for markedly smaller dose commitments than are their respective base cases.

It cannot be too strongly emphasized that even the largest of these estimated dose commitments is miniscule in comparison with the dose commitments from exposure to the natural background radiation. The LWR base-case plant is estimated to deliver, during its 20 years of operation, 2.6×10^4 person-rem to the local (50 mile) population; during that 20 years the local population would receive a total body dose commitment of about 2.9×10^6 person-rem from the natural background. In a similar period the 3.1×10^8 people in the large regional population would receive some 8.0×10^8 person-rem from the unavoidable natural background and 1.3×10^5 person-rem from tritium emitted by the LWR base-case plant.

At the same time it must be noted that the annual dose commitments to the maximum individuals from complete release of tritium by the base-case plant (LWR-I) are relatively high. They are below (but not comfortably below) the 40 CFR 190 limit of 25 millirem/year set by the Environmental Protection Agency (EPA) for dose commitments to individuals for emissions from fuel cycle facilities. It should be emphasized that these annual dose commitments are estimated with the very conservative assumption that all of the individual's food is grown at his location. More realistic assumptions would lead to lower estimated doses. Moreover, corrections for thermal buoyancy of the stack plume would lower the estimates, and, of course, lower estimates would result if a taller stack were used. Since tritium would not be the only radionuclide released to the atmosphere from such a plant,

Table 5.5 Dose commitment to individuals and to populations from tritium released by hypothetical plants in year 2020

Source	Tritium release (Ci)	Maximum individual ^a (millirem/year)		Dose commitment (person-rem) from year 2020 release ^b		
		Actual	Hypothetical	Local population (1.16 × 10 ⁶)	Regional population (3.1 × 10 ⁶)	Global population
LWR plant tritium						
Case I	8.1 × 10 ⁵	18	24	1300	6500	1620
Case II	1.0 × 10 ⁵	2.3	3.1	168	840	210
Cases III, IVA, and VA	3.8 × 10 ⁴	8.5 × 10 ⁻¹	1.1	61	300	76
Cases IVB and VB	3.9 × 10 ³	8.7 × 10 ⁻²	1.2 × 10 ⁻¹	6.3	31	8
Case VI	1.8 × 10 ⁴	4.0 × 10 ⁻¹	5.4 × 10 ⁻¹	69	145	36
LMFBR plant tritium						
Case I	1.3 × 10 ⁵	2.9	3.9	210	1050	260
Case II	1.6 × 10 ⁴	3.5 × 10 ⁻¹	4.3 × 10 ⁻¹	26	130	32
Cases III, IV, and V	8.4 × 10 ³	1.9 × 10 ⁻¹	2.5 × 10 ⁻¹	14	67	17
Case VI	5.1 × 10 ³	1.1 × 10 ⁻¹	1.5 × 10 ⁻¹	8.2	41	10
Natural background		125	125	1.45 × 10 ⁵	3.9 × 10 ⁷	
Natural tritium	4 × 10 ⁶					8000

^aNote that dose commitments to maximum individuals are independent of year of release. All other commitments are estimated with some conservatism by assuming release in year 2020 with larger populations than were present in earlier years.

^bNote that the global population dose commitments are those delivered over a long time period to a population assumed to increase until year 2075. All other commitments are essentially delivered during the year of release.

this limit on dose to individuals may preclude complete release of tritium from plants processing 1500 metric tons of heavy metal (MTHM) per year of LWR fuel. It is apparent, however, that all other LWR plants and all LMFBR plants offer dose commitments at or below 3 millirem/year, even to our maximum hypothetical individual. All of these would, accordingly, be comfortably below the EPA 40 CFR 190 limit.

5.2.6 Summary of Dose Commitments from Storage and Disposal Options

From storage options. As indicated in Sect. 4.2, one of the options for management of tritium retained by the several plants is by long-term storage in tanks. Consideration was given to such storage for 50 and for 100 years with the residual undecayed tritium assumed to be released to the atmosphere as HTO vapor through the plant stack. Such vaporization after 50 years would result in release of 6% of the tritium originally stored, whereas vaporization after 100 years would release 0.36% of that originally stored.

It is assumed quite simplistically that the contents of each tank are vaporized at the rate at which the tank was filled and at a time when the (average) age of the contents is 50 years. The period during which the vaporization would proceed would last about 20 years, and it would be completed shortly after 2070. If the same assumptions are made, but the storage times are 100 years, the vaporization process would proceed in the period of years 2100 through 2120. There appear to be no estimates of the local (50 mile) or the large regional population for these time periods. It has, accordingly, been assumed that both the local and the regional population will increase at a rate that equals the growth rate for the population of the 30 to 50°N latitude band shown in Table 5.2. Moreover, it has been assumed, with some conservatism for the 50-year storage case, that the population is constant at the levels for year 2075 and following during all these releases. On these bases, the local population is assumed to be 1.65×10^6 persons and the large regional population is assumed to be 4.4×10^8 persons.

Table 5.6 shows the dose commitments to individuals and to the several population groups that would result from each year of such releases. It is clear that dispersal of the stored tritium over a 20-year interval would result in relatively low (and presumably acceptable) doses to the maximum individuals. The total inventory of tritium dispersed during this period from reprocessing of LWR fuel would ultimately be responsible for 13 000 to 14 300 person-rem to the combined population groups. Table 5.7 shows dose commitments to the combined populations from 20 years of plant operation and from dispersal of the tritiated waters after 50 years of storage. Dose commitments from 20 years of plant operations and from dispersal of the tritiated waters after storage for 100 years are shown in Table 5.8.

It is apparent from these tables that dispersal after storage for 50 years would be responsible for dose commitments that are of the same order as those from plant operation and that the more efficient the plant retention system the more preponderant become the commitments resulting from subsequent dispersal. Dose commitments incurred on dispersal after 100 years of storage make only trivial contributions to the total except for the plants (LWR-IVB and VB) with very efficient retention systems. Such 100-year storage and atmospheric dispersal would seem to be nearly equivalent to satisfactory permanent disposal.

From disposal options. The several options considered for disposal of the retained tritium (Sect. 4.2 and Table 4.1) may vary appreciably in the degree to which they can keep

Table 5.6. Dose commitment to individuals and to populations from tritium released in year 2070 after storage for 50 years

Source	Tritium release (Ci)	Maximum individual ^a (millirem/year)		Dose commitment (person-rem) from year 2070 release ^b		
		Actual	Hypothetical	Local population (1.65 × 10 ⁶)	Regional population (4.4 × 10 ⁸)	Global population
LWR plant tritium						
Case II	4.25 × 10 ⁴	9.5 × 10 ⁻⁴	1.3 × 10 ⁻³	95	490	104
Cases III, IVA, and VA	4.65 × 10 ⁴	1.0 × 10 ⁻³	1.4 × 10 ⁻³	105	530	113
Case IVB and VB	4.65 × 10 ⁴	1.0 × 10 ⁻³	1.4 × 10 ⁻³	105	530	113
Case VI	4.75 × 10 ⁴	1.1 × 10 ⁻³	1.4 × 10 ⁻³	107	540	115
LMFBR plant tritium						
Case II	6.65 × 10 ³	1.5 × 10 ⁻⁴	1.8 × 10 ⁻⁴	11	50	16
Cases III, IV, and V	7.13 × 10 ³	1.6 × 10 ⁻⁴	1.9 × 10 ⁻⁴	12	53	17
Case VI	7.35 × 10 ³	1.66 × 10 ⁻⁴	2.0 × 10 ⁻⁴	12	55	18

^aNote that dose commitments to maximum individuals are independent of year of release. All other commitments are estimated with some conservatism by assuming release in 2020 with larger populations than were present in earlier years.

^bNote that the global population dose commitments are those delivered over a long time period to a population assumed to be constant at the year 2075 level. All other commitments are essentially delivered during the year of release.

Table 5.7. Ultimate dose commitments to the combined populations from 20 years of reprocessing operations and from dispersal after storage for 50 years

Case	Dose commitment (person-rem)		
	From plant ^a	From dispersal ^b	Total ^c
LWR-I	1.9×10^5		1.9×10^5
LWR-II	2.4×10^4	1.4×10^4	3.8×10^4
LWR-III, IVA, and VA	8.7×10^3	1.5×10^4	2.4×10^4
LWR-IVB and VB	9.1×10^2	1.5×10^4	1.6×10^4
LWR-VI	5.0×10^3	1.5×10^4	2.0×10^4
LMFBR-I	3.0×10^4		3.0×10^4
LMFBR-II	3.8×10^3	1.5×10^3	5.3×10^3
LMFBR-III, IV, and V	2.0×10^3	1.6×10^3	3.6×10^3
LMFBR-VI	1.2×10^3	1.7×10^3	2.9×10^3

^aDose commitments conservatively estimated as 20 times those from release in year 2020.

^bDose commitments conservatively estimated by assuming populations as of 2075 (see text).

^cTotal dose commitments to local, regional, and global populations.

Table 5.8. Ultimate dose commitments to the combined population after 20 years of reprocessing operations and from dispersal after storage for 100 years

Case	Dose commitment ^a (person-rem)		
	From plant	From dispersal ^b	Total
LWR-I	1.9×10^5		1.9×10^5
LWR-II	2.4×10^4	7.8×10^2	2.5×10^4
LWR-III, IVA, and VA	8.7×10^3	8.4×10^2	9.6×10^3
LWR-IVB and VB	9.1×10^2	8.4×10^2	1.8×10^3
LWR-VI	5.0×10^3	6.0×10^2	5.6×10^3
LMFBR-I	3.0×10^4		3.0×10^4
LMFBR-II	3.8×10^3	90	3.9×10^3
LMFBR-III, IV, and V	2.0×10^3	96	2.1×10^3
LMFBR-VI	1.2×10^3	102	1.3×10^3

^aTotal dose commitments to local regional and global populations.

^bDose commitments estimated for populations constant at the year 2075 levels.

the tritium isolated from the environment for the 150 years required for its decay to 0.022% of its original activity. Surface trench burial, which may be permissible only for the most dilute and voluminous of the tritiated waters and which seems to be quite expensive for those, is probably the least dependable of the disposal methods. Hydrofracture, intermediate-depth burial in concrete monoliths, and deep-well disposal at well-chosen sites all seem likely to

provide effective confinement. Confinement in a geologic repository or deep-sea disposal would seem to provide all but certain effective isolation from man's environment. No attempt has been made in this study to evaluate in detail the likelihood or the magnitude of possible releases of tritium to the environment from any of the disposal options. We have assumed that no appreciable interaction of tritium with man will result from any disposal methods and sites. This is, of course, equivalent to the assumption that a very high probability of safe disposal must be demonstrated before such disposal will be permitted. Consequently, we have assumed that no appreciable dose commitments will follow operation of any disposal systems.

5.3 Cost-Effectiveness Considerations

There are no regulations that define the circumstances under which the options for retention and disposal of tritium from fuel reprocessing plants would be considered cost effective. Moreover, it is not certain whether, at a time so far in the future as the year 2000, cost-effectiveness will be considered in the decision that might require control of tritium from such plants.

The Nuclear Regulatory Commission in Appendix I to 10 CFR 50 provided numerical guidance on design objectives for light-water cooled nuclear power reactors to meet the requirement that radioactive material in effluents released to unrestricted areas be kept "as low as reasonably achievable." That document included (in addition to specific dose limits to individuals maximally exposed to liquid effluents, to gaseous effluents, and to radioiodine and particulates) the following requirements:

In addition to the provisions of paragraphs A, B, and C above, the applicant shall include in the radwaste system all items of reasonably demonstrated technology that, when added to the system sequentially and in order of diminishing cost-benefit return, can for a favorable cost-benefit ratio effect reductions in dose to the population reasonably expected to be within 50 miles of the reactor. As an interim measure and until establishment and adoption of better values (or other appropriate criteria), the values \$1000 per total body man-rem and \$1000 per man-thyroid-rem (or such lesser values as may be demonstrated to be suitable in a particular case) shall be used in this cost-benefit analysis.

It is clear that the man-thyroid-rem was included because of possible releases of ¹³¹I whose dose commitment to thyroid markedly exceeds that to total body. Appendix I suggested that additional hearings would be conducted to establish more firm values for the worth of a man-rem, but no such hearings have been held. Accordingly, the light-water cooled reactors are required to add radwaste equipment if, and only if, the resulting decrease in dose commitment to the population "reasonably expected to be within 50 miles of the reactor" from the radionuclides released each year is obtained at an annualized cost of \$1000 or less per man-rem or man-thyroid-rem. The annualized cost includes fabrication and installation of the equipment (including cost of money) as well as cost of maintenance and operation of the equipment. Appendix I contained (presumably because the \$1000 values were considered to be interim figures) no provision for changes in the value of money.

How, or even whether, a similar requirement would be applied to tritium releases from fuel reprocessing plants is, of course, speculative. Tritium, unlike radioiodine, delivers essentially equal dose commitments to total body and to thyroid. It might, therefore, be argued that decrease in total body dose from tritium by a man-rem is worth \$2000. Since it

was previously established (Sect. 5.2.2) that release to the atmosphere of 1 Ci/year of tritium (as HTO) was responsible for a 1.61×10^{-3} whole body person-rem, and an equal amount to the thyroid, it is clear that the retention worth of 1 Ci of tritium is near \$3.20.

It is clear (Tables 4.33 and 4.34) that relatively few of the retention plus disposal options would be considered cost effective for LWR fuel reprocessing by this standard. Moreover, for LMFBR fuel reprocessing, only water recycle (retention case LMFBR-II) with deep-well disposal or hydrofracture are close to being cost effective.

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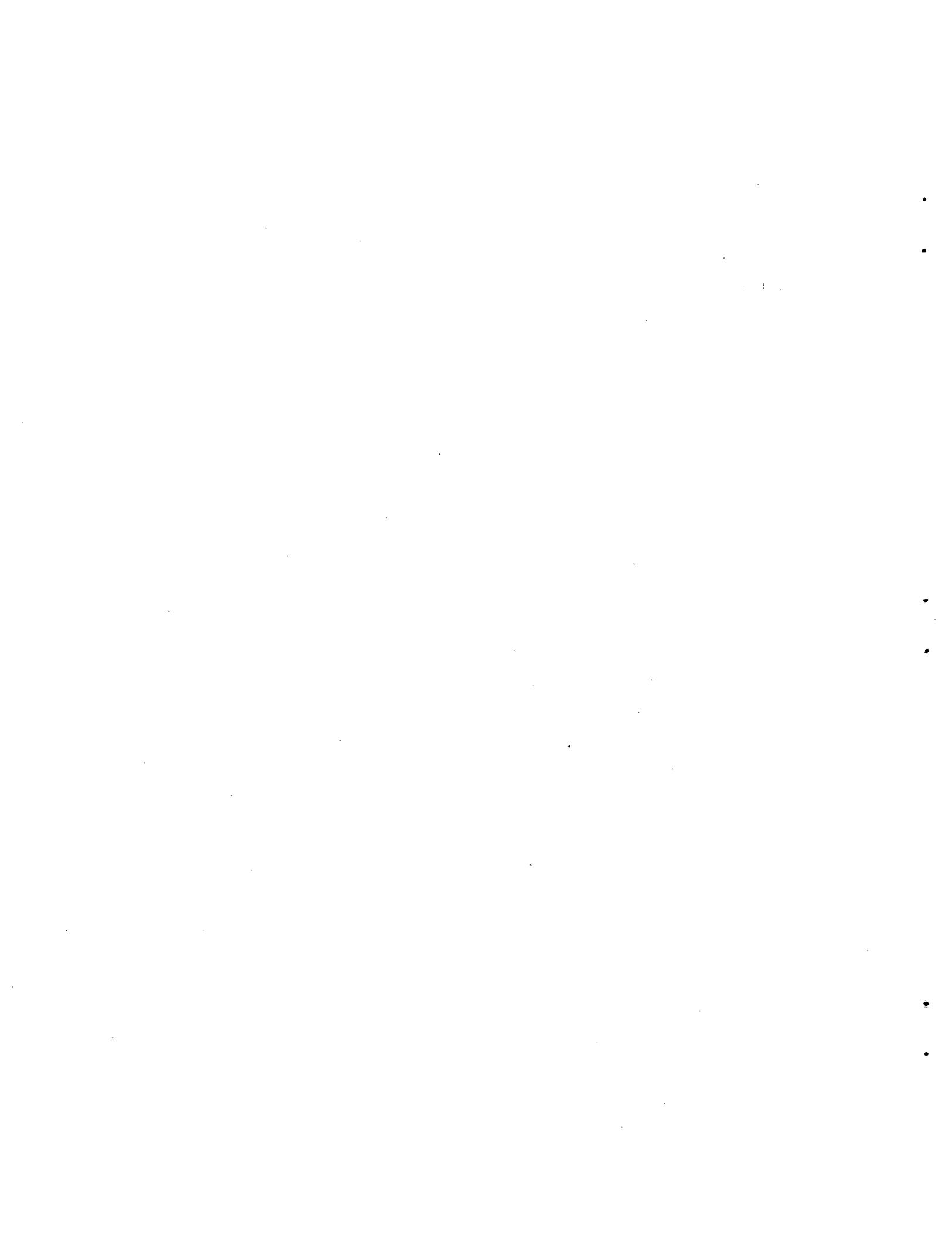
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APPENDIX A
TECHNICAL ASSESSMENT OF ISOTOPE SEPARATION PROCESSES

A.1 Dual-Temperature Exchange and Its Modification

The dual-temperature exchange (GS) process passes H_2S gas countercurrent to tritiated water in gas-liquid contactor columns. Since the extent of exchange among the hydrogen isotopes is a function of temperature, two columns, one at 30 to 40°C and one at 120 to 140°C, are employed under a total pressure of ~ 19 atm. As shown in Fig. A.1, the gaseous

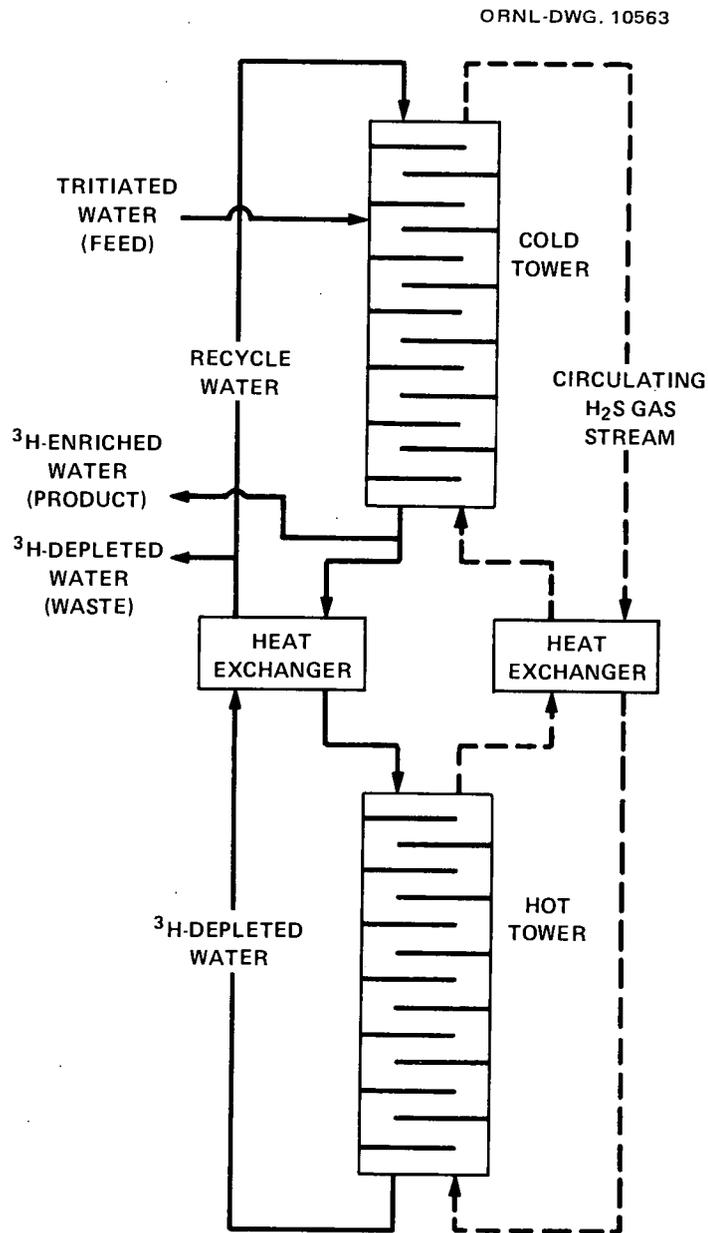


Fig. A.1. Schematic diagram of dual-temperature H_2O - H_2S exchange process.

H₂S passes up the hot tower where it increases in tritium concentration, through a heat exchanger where it is cooled, up the cold tower where its tritium concentration is diminished, and after it is heated in the heat exchanger, to the bottom of the hot tower. Tritiated feed water is introduced into the upper portion of the cold tower and passes down the tower, increasing in tritium concentration. A portion of this water is withdrawn as concentrated product at the bottom of the cold tower; the balance is warmed in a heat exchanger and is fed to the top of the hot tower. The water flows down the hot tower and is depleted in tritium. From the hot tower bottom, this depleted stream, after being cooled in the heat exchanger, is split; one portion is withdrawn as tritium-depleted waste, and the balance is sent to the top of the cold tower.

The relatively high effective separation factor (estimated to be 1.29 with the cold tower at 30°C and the hot tower at 130°C) is an advantage of the process. Another advantage is the high throughput that is possible. The process can have problems – with possible corrosion of carbon steel by the H₂S in the highly humid atmosphere and with process control. However, practical experience with the process shows that proper material selection, careful equipment design and maintenance, and proper process design can result in high on-stream availability.^{1,2} The GS isotope separation facility at the DOE's Savannah River Laboratory (Aiken, South Carolina) has been on stream 96% of the time over a period of several years.

A modification of this process that enhances the separation of tritium is one (Fig. A.2) in which the top of the cold (30°C) tower is fed with tritium-free water. A portion of the concentrate from the cold tower bottom constitutes the product, whereas the effluent from the hot tower bottom constitutes the depleted waste stream.³

The increased separation efficiency (for tritium) of this modification promises to be valuable in many applications, since it reduces (in some cases markedly) the number of hot and cold towers that would otherwise be required to achieve the necessary separation factors. However, this increased efficiency is obtained by the addition of tritium-free fresh water to the system. A concentrated tritium product can be obtained from this system for storage or disposal, but the remainder of the tritium is available only in a very large volume of water. As described in Sect. 4.1, this volume of "waste" is so large as to preclude evaporation and must be managed by discharge to surface waters (not permitted for this study) or by deep-well disposal onsite.

The dual-temperature process used at Savannah River and the potentially even more efficient Exxon Nuclear Company version³ seem to offer good promise of process readiness by the 1990's. It should be noted, however, that the process has not been used for the recovery of tritiated concentrates from dilute reprocessing plant waters. Therefore, the following observations can be made:

1. Major design modifications may be necessary to ensure optimal recovery of tritium from the dilute process streams.
2. Very large quantities of cooling water are required.
3. Careful design is required to ensure safe handling of the very large quantities of toxic H₂S that must be used.
4. The process requires essentially pure water, and the effect of nitric acid (at ~0.01 *M*) from the process solution condensate needs to be evaluated. It is possible that an additional distillation of the condensate after neutralization may be required.

- **DEVELOPER** DUPONT, ENC VARIATION, THEORETICAL
- **PROCESS HIGHLIGHTS** ENRICHMENT OF HTO BASED ON HT EXCHANGE BETWEEN H₂O AND H₂S AT DIFFERENT TEMPERATURES
- **PROCESS ADVANTAGES** TECHNICALLY WELL ESTABLISHED – SOME R&D REQUIRED FOR COMMERCIAL PROCESS
- **PROCESS DISADVANTAGES** LOWER SEPARATION FACTOR; H₂S CORROSIVE TO MATERIALS OF CONSTRUCTION; LARGE VOLUME OF FRESH WATER REQUIRED

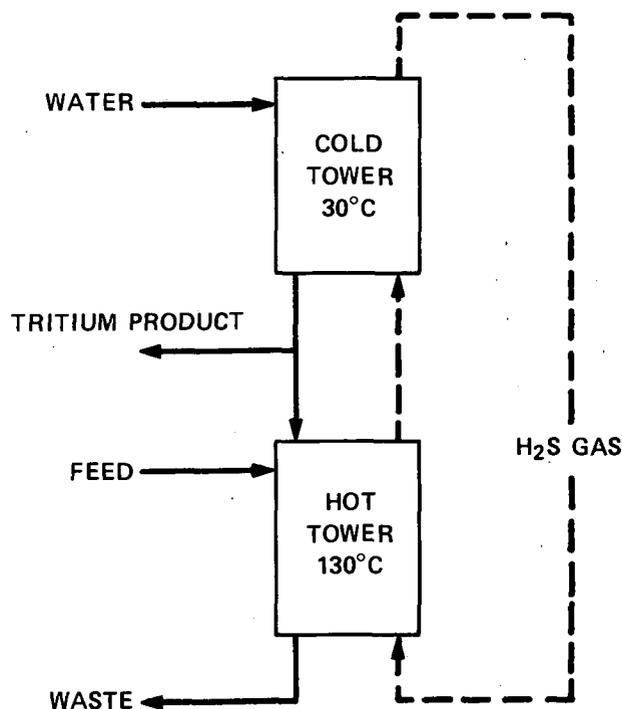


Fig. A.2. Variation of the dual temperature exchange process.

In addition, the effect on the Purex process of H₂S (at ~1 ppm) in water returned to the processing plant needs to be determined.

A.2 Combined Electrolysis-Catalytic Exchange (CECE) Process

Chemical exchange between a gaseous mixture of hydrogen isotopes and water containing these isotopes favors the accumulation of the heavier isotopes in the water phase. Moreover, when water is decomposed electrolytically into hydrogen and oxygen, the reaction kinetics also favor retention of the heavier isotopes in the liquid water. The CECE process

uses both of these effects, with gaseous hydrogen being produced by electrolysis in quantities sufficient for chemical exchange (promoted by a hydrophobic catalyst) with the liquid water.

A schematic diagram of the process is shown in Fig. A.3. The equipment includes for each separation stage, an electrolysis cell, a countercurrent packed-bed column for the

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- **DEVELOPER** MOUND LABORATORIES/AECL
- **PROCESS HIGHLIGHTS** ISOTOPIC EXCHANGE BETWEEN WATER AND HYDROGEN TRANSFERRING TRITIUM FROM GAS TO LIQUID PHASE. AN ELECTROLYTE UNIT PROVIDES THE HYDROGEN (TRITIUM-ENRICHED GAS)
- **PROCESS ADVANTAGES**
 - HIGH SEPARATION AND CONCENTRATION FACTORS
 - HIGH CAPACITY POTENTIAL
 - NONCORROSIVE SYSTEM
- **PROCESS DISADVANTAGES**
 - PILOT-PLANT STAGE
 - RADIATION EXPOSURE POTENTIAL TO ELECTROLYSIS UNIT AND HYDROPHOBIC CATALYST

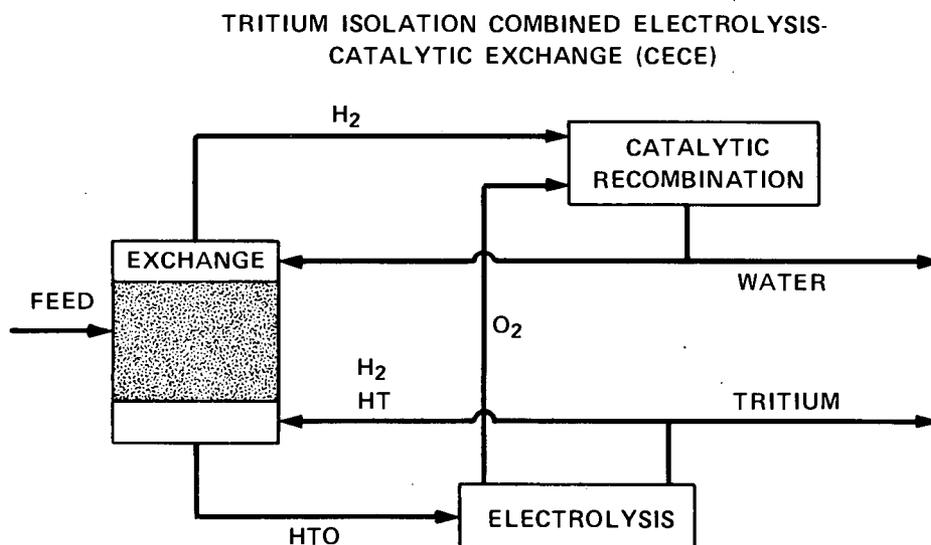


Fig. A.3. Combined electrolysis-catalytic exchange.

H_2 - H_2O exchange, and a catalytic recombiner to provide reflux water by oxidation of part of the gaseous hydrogen. As the water passes down each column, it becomes increasingly richer in tritium; the gaseous hydrogen, already containing less tritium than the water from which it was generated, is further depleted in tritium as it passes up the column. The dilute

tritium-contaminated (feed) stream enters the first stage contactor at the level where its tritium concentration corresponds to that in the reflux water descending the column. The tritium-poor hydrogen stream leaving the top of the column can be routed for disposal (assumed to be as HTO) to the atmosphere. The CECE process needs only five overall stages to concentrate 90% of the entering tritium into 1% of the original volume.

Development of the CECE process at Mound Laboratories⁴⁻⁷ and at Chalk River⁸⁻¹⁰ has focused on three major areas of investigation: (1) effects of tritium exposure on electrolysis cell operation, (2) effects of tritium exposure on the hydrophobic exchange catalyst, and (3) operation of a pilot CECE unit.

The solid polymer electrolyte (SPE) cell manufactured by General Electric has been chosen for use in the CECE system because of the low volume of liquid held in the cell and because the electrolyte is fixed within the cell. Since the cell membrane in which the electrolyte resides is constructed of a polymeric material, extensive tests of stability to tritium beta irradiation have been conducted. These have so far shown no measurable loss in cell performance.

The hydrophobic exchange catalyst also contains organic polymer that might be damaged by radiation. Maximum exposures to tritium irradiation have so far shown only small effects; some 80% of its original activity is retained by the catalyst system.

The CECE process has been under development for about six years. It offers high separation factors per stage and more simple and compact processing equipment than do most other processes. However, there are remaining areas that need further study:

1. The process is still at a small pilot-plant stage, and costs for a large installation are not firm.
2. Although tritium will be the major radioisotope present in the feed to the process, it will not be the only one. The radiation effects of other radioisotopes on the polymer need to be defined.
3. The presence of dilute nitric acid in the water needs to be determined, and limits (if any) in its concentration need to be defined.

A.3 Molecular Excitation (Laser)

The most recently proposed method for the separation of hydrogen isotopes is selective molecular excitation. Since tritium-substituted compounds such as water have greatly different energy levels than do their protium analogs, it is possible, using narrow-band laser radiation, to excite one isotopic species selectively and permit that species to react preferentially. Extremely high separation factors are theoretically possible.¹¹⁻¹³

Laser isotope separation is attractive because of the potential of single-stage separation factors by higher orders-of-magnitude than those of the other known processes for hydrogen isotope separation. Moreover, the separation energy need only be applied to the minor constituent — a decided advantage when trace quantities of a species are to be removed from a large volume of feed material.

A plausible method to detritiate low-level aqueous waste by molecular photoexcitation has been formulated. The process, which is conducted in the gas phase, consists of isotopically selective photodissociation of HTO in the presence of excess hydrogen to scavenge the photo products (Fig. A.4). This photodissociation is achieved by a two-step process employing

- DEVELOPER MOUND LABORATORIES
- PROCESS HIGHLIGHTS SELECTIVE PHOTO-DISSOCIATION OF GAS PHASE HTO VIA INFRARED LASER AND ULTRAVIOLET FLASH LAMP; HT AND O₂ ARE SEPARATED AND THE HT IS ENRICHED BY RECYCLING
- PROCESS ADVANTAGES LOW ENERGY COSTS – HIGHEST SEPARATION/CONCENTRATION FACTOR
- PROCESS DISADVANTAGES PROCESS NOT DEMONSTRATED AND IS IN EARLY DEVELOPMENT STAGE

TRITIUM ISOLATION
LASER ISOTOPE SEPARATION

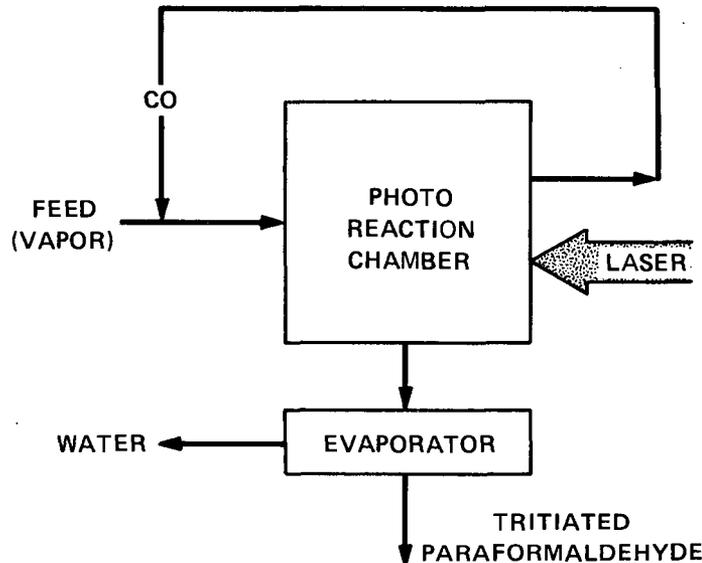


Fig. A.4. Laser isotope separation.

an infrared laser and an ultraviolet flashlamp. The infrared laser radiation selectively excites one of the vibrational modes of HTO, whereas the ultraviolet radiation has sufficient energy to photodissociate only the vibrationally excited HTO but not the ground-state water. The photoproducts, T and OH, both react rapidly with H₂ to produce HT and H₂O respectively. The tritium can be enriched in the hydrogen stream by recycling, and the tritiated product is obtained by a physical separation of the water from the hydrogen gas. This process is in the small proof-of-principle stage, and work has largely been centered on the development of laser systems.¹³

A.4 Fractional Distillation

Distillation is a well understood technique employing simple equipment that is used in many industrial processes to separate diverse constituents of solutions through differences in boiling points. Enrichment of tritium by distillation of tritiated water differs considerably from the ordinary industrial distillation processes because of the extremely low initial tritium concentration and because of the very small differences in normal boiling points ($\sim 1-3^\circ\text{C}$) between the H_2O , HDO , and HTO .

The separation factor is estimated to be ~ 1.05 at 60°C and is expected to diminish with either increasing temperature or increasing total pressure.¹⁴ Accordingly, fractional distillation at reduced pressure appears attractive from the viewpoint of increasing the separation factor. The gain in the separation factor obtained in this way, however, would be obtained at the expense of a larger plant to accommodate the increased volumetric flow for a given mass throughput.

Fractional distillation of water has been employed extensively in the production of heavy water, but the large plant size and its high thermal energy consumption lead to high capital and operating costs. These high costs are due primarily to the large number of stages, the high reflux ratios, and the low separation factor. Some reduction in plant size at the same capacity may be possible by using a packing material that has a higher throughput-to-volume ratio per theoretical plate than that of the bubble-cap plates. More effective utilization of process heat or use of vapor recompression systems would also contribute to lower operating costs.

A.5 Conventional Electrolysis

Decomposition of water by electrolysis in the presence of an electrolyte is a well established process.¹⁴ When deuterium is present, hydrogen is discharged preferentially over deuterium at the cathode because of the higher overvoltage associated with the discharge of deuterium. In addition, the equilibration of discharged HD with the liquid at the cathode surface also favors the retention of deuterium in the liquid. The equilibrium constant is reported to be 3.88 at 25°C .⁷ The separation factor for the $\text{H}_2\text{O}-\text{D}_2\text{O}$ system by electrolysis in large heavy water plants can range up to 8. This separation factor is influenced by the electrode material as well as by the choice of operating conditions.

The enrichment of tritium by electrolysis has been studied on a relatively small scale by a number of investigators. The reactions are, of course, similar to those for the enrichment of deuterium; high separation factors can be obtained and the process is certainly feasible. However, the use of conventional electrolysis alone to enrich tritium from very low concentrations in large volumes of water does not appear to be an attractive option. The application of electrolysis to D_2O production recovers only about 30% of the D_2O in the feed. This could be improved at the expense of additional electrolysis stages; however, the electrical power consumption seems likely to prove excessive even for an optimum electrolytic cascade.

Conventional electrolysis appears to be an attractive finishing step when used to upgrade the enriched product (e.g., 10 to 50% D_2O) from more economical processes (dual temperature $\text{H}_2\text{S}-\text{H}_2\text{O}$ exchange; $\text{H}_2-\text{H}_2\text{O}$ exchange, etc.). Such combinations have been used in the large-scale production of heavy water.

A.6 Bipolar Electrolysis

Special electrodes are placed between the terminal electrodes in the bipolar electrolysis process (Fig. A.5) to provide a number of compartments and electrolytic stages in an

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- DEVELOPER ORNL
- PROCESS HIGHLIGHTS COUNTERCURRENT ELECTROLYTE FLOW, ELECTRODES HYDROGEN PERMEABLE
- PROCESS ADVANTAGES LESS POWER CONSUMPTION, HIGH SEPARATION FACTOR
- PROCESS DISADVANTAGES BENCH-SCALE STATUS, LOW CAPACITY

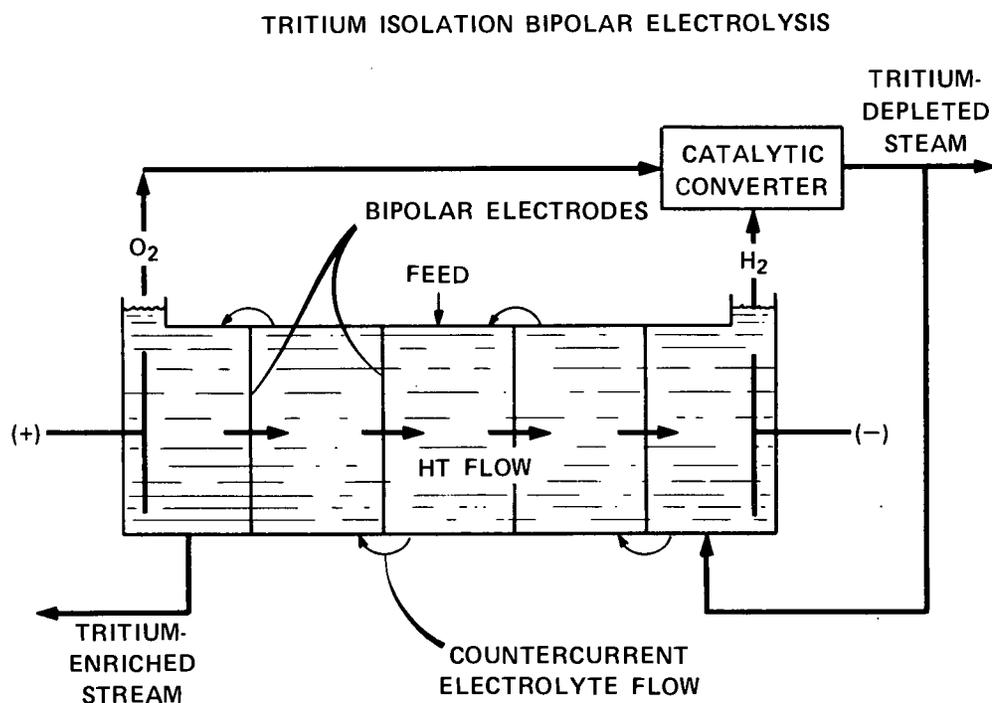


Fig. A.5. Bipolar electrolysis.

electrolysis cell.^{15,16} The bipolar electrodes are permeable to elemental hydrogen and its isotopes, but are impermeable to the aqueous electrolyte. At such an electrode, the hydrogen isotopes are electro-reduced to the elemental (atomic) state at the cathodic surface, permeate (via diffusion) the electrode, and are electro-reduced at the anodic surface. Since all three of these processes are more rapid for the lighter isotope of hydrogen, the protium moves preferentially toward the terminal cathode. Hydrogen generated at the terminal cathode is catalytically recombined with oxygen from the anode. The water (depleted in tritium)

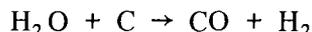
is returned in part to the cathode compartment as reflux and is in part discarded as tritium-depleted waste for disposal. The reflux water, along with the feed stream (with similar concentration) introduced at the cascade compartment, provides an electrolyte flow counter-current to the protium flow. As a consequence, tritium is enriched in the water as it approaches the terminal anode compartment. By using an adequate number of compartments or by connecting several such cells in series, a cascade system can be created to provide tritium-rich and tritium-poor waters of the desired composition. Measured separation factors for protium-tritium range from 8 to 20 (Ref. 16).

Bipolar electrolysis offers a substantial reduction (a fivefold factor) in energy required from that for conventional electrolysis.¹⁷ This technique may well be capable of development into an attractive process for the enrichment and recovery of tritium. However, at present, the process is in a small-scale laboratory stage. Laboratory-scale units are capable of processing only 250 g/d of H₂O; consequently, no conceptual plant cost data are available. A major drawback for the system appears to be the low water throughput of the cells; very large electrodes or many units in parallel would be required to provide the water throughputs necessary for the reprocessing plants.

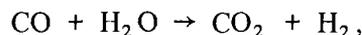
A.7 Cryogenic Distillation of Hydrogen

The enrichment of tritium by fractional distillation of liquid hydrogen is similar in principle to the enrichment of this isotope by fractional distillation of water. However, hydrogen distillation must obviously be done at very low temperatures (−250°C if distillation is to be done at atmospheric pressure). The distillation of liquid hydrogen requires a much smaller plant than does water distillation, both because hydrogen distillation uses higher pressures (1 to 2 atm) and because the separation factor per stage is much higher. A separation factor of 2 to 3 per stage has been estimated for the distillation separation of H₂ from HT. An advantage of cryogenic distillation over the dual-temperature H₂S-H₂O exchange lies in the lack of corrosive compounds in the liquid hydrogen. On the other hand, materials for long-term service at −250°C may require additional development.

It is obviously necessary to convert the large volume of tritiated water to elemental hydrogen if the process is to be used. Such conversion could be done either by electrolysis or by use of the water-gas and water-gas shift reaction sequence. The reactions are



and



and the H₂ must be separated from the CO₂ and any impurities introduced by the carbon. The conversion of water to hydrogen, by whatever process, introduces some economic penalty¹⁴ as does the necessary cooling of the large volumes of H₂ to cryogenic temperatures. It should be noted that cryogenic distillation is actively studied both at Mound Laboratory¹⁸ and at Chalk River.¹⁹ However, such distillation has never been used for the large-scale production of deuterium or heavy water in the United States, and extensive process development seems certain to be required. The very large inventory of molecular hydrogen and the

possible consequences of its escape are probably the least attractive features of this separations process.

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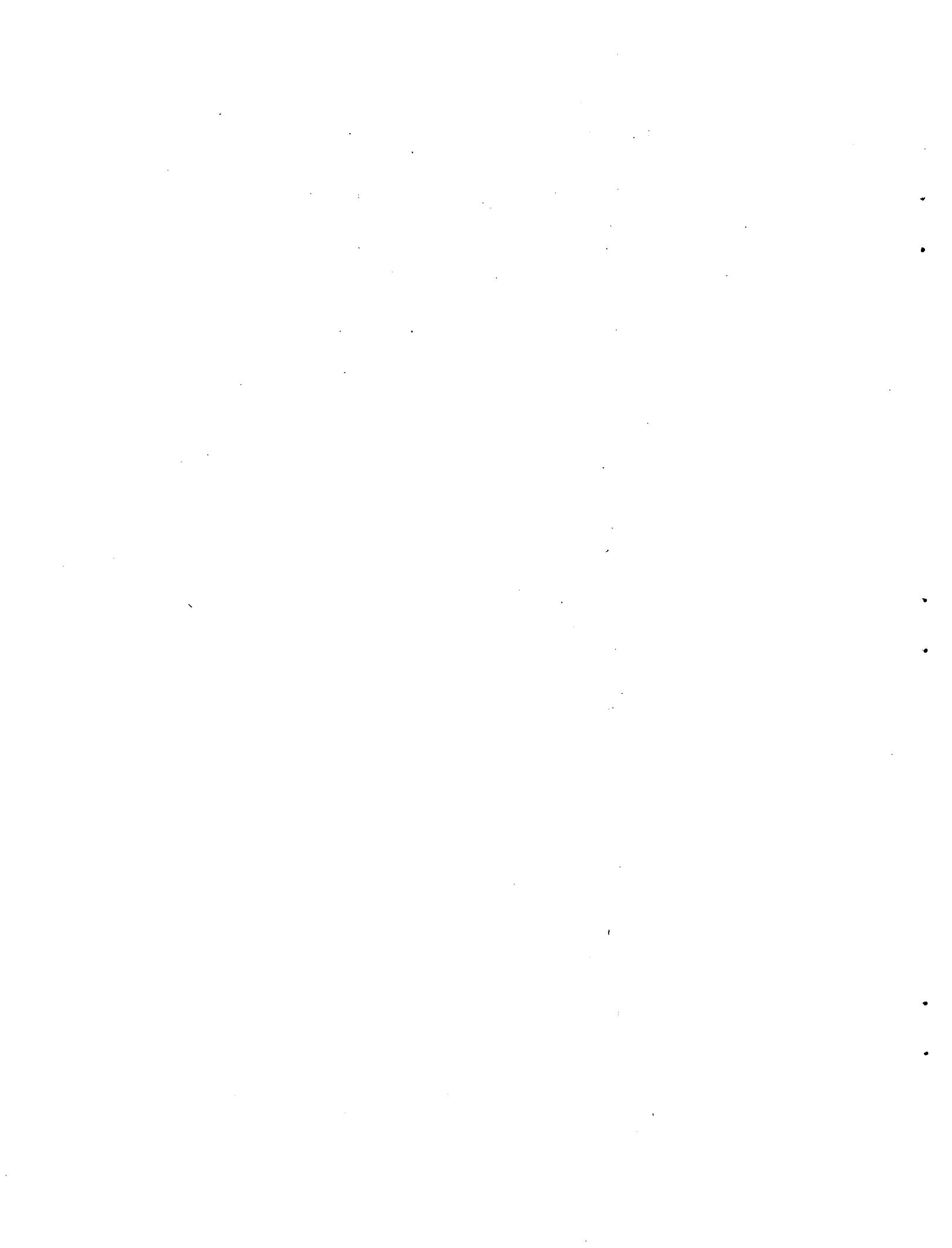
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APPENDIX B
METHODOLOGY FOR ESTIMATING CAPITAL AND
OPERATING COSTS OF RETENTION OPTIONS

B.1 Estimation of Incremental Capital Costs

The Exxon Nuclear Co., Inc. (ENC) methodology for estimating incremental capital costs is initiated by dividing the primary process operations into more than 30 process or equipment modules (Table B.1). The particular reprocessing plant mission dictates the

Table B.1. Module identification for retention options^a

Module and function	Reactor type	
	LWR ^b	LMFBR
1 Fuel receiving	X	X
2 Sodium deactivation	—	X
3 Fuel shearing	X	X
4 Tritium isolation (voloxidation)	—	—
5 Tritium isolation (isotope separation)	—	—
6 Dissolution	X	X
8 Feed preparation (U)	X	X
9 Dissolver off-gas	X	X
10 Vessel off-gas	X	X
12 Solvent extraction (U)	X	X
13 Solvent extraction (Pu)	X	X
14 Solvent treatment	X	X
15 Acid and water recycle	X	X
16 HLW concentration	X	X
17 ILW treatment	X	X
18 LEU purification	X	—
19 LEU conversion	X	—
20 Fission product conversion	X	X
22 Head-end off-gas system	X	X
23 U-Pu conversion	—	—
25 HLW solution storage	X	X
26 HLW solidification	X	X
27 Fuel and waste storage	X	X
28 Fission product storage	X	X
29 Cladding storage	X	X
30 Acid makeup facility	—	—
31 Hull dryer system	—	—
32 Tritiated steam generator	—	—

^aFrom *Evaluation of Reprocessing Configurations for Nuclear Energy Center*, ORNL/Sub-7501/6, Vols. I through III, Exxon Nuclear Company, Richland, Washington (June 1980); modules identified with X are those required for base-case (LWR-I and LMFBR-I) plants.

^bFueled with low-enrichment uranium (LEU).

number and the capacity of these modules. The many process modules are identified and their characteristics and capacities are specified by considering the water and tritium balances for each of the retention options (see Sect. 4.1). Each module that needed to be added to the base case or that differed in size or function from the corresponding module in the base case was identified and characterized. The affected modules are identified in Table B.2 for each of the retention options. Incremental capital costs were derived from the affected module costs and their variance from the base-case module costs.

Table B.2. Process modules added or modified in retention option

LWR and LMFBR	Module affected ^a
Case I	Module 15, vaporizer increase (mod)
Case II	Module 15, vaporizer elimination (mod)
Case III	Module 4, tritium isolation—voloxidation (add)
Cases IVA and B	Module 5, tritium isolation— ³ H separation (add) Module 10, drier (add) Module 15, vaporizer decrease (mod)
VA and B	Same items as IV
VI	Module 10, vessel off-gas system (add) Module 14, wash column and centrifuge (add) Module 15, separate fractionator system for partitioned tritium (mod) Module 16, concentrator (add) Module 22, hydrogen (T ₂) combiner and air treatment system (add) Module 30, acid makeup facility, NO _x scrubber (add) Module 31, clad hull drier system (add) Module 32, tritiated water steam generator system (add)

^aParentheses denote modification (mod) or addition (add).

B.2 Estimation of Total Direct Field Costs

Careful cost estimates (using the best available information) were prepared for the major process equipment in each of the affected modules. In addition, the module costs included all items required within the module. Examples of such items include shielding walls, shielding windows, manipulators, cell linings, wall penetrations, piping jumpers, and small equipment items such as steam jets, pumps, etc. Cost estimates for some structural materials (i.e., concrete and rebar) were based on estimates of the quantity required. Cost estimates for in-cell support equipment (piping, instrumentation, and electrical materials)

were obtained from a recent study¹ by applying suitable factors to the cost estimates for major equipment items.

“Balance of building” costs were then estimated to cover the costs of such plant segments as service galleries, laboratories, control rooms, utility distribution systems, and maintenance facilities.

“Balance of plant” costs covered such items as site preparation, boiler plants, cooling water systems, utilities, and warehouses. The “balance of building” and “balance of plant” costs were factored from data available from Exxon Nuclear’s proposed commercial plant.

Analyses of previously reported^{1,2} cost estimates yielded the ratios of costs of “other materials,” “balance of building,” and “balance of plant” to costs of major process equipment (Table B.3). These ratios, along with estimates of major process equipment costs, were used to obtain total direct field costs.

Table B.3. Ratio of other materials, balance of building, and balance of plant costs to process equipment costs

Cost types	Ratio
Equipment cost	1.0
Other materials	2.3
Balance of building	1.5
Balance of plant	0.8
Total	5.6

It is clear that, when these factors are applied, the total direct field costs are 5.6 times greater than major process equipment costs.

Direct field costs represent that portion of project costs expended for design, fabrication, and installation of major equipment and for purchased material plus directly applied construction labor. Field distribution costs and home office costs must be added to direct field costs.

Field distributable costs are incurred in providing temporary construction facilities, utilities, communications, and equipment rental. These costs are estimated to be 30% of the direct field costs.¹

Home office costs must be added to the direct field costs to cover overall project management personnel and facility costs, purchasing, expediting, vendor quality-control scheduling, estimating, and accounting. Home office costs are estimated to be 25% of direct field costs.¹

A contingency must be added to cover items that may be specifically unpredictable but, based on experience, can be anticipated to occur. These items include adverse weather, labor problems, material shortages, rework, and problems related to scale-up from laboratory or pilot-plant stage to production systems and equipment. Problems related to scale-up

are of particular importance in the cases considered herein, which is the most significant reason for adopting a high contingency of 55%. This contingency is applied to the total of direct field costs plus field distributable costs and home office costs.

The total project cost assessed in this manner is clearly 2.40 times the total direct field costs.

B.3 Sample Calculation of Total Project Cost

The following example is shown in some detail to illustrate the estimation of total costs for this study.

Retention option LWR-III requires the addition of Module 4 containing a voloxidizer and a voloxidizer off-gas system and substitution of a much smaller vaporizer (for the large one of LWR-I) in Module 15.

A prototype voloxidizer capable of treating 1 MTHM/d of LMFBR fuel (or 4 MTHM/d of LWR fuel) has been estimated to entail the following costs:

Design:	\$540 000, including 20% contingency (1981 dollars)
Fabrication:	\$1 480 000, including 20% contingency (1982 dollars)
Installation:	\$730 000, including 20% contingency and \$80 000 in supervisory costs (1983 dollars)

Removing the contingency and the supervisory costs, backing the costs to first quarter 1980 dollars (at 10% inflation rate per year), and rounding to the nearest \$100 gives the following values:

Design	\$409 100
Fabrication	\$1 019 300
Installation	\$407 000
Total	\$1 835 400 for 4 MTHM/d of LWR fuel (with voloxidizer)

When scaled to a voloxidizer capable of 8 MTHM/d of LWR fuel, the fabrication cost (using a factor of $2^{0.7}$) becomes \$1 655 900. The design and installation costs are assumed to be independent of size in this range. As a consequence, the cost for the LWR voloxidizer becomes:

Design	\$409 100
Fabrication	\$1 655 900
Installation	\$407 000
Total	\$2 472 000

The tritium off-gas system is estimated to cost \$2 400 000. The total major equipment costs for Module 4 are, therefore, \$4 872 000. For option LWR-III, however, Module 15 requires fabrication and installation of a small (\$28 000) vaporizer instead of a larger one (\$209 000) for the base-case LWR-I. Incremental costs of major equipment for case LWR-III, therefore, are:

	\$4 872 000
	+ 28 000
	- 209 000
Total major equipment costs	\$4 691 000

To obtain total direct field costs, the following additions are made:

Major equipment	\$4 691 000
Additional materials (factor 2.3)	\$10 789 000
Balance of process building (factor 1.5)	\$7 037 000
Balance of plant (factor 0.8)	\$3 753 000
Total direct costs	\$26 270 000
Field distributable (30%)	\$7 881 000
Subtotal	\$34 151 000
Home office (25%)	\$8 538 000
Subtotal	\$42 689 000
Contingency (55%)	\$23 479 000
Total project costs	\$66 168 000

The incremental capital cost of LWR-III over the base-case (LWR-I) is, accordingly, \$66 168 000.

B.4. Estimation of Incremental Operating Costs

The annual operating cost of the facilities considered is the sum of many elements. The key elements considered for this evaluation include labor, consumables, utilities, and equipment replacement. Elements of the annual operating cost specifically excluded from this estimate include taxes, licensing fees, decommissioning, environmental monitoring, insurance, and waste disposal.

In all cases, only the incremental operating cost difference from the base case was determined.

The incremental operating costs were based on the following assumptions:

1. Labor costs were estimated at \$48 000 per man-year. This figure includes elements such as benefits, hiring expense, travel, and relocation. The estimates of staff required to operate a facility were based on the following:
 - Certain functions (e.g., management, laboratory, guard force, and environmental monitoring requirements) are assumed to remain constant for all cases; therefore, no incremental costs were assigned to these functions.
 - The operating and maintenance staffs were adjusted to account for the different operating modes of the cases considered. The increments estimated for the several operations are listed in Table B.4.

Table B.4. Incremental labor

Case	Staff increment	
	Operations	Maintenance
LWR-I	—	—
LWR-II	—	—
LWR-III	4	2
LWR-IVA	8	4
LWR-IVB	8	4
LWR-VA	8	4
LWR-VB	8	4
LWR-VI	4	2
LMFBR-I	—	—
LMFBR-II	—	—
LMFBR-III	4	2
LMFBR-IV	8	4
LMFBR-V	9	4
LMFBR-VI	4	2

2. Consumables were estimated empirically from the following elements of cost:
 - Office supplies at 0.05 X labor costs;
 - Maintenance and operating supplies (SWP clothing, small tools, decontamination materials, etc.) at 0.05 X labor costs;
 - Chemical usage for the various cases was evaluated; all chemical costs were determined to be negligible except for the oxygen requirements for case III and the sucrose requirements for case VI for both LWR and LMFBR fuels.
3. Annual utility costs were estimated empirically using the following principal cost elements:
 - Steam at \$5.00 per 1000 lb;
 - Electricity at \$0.02/kWh; cooling water requirements were converted to kilowatt hours by considering the water pumping rates involved;
 - Annual equipment replacement was estimated at 0.75% of the total project capital cost for each option.

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APPENDIX C
DESCRIPTION OF OPTIONS FOR TERMINAL DISPOSAL

C.1 Shallow Trench Burial

Near-surface burial has been used routinely for several decades for the disposal of low- and intermediate-level radioactive wastes in the United Kingdom^{1,2} and Canada,³⁻⁵ as well as at several federal installations in the United States.⁶⁻¹¹ Moreover, the disposal of low-level radioactive waste has been practiced at a total of seven commercial sites in the United States.^{6,12,13} It has been reported that some 645 000 Ci of tritium was buried in 1971 at one of the commercial sites.¹³ Only three commercial burial sites are accepting wastes at present; these are those at Hanford (Washington), Beatty (Nevada), and Barnwell (South Carolina). The availability of any of these sites in the future for the burial of wastes containing moderate quantities of tritium appears questionable.

The most economical fixation technique for this disposal option would appear to be the incorporation of tritiated water into concrete that is then poured into steel drums. The concrete could hold some 20 to 25% of water by weight and provide a dilution factor of ~1.8. If the proposed regulations¹⁴ (see Sect. 2, Table 2.1, this report) are adopted by the Nuclear Regulatory Commission, the two acceptable modes of shallow burial would be:

1. Class E – no administrative control, worker/reclaimer access; the tritium concentration may not exceed 0.05 Ci/m³.
2. Class D – administrative control for 150 years, followed by reclaimer access; the tritium concentration may not exceed 94 Ci/m³.

Table C.1 shows the volumes of tritiated water concentrate, tritium concentrations, volumes of tritiated concrete, and specific activities of the concrete for each of the LWR

Table C.1. Quantities and specific activities of tritiated water

Retention case	Tritiated water			Concrete	
	Volume (m ³)		Concentration (Ci/m ³)	Volume (m ³ /year)	Specific activity (Ci/m ³)
	Annual	Total			
LWR-II	3 880	77 600	0.182	7 500	94
LWR-III	14.4	290	53 600	27	28 600
LWR-IV	900	18 000	858	1 600	480
LWR-V	86.4	1 728	8 930	160	4 800
LWR-VI	2 660	53 160	298	4 900	160
LMFBR-II	8 190	163 740	13.8	15 000	8
LMFBR-III	14.4	290	8 300	27	4 400
LMFBR-IV	900	18 000	133	1 600	74
LMFBR-V	86.4	1 728	1 377	160	740
LMFBR-VI	2 635	52 700	41.7	4 700	24

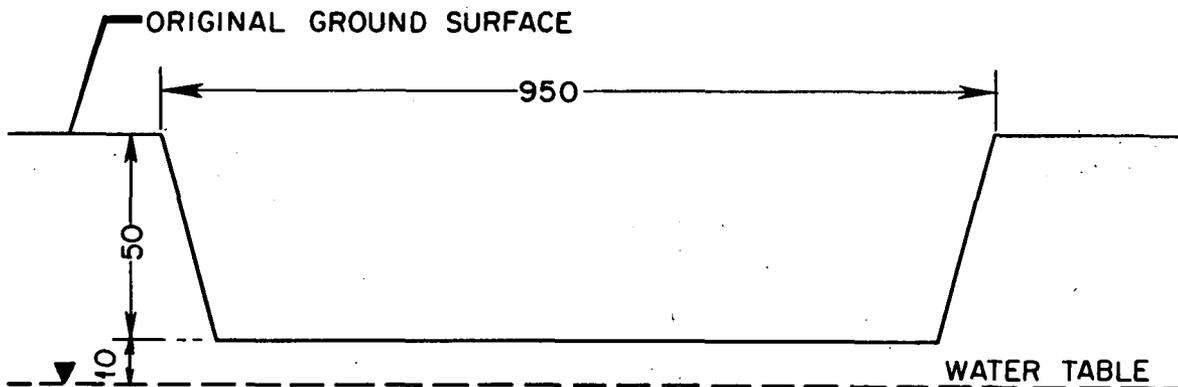
and LMFBR retention options. None of the concretes can qualify for Class E burial even after quite unreasonable long periods of interim storage. Four of the options (LWR-II, LMFBR-II, LMFBR-IV, and LMFBR-VI) yield concretes that qualify immediately for Class D burial; concrete from LWR-VI could qualify after interim storage for ten years. All cases yield large volumes of concrete of relatively low specific activity. None of the other concentrates (those from LWR-III, LWR-IV, LWR-V, LMFBR-III and LMFBR-V) can qualify for surface-trench burial.

C.2. Concrete Monoliths at Intermediate Depth

The proposed guidelines¹⁴ of the Nuclear Regulatory Commission suggest that burial of the tritiated water by fixation in concrete monoliths at intermediate depths may be an acceptable method of disposal. Such emplacement in concrete monoliths is presently under examination for the disposal of partially decontaminated salt at Savannah River and at Hanford. For such burial to be employed, the top of the monolith must be more than 10 m below grade, and the water table must be well below the bottom of the monolith. These requirements seem to be met at many sites (including Savannah River and Hanford) but are unlikely to be met at the hypothetical site at Oak Ridge, Tennessee.

Such disposal might reasonably be done in the following manner.¹⁴ The entire disposal area would be excavated to a depth of ~20 m; the bottom of the excavation must be above the water table. A layer (~1.5 m thick) of compacted and essentially impermeable clay is placed in the bottom of the excavation, and a permeable (sand- or gravel-filled) layer is then emplaced to permit the collection and monitoring of any water that might penetrate the disposal area (Figs. C.1 through C.4). Additional layers of compacted, impermeable clay would then be emplaced to a depth of 8 m above the permeable layer. The pit would then be backfilled with native soil to the original ground surface. For the disposal to proceed,

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NOTE: DIMENSIONS ARE IN FEET

Fig. C.1. Excavated pit.

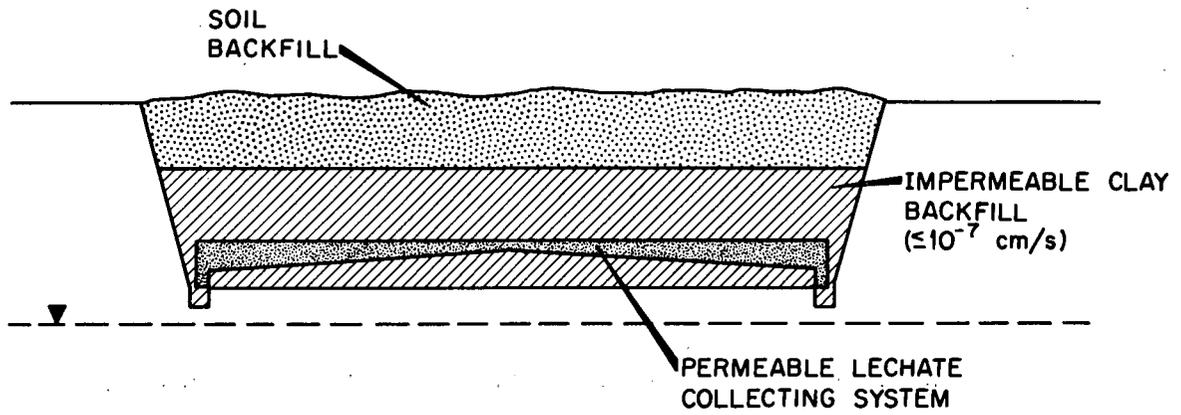


Fig. C.2. Backfilled pit with constructed leachate collection system.

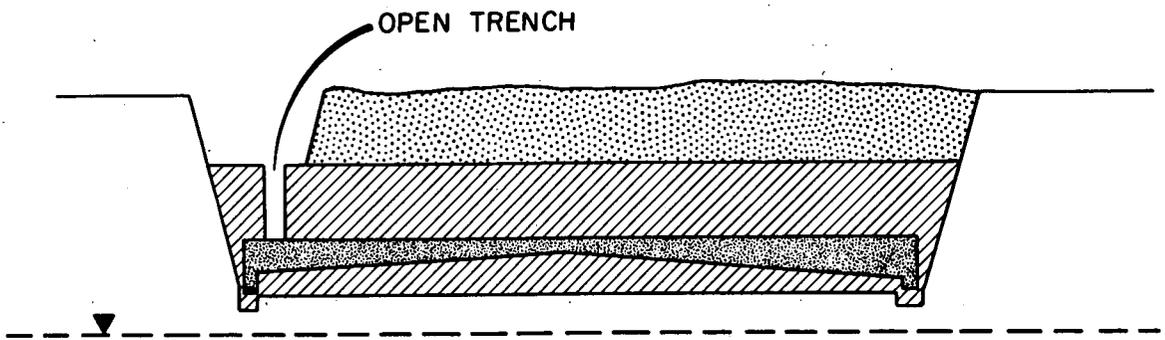


Fig. C.3. Trench construction for concrete placement.

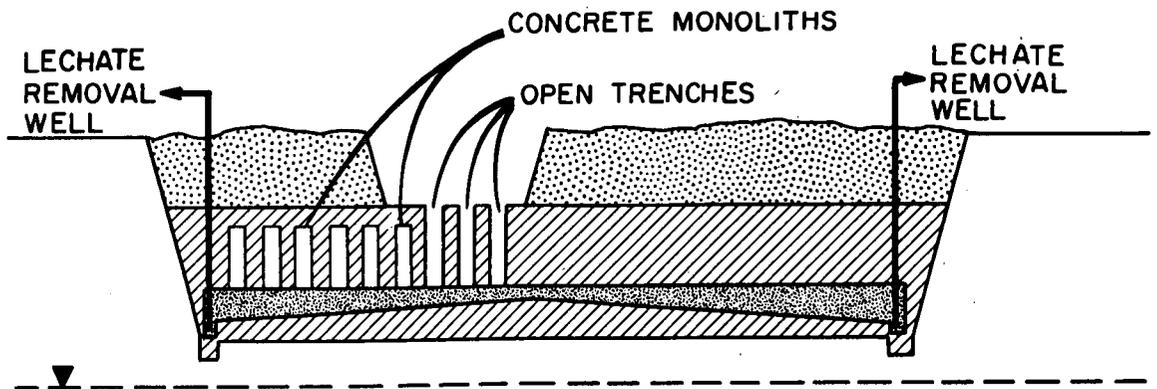


Fig. C.4. Completed trenches with concrete emplaced and backfilled; open trenches ready for concrete placement.

the section to be used would be reexcavated to the top of the impermeable clay layer, and trenches (maximum 14 m in length, 6 m wide) would be excavated in the thick clay layer. The tritiated concrete would be poured into the trench; after the concrete has solidified, the trench would be covered with a compacted layer (1.5 m thick) of essentially impermeable clay. The used portions of the pit would then be backfilled with native soil to the original ground level. Twenty years of plant operation would require some 600 trenches to accommodate the largest volume of tritiated waste that would seem realistic to handle in this manner (option LMFBR-II). If the trenches could be placed in a square array with a 4-m separation between adjacent sides and an 8-m separation between adjacent ends, the final repository would occupy some 13.5 hectares (34 acres).

The proposed guidelines¹⁴ show the concentration limits for Class C (intermediate-depth burial; no administrative control; no reclaimer access except well water) to be the same (94 Ci/m³) as for Class D. Accordingly, only cases LWR-II, LMFBR-II, LMFBR-IV, and LWR-VI provide concretes that are immediately suitable for such disposal; all other concentrates would yield such concretes only after relatively long-term storage as liquids. On the other hand, all concentrates are well below the maximum concentration for Class B (intermediate-depth burial; administrative control for 150 years followed by no reclaimer access except well water) for which the proposed limit is 430 000 Ci/m³.

It seems plausible that, given a site in which the water table is sufficiently below grade, such an emplacement would suitably immobilize tritium and prevent its adverse interaction with the biosphere. The essential lack of moving water, the massive size of the monoliths, and, especially, the thick, protective, and essentially impermeable layer of clay would seem to preclude appreciable tritium migration. A considerable assessment of the extent and permanence of the tritium confinement would, however, seem necessary before such a procedure could be certain of acceptance.

A greater problem could be posed by the logistics of pouring the concrete without sensible loss of tritiated water vapor and without undue exposure of the staff of the disposal system. Equipment can be designed to mix batches of concrete without appreciable employee exposure, but safely pouring the monoliths may prove to be a formidable undertaking. Indeed, it may prove that the more concentrated waters (options LWR-III and -V and LMFBR-III and -V) are not manageable by this method. For those concentrated streams (two of which aggregate 290 m³ of water during the 20-year period and two others that aggregate 1728 m³), it may prove necessary to solidify the tritiated waters in concrete within sealed drums and to emplace the drums within the trenches for incorporation within concrete monoliths of appropriate size.

C.3 Discharge to Deep Wells

The Committee on Waste Disposal of the National Academy of Sciences generally endorsed the proposal that portions of deep sedimentary basins and coastal plain areas could be used as disposal reservoirs for liquid wastes containing radioactivity.¹⁵ Several reconnaissance studies of sedimentary basins were performed by the U.S. Geological Survey.¹⁶ In a subsequent review,¹⁷ the National Academy of Sciences noted that truly satisfactory disposal sites would be permeable sedimentary rock layers at least 3000 ft deep in geologic basins confined above by relatively thick impermeable strata (i.e., shale or salt deposits) so that movement of liquid would be restricted vertically but not horizontally.

The U.S. Environmental Protection Agency is not categorically opposed to waste disposal by deep-well injection¹⁸ but is on record as "being opposed to emplacement of materials by subsurface injection without strict controls and a clear demonstration that such emplacement will not interfere with present or potential use of the subsurface environment, contaminate groundwater resources, or otherwise damage the environment."

The disposal of chemical and petrochemical wastes in deep wells has increased markedly in the United States during the past 25 years;¹⁸ however, this method has seldom been used in this country for the disposal of radioactive wastes.¹²

Trevorrow et al.¹⁹ examined the possibility of disposing tritium-bearing aqueous waste in deep wells. They concluded that suitable sites were abundant in the United States; however, the only deep-well disposal of tritium to date has been done at the Idaho National Engineering Laboratory. There, low-level liquid wastes from the Idaho Chemical Processing Plant have been disposed of at a rate of $\sim 11\,400\text{ m}^3/\text{year}$ (3×10^6 gal/year) through a well ~ 600 ft deep into the Snake River aquifer.^{16,17,20,21} Nearly all the activity except tritium is removed from the wastes by distillation and ion exchange processes before discharge. Since 1952, approximately 22 000 Ci of tritium at an average concentration below 5×10^{-4} Ci/m³ have been discharged into the aquifer; the discharge rate since 1961 has been such that a quasi-steady-state inventory of 14 000 Ci of this isotope has been maintained in the aquifer.²¹ The distribution and migration of radionuclides (along with stable sodium, chlorine, and chromium) in the aquifer has been monitored for many years by sampling from some 45 observational wells near and downstream from the discharges. The tritium plume (with boundaries taken as 2×10^{-6} Ci/m³) occupied an area of about 15 square miles in 1972.²¹

Radioactive liquid wastes have been disposed of into deep water-bearing strata at a number of sites in the USSR.²²⁻²⁵ Spitsyn et al.²³ state that some 1.2×10^6 m³ of liquid with a total beta activity of 5×10^7 Ci were disposed of at Melekess between 1963 and 1970. By 1972, wastes containing as much as 1000 Ci/m³ of mixed fission products were apparently being injected at least at one site, and consideration was being given toward injecting wastes with higher specific activities.^{24,25} None of these papers mentions tritium, and none appear to tabulate waste composition, although reference 24 lists ¹⁴⁴Ce-¹⁴⁴Pm, ⁹⁵Zr-⁹⁵Nb, ¹⁴⁷Pm, ⁸⁹Sr, and ⁹⁰Sr as the major heat sources in the waste. Monitoring or migration data are not discussed in any of the Spitsyn et al. papers; however, the increasing concentrations injected seem to suggest that the Russians are satisfied with this aspect of liquid waste disposal.

C.4. Disposal by Shale Fracturing (Hydrofracture)

Oak Ridge National Laboratory (ORNL) has used the hydrofracture technique for the disposal of intermediate-level wastes for several years. By this method, which is probably unique to ORNL, the waste solution is mixed with a solids blend of cement and other additives and is injected into an impermeable and isolated shale formation at a depth of 215 to 300 m. The injection pressure is sufficient to propagate a thin, horizontal crack in the shale. As the injection of grout continues, this crack is filled by the grout and is further extended so that a thin, approximately horizontal grout sheet several hundred feet in diameter is formed. The grout sheet sets within a few days after completion of the injection

and fixes the wastes in the shale formation. The preparation of a bore hole for the injection consists of drilling a hole to and through the shale formation, lowering and cementing a casing to the total depth, and cutting the casing at the depth at which the fracture (and resulting grout sheet) is desired. A preliminary shale fracture is done by water pressure before injection of the grout. Subsequent cuts and fractures at increasingly shallower depths permit the formation of essentially parallel sheets a few feet apart.²⁶⁻²⁸

Intermediate-level waste, which is defined as a solution up to 400 Ci/m³ of mixed fission products, is generated at a rate of ~4 million gal/year from a number of processes at ORNL.²⁸ This waste is neutralized with caustic to form a sludge (~400 000 gal have been accumulated in 30 years) and a supernate. The supernate is evaporated and concentrated by about 25-fold.^{26,28} About 300 m³/year (80 000 gal) of this concentrate, which is alkaline and whose major constituent is NaNO₃, is disposed of by hydrofracture.

Experimental injections into the shale formation were performed in 1964 and 1965.²⁶ Since late 1966, many injections at depths from 250 to 266 m have been made; the largest injection²⁸ (in contained radioactivity) disposed of ~300 m³ of waste (~480 m³ of grout) containing 8900 Ci of ⁹⁰Sr, 89 000 Ci of ¹³⁷Cs, and ~4 g of ²³⁹Pu.

The grout composition can be varied (within limits) to provide the desired retention of particular radionuclides. Fly ash has been shown to improve the retention of ⁹⁰Sr in the grout, and attapulgite clay is used to retain excess water. Finely ground, local Conasauga shale has been shown to improve the retention of cesium. The dry solids mix used for intermediate-level ORNL wastes is (by wt %) 38.5% fly ash, 15.4% attapulgite, 7.7% shale, and 0.05% retarder to delay setup during injection.²⁸

The well-head pressure (typically ~2500 psi to fracture and somewhat less to propagate the fracture) is carefully monitored during injection for any sudden drops that might indicate a vertical fracture. Such vertical fractures are not uncommon in the oil industry, but the 30 experimental and operational injections at the ORNL site appear to have each produced essentially horizontal grout sheets.²⁸

Slight modifications of this successful technique offer several attractive features for the disposal of tritiated water. No water movement has been observed in test wells in the disposal area at depths below 60 m. Tritium would be effectively immobilized in thin sheets in an essentially impermeable formation at depths of more than 180 m and far below the level where water migration occurs. The ORNL practice is to make a single large (300 m³ of aqueous waste) injection approximately once per year. However, Weeren has reported²⁹ that an injection every ten days would be reasonably simple to accomplish. The total volume that may be disposed of in a single well is probably ~40 000 m³. Tritiated waters from all options could be readily accommodated by hydrofracture, although LWR-II and LMFBR-II options would require two and four wells, respectively, during the 20-year plant lifetime. The technique has proved successful (though not with high concentrations of tritium) at the Oak Ridge site; however, careful local testing would be necessary before this technique could be approved for use at other sites.

C.5 Geologic Disposal

The placement of radioactive wastes into stable geological formations can isolate these wastes from the biosphere for geologic time periods. Geologic storage is being examined

both here and abroad for the disposal of high-level and long-lived radioactive wastes. It would not be economical to prepare a geological repository for relatively short-lived radioactive species like tritium. If, however, geological repositories were opened for long-lived, high-level wastes, they might also be used for high-level, shorter half-life species.

Several geological formations have been considered for radioactive waste storage, including salt beds, shale formations, and hard bedrocks such as granite and basalt. Salt deposits have been considered for radioactive waste repositories because the existence of the salt deposit itself indicates that there is little ground-water movement in the formation. Salt also has good heat conduction properties, and its plasticity makes it self-sealing to several types of deformations. A demonstration facility for low- and intermediate-level radioactive waste disposal is currently in operation at the ASSE Salt Mine in the Federal Republic of Germany.³⁰

No geological nuclear waste repository has been built so far in the United States. Extensive studies are in progress to identify potential repository sites, categorize their geology, and determine construction costs. These studies are being managed for the DOE's Office of Nuclear Waste Isolation (ONWI).³¹

Conceptual designs of repositories have been completed. From the surface, a geological nuclear waste repository would resemble a relatively large mine. There would be a railroad siding, facilities for deep excavation, and buildings for unloading, handling, and repackaging nuclear wastes prior to isolation. Material excavated during construction of the repository would be placed in a temporary protected storage pile. (Depending on the nature of the geological formation where the repository is located, this material could be granite, basalt, rock salt, etc.) Where necessary, facilities would have concrete shielding to protect workers from nuclear radiation. Ventilation systems would be provided with filters to prevent the release of radioactive particles. A total of about 200 to 400 acres would comprise the surface area of the repository.

From the surface, several vertical shafts would lead to tunnel-like storage zones about a half mile deep. Sealed in canisters and further protected by engineered barriers, the wastes would be placed in holes excavated along the tunnels.

As each storage zone is filled, the holes, tunnels, and shafts would be backfilled and sealed. However, for some period of time prior to final closure, the wastes may be maintained in a "retrievable" condition. Following final closure, several independent barriers (some designed by man and others provided by nature) would serve to isolate the waste from man and his environment.³¹

The storage holes for high-level waste are spaced to reduce the thermal loading in the repository medium. Since the heat loading of even the highest tritium concentration is negligible, it should be possible to place tritium containers in positions interstitial to the high-level waste containers. The temperatures reached at such locations are not high enough to breach the tritium containers.

C.6 Disposal at Sea

The United States apparently began the practice of disposing packaged, solid radioactive wastes into the deep oceans very early, but only relatively small amounts of radioactive materials have been given such disposal by this country. Between 1946 and 1953, some

59 000 containers with a total of some 15 000 Ci of mixed radionuclides were dumped into the Pacific Ocean. Disposal in the Atlantic began in 1951, and by 1963 had reached a total of 30 000 containers with ~46 000 Ci of activity. However, in the 1963 to 1970 period, only 70 containers (30 Ci) were dumped into the Atlantic and 278 containers (186 Ci) into the Pacific.¹² Joseph et al.³² (and later Olivier³³) reported that U.S. operations had dumped some 80 000 Ci and 15 000 Ci, respectively, into the Atlantic and into the Pacific during these same time periods.

From 1949 to 1960, the United Kingdom discharged suitably packaged solid wastes to the sea.^{3,34} Solids of sufficiently low level were dumped at authorized locations in the English Channel at a depth of some 200 m; the English Channel location was authorized to receive up to 5000 t/year of waste (not exceeding 200 Ci of alpha activity and 4000 Ci of beta-gamma activity). An Atlantic Ocean site with a depth in excess of 3000 m was authorized to receive up to 1500 t of waste, with radioactivity limits governed only by radiation levels from the containers such as "to prevent any hazard to personnel during shipment and dumping operations."³⁴ Between 1951 and 1967 the United Kingdom is reported to have discharged some 40 000 Ci into the Atlantic Ocean.³³ As of 1970, Japan is reported to have disposed at sea (at a depth of 3000 m) some 1500 concrete-filled drums containing "several hundred" curies of radioisotopes.³⁵

A panel of experts, established by the International Atomic Energy Agency, examined the scientific and technical aspects of such disposals of radioactive materials. The panel reported³⁶ that low- and intermediate-level radioactive wastes could be safely disposed of into the sea under carefully controlled and specified conditions; they also recommended criteria for selecting disposal sites and guidance for the design of sea disposal containers. A second panel³ confirmed and extended the conclusion stated in 1961.

Preliminary studies began within the European Nuclear Energy Agency in 1965 to develop a safe and economical method for ocean disposal of solid wastes and to demonstrate the method at an international level. The first such operation, which took place in 1967, has been documented in detail.³⁷ Five member countries took part in the operation; some 35 800 containers weighing ~11 000 t and containing ~8000 Ci of solid waste were deposited at a depth of ~5000 m in the western Atlantic Ocean.

The general framework for the operational control of sea disposal was firmly established during eight sea-disposal campaigns carried out under the supervision of the Nuclear Energy Agency from 1967 to 1976. Since the first operation in 1967, up to eight European countries have taken part in these campaigns; these include Belgium, France, Federal Republic of Germany, Italy, The Netherlands, Sweden, Switzerland, and the United Kingdom.³³ Three dumping sites have been used for these operations, following recommendations from a group of international experts from NEA countries. The sites were all located in the north-eastern Atlantic at an average depth of ~5 km. The area used in 1976 is defined as a circle, 70 nautical miles in diameter, and centered on the point of 46°15'N and 17°25'W. The area has an average depth of ~4.5 km and is located ~1000 km from the European coasts. Its precise position was determined after a careful study of the network of known undersea cables in front of the European Continent. The quantities of wastes disposed of under NEA-supervised operations are shown in Table C.2.

These operations, both at national and international levels, anticipated to a great extent the system subsequently developed for the dumping of all types of waste within the

Table C.2. Quantities of radioactive waste dumped during NEA-supervised sea-disposal operations^a

Year	Dumped weight (t)	Approximate activity (Ci)	
		Alpha ^b	Beta-gamma ^c
1967	10 840	250	7 600
1969	9 180	500	22 000
1971	3 970	630	11 200
1972	4 130	680	21 600
1973	4 350	740	12 600
1974	2 270	420	100 000 ^d
1975	4 460	780	60 500 ^e
1976	6 770	880	53 500 ^f
Total	45 970	4880	289 000

^aSource: J. P. Olivier, "Seabed Disposal Practices for Packaged Radioactive Waste," *Proceedings*, International Symposium on Management of Wastes from the LWR Fuel Cycle, CONF-76-0701 (July 1976).

^bActinides.

^cIncluding tritium.

^dAlmost exclusively tritium.

^eIncluding ~30 000 Ci of tritium.

^fIncluding ~21 000 Ci of tritium.

guidelines of the London Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter.³⁸ This convention, which is concerned with the disposal of both chemical and radioactive pollutants, was organized in August 1975. Together with several other international conventions, it has served as a dominate force in preventing the seas from being overly contaminated with radioactive materials.³⁹ Finally, the International Atomic Energy Agency (IAEA) adopted specific recommendations for the application of the London Convention to radioactive wastes, including a definition of certain categories of radioactive wastes regarded as unsuitable for deep ocean disposal.⁴⁰

Unsuitable waste for sea dumping is expressed in terms of radioactivity per unit mass. Unsuitable wastes are those that contain more than 10 Ci/t of alpha emitters with half-lives longer than 50 years, those containing more than 1000 Ci/t of beta-gamma emitters (excluding tritium) and an additional limit of not more than 100 Ci/t of ⁹⁰Sr plus ¹³⁷Cs, and those containing more than 10⁶ Ci of tritium per metric ton.³³ It is clear (see Table C.1) that all of the concentrates expected from the options discussed in this study are far below the established concentration limits.

The quantities of tritium adjudged to be safely releasable to the North Atlantic Ocean are impressively large. In an assessment carried out under NEA supervision,⁴¹ the limiting capacity (Slansky's concept⁴²) of the North Atlantic was estimated to be 10¹⁵ Ci of tritium per year. The disposal of that quantity of tritium each year would lead (with tritium assumed quite conservatively to be released from its solidified state in the sealed drum at the moment

of dumping) to dose levels equivalent to the appropriate ICRP dose limits for the most exposed members of the public. While it is clear that no one advocates the disposal of tritium in these quantities, it is obvious that the North Atlantic Ocean might safely contain very large quantities of this isotope. If the world were producing 5000 GW(e) years of power from LWRs, the reprocessing plants might collect as much as 1×10^8 Ci of tritium for disposal each year. The accumulated tritium from nuclear power programs by the year 2000 has been estimated to be about 5×10^8 Ci.^{33,43}

It seems apparent that sea bed disposal of tritium solidified in concrete and contained in steel drums should be a satisfactory disposal method.

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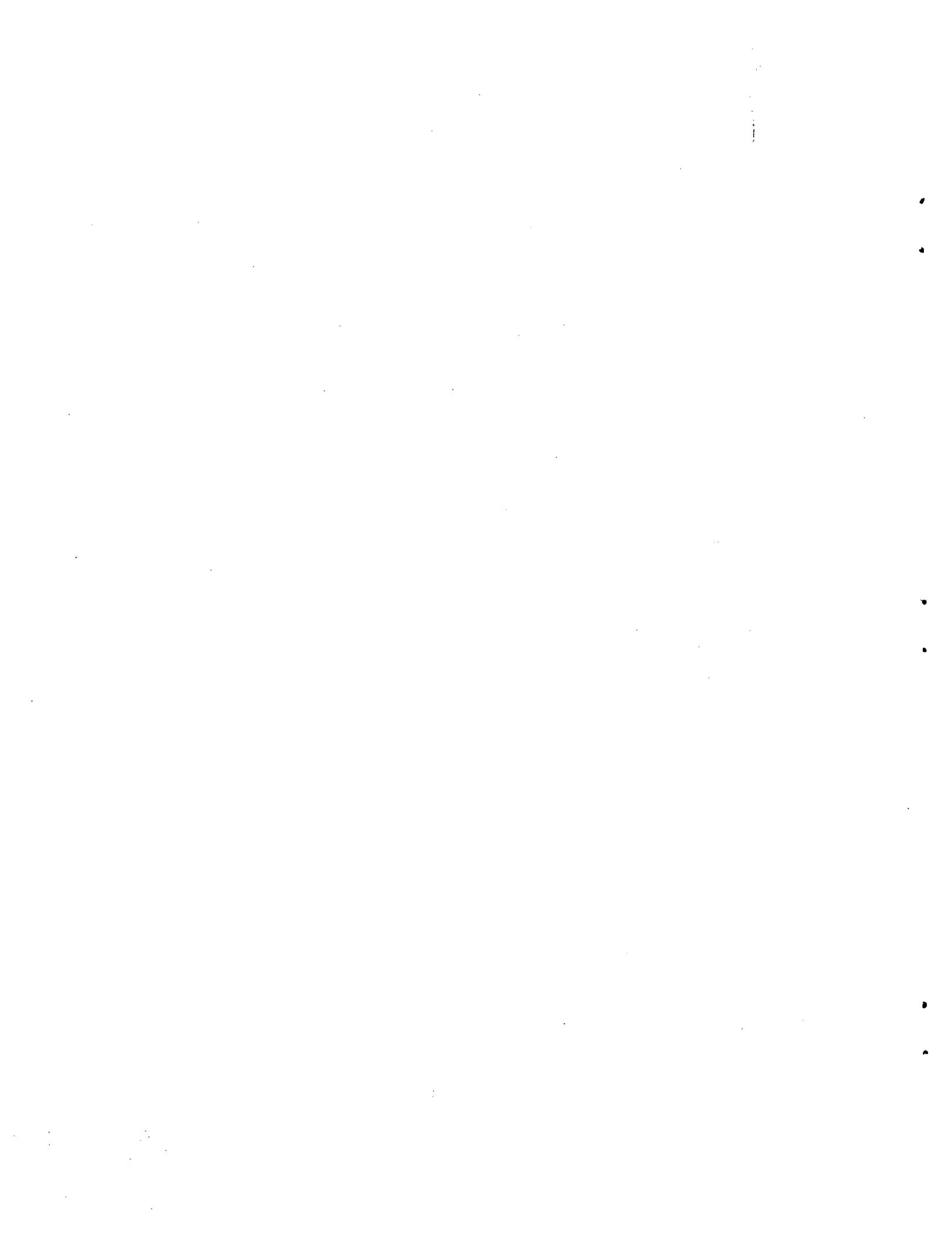
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APPENDIX D

METEOROLOGICAL AND OTHER PARAMETERS TO AIRDOS CALCULATIONS

All tritium is assumed to be released as tritiated water (HTO) from a single stack with characteristics similar to those of Table D.1. A standard equation¹ is used by the computer to estimate the rise of the momentum-dominated plume to obtain the effective stack height.

Table D.1 Assumed stack characteristics

Height, m	100.0
Diameter, m	2.90
Effluent velocity, m/s	20.40
Heat emission, cal/s	0.0

Dispersion in the airborne plume as it blows downwind is estimated from the standard Gaussian plume equation² as modified by Gifford.³ Meteorological data, accepted as appropriate for the Oak Ridge area and shown in Tables D.2 through D.5, were used as input to CONCEN for this operation. The horizontal and vertical dispersion coefficients (σ_y and σ_z) required for calculation of dispersion and depletion are those from the Atmospheric Turbulence and Diffusion Laboratory at Oak Ridge, Tennessee.^{4,5}

As the plume moves downwind, it is depleted in tritium through scavenging during precipitation of rain or snow and by deposition through interaction with vegetation or surface water as well as (very slightly) by radioactive decay. For these calculations, a scavenging coefficient of 2.8×10^{-6} /s and a deposition velocity of 10^{-3} m/s were assumed. The effective decay constant in the plume is the reciprocal (1.54×10^{-4} /d) of the mean time for radioactive decay.

Values of χ/Q (in s/m^3) are estimated for each of the 16 compass directions at ten distances from 1207 m (0.75 miles) to 72 420 m (45 miles). These values are shown in

Table D.2. Meteorological data input

Average air temperature, K	294.0
Average vertical temperature gradient of the air, K/m	
In stability, class E	0.0728
In stability, class F	0.1090
In stability, class G	0.1455
Rainfall rate, cm/year	138.00
Height of lid, m	1125

Table D.3. Frequency of atmospheric stability classes for each direction

Sector ^a	Fraction of time in each stability class						
	A	B	C	D	E	F	G
1	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
2	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
3	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
4	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
5	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
6	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
7	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
8	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
9	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
10	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
11	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
12	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
13	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
14	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0
15	0.0700	0.1400	0.1300	0.2800	0.2000	0.1400	0.0

^aSectors are numbered counterclockwise starting at 1 for due north.

Table D.4. Frequencies of wind directions and reciprocal-averaged wind speeds

Wind toward ^a	Frequency	Wind speeds for each stability class (m/s)						
		A	B	C	D	E	F	G
1	0.040	1.67	1.67	1.67	1.46	1.25	1.25	0.0
2	0.011	1.41	1.41	1.41	1.29	1.16	1.16	0.0
3	0.016	1.33	1.33	1.33	1.26	1.19	1.19	0.0
4	0.011	1.25	1.25	1.25	1.20	1.15	1.15	0.0
5	0.048	1.54	1.54	1.54	1.35	1.16	1.16	0.0
6	0.117	1.82	1.82	1.82	1.47	1.12	1.12	0.0
7	0.164	1.82	1.82	1.82	1.42	1.02	1.02	0.0
8	0.044	1.67	1.67	1.67	1.50	1.32	1.32	0.0
9	0.024	1.70	1.70	1.70	1.58	1.45	1.45	0.0
10	0.020	1.67	1.67	1.67	1.49	1.30	1.30	0.0
11	0.039	1.79	1.79	1.79	1.58	1.37	1.37	0.0
12	0.053	2.04	2.04	2.04	1.77	1.49	1.49	0.0
13	0.082	1.89	1.89	1.89	1.61	1.33	1.33	0.0
14	0.102	2.04	2.04	2.04	1.63	1.21	1.21	0.0
15	0.150	1.89	1.89	1.89	1.49	1.09	1.09	0.0
16	0.088	2.00	2.00	2.00	1.60	1.19	1.19	0.0

^aWind directions are numbered counterclockwise starting at 1 for due north.

Table D.5. Frequencies of wind directions and true-average wind speeds

Wind toward ^a	Frequency	Wind speeds for each stability class (m/s)						
		A	B	C	D	E	F	G
1	0.040	2.58	2.58	2.58	2.09	1.59	1.59	0.0
2	0.011	1.85	1.85	1.85	1.55	1.25	1.25	0.0
3	0.016	1.73	1.73	1.73	1.53	1.33	1.33	0.0
4	0.011	1.52	1.52	1.52	1.37	1.22	1.22	0.0
5	0.048	2.29	2.29	2.29	1.86	1.43	1.43	0.0
6	0.117	2.99	2.99	2.99	2.39	1.79	1.79	0.0
7	0.164	3.12	3.12	3.12	2.47	1.81	1.81	0.0
8	0.044	2.52	2.52	2.52	2.26	2.00	2.00	0.0
9	0.024	2.57	2.57	2.57	2.29	2.01	2.01	0.0
10	0.020	2.50	2.50	2.50	2.05	1.59	1.59	0.0
11	0.039	2.94	2.94	2.94	2.40	1.87	1.87	0.0
12	0.053	3.52	3.52	3.52	2.91	2.29	2.29	0.0
13	0.082	3.24	3.24	3.24	2.66	2.08	2.08	0.0
14	0.102	3.54	3.54	3.54	2.68	1.82	1.82	0.0
15	0.150	3.39	3.39	3.39	2.58	1.78	1.78	0.0
16	0.088	3.54	3.54	3.54	2.65	1.76	1.76	0.0

^aWind directions are numbered counterclockwise starting at 1 for due north.

Table D.6; they give, when multiplied by the source term (in Ci/s), the sector-averaged concentrations (Ci/m³) of tritium in ground level air.

Subroutine DOSEN, which evaluates doses in AIRDOS-EPA, treats tritium (and ¹⁴C) as special cases because the stable isotopes of these elements constitute significant fractions of man's body as well as of his food and drink.⁶ This subroutine assumes that tritium follows water precisely through the environment and that tritium doses from ingestion of food and water as well as those from inhalation and skin absorption are proportional to the concentration of tritium in the air.⁷

For these dose estimates, it is assumed^{6,7} that a person's daily intake consists of 0.532 kg of vegetables, 0.258 kg of meat, and 0.307 kg of milk; his total intake of water in this food is assumed to be 1638 g/d. The total body dose conversion factor for ingestion is taken to be 8.3×10^{-5} rem/ μ Ci.⁷ The absolute humidity is assumed to average 8.0 g/m³ of air, although Till et al.⁸ suggest that 8.4 g/m³ may be more realistic for the Oak Ridge, Tennessee, area. If all of the individual's food is in equilibrium with atmospheric tritium at his location, he would (by ingestion of that food) receive 6.18×10^{-6} rem/year (6.18×10^{-6} rem/year (6.18×10^{-3} millirem/year) for each picocurie of tritium contained in a cubic meter of air.

The individual is also assumed to ingest 1512 g/d of drinking water. The assumption that this water has the same specific activity as that of water in the air is generally considered to be markedly ultraconservative,⁸ and the DOSEN subroutine has used, for these calculations,

Table D.6. Ground-level χ/Q values for tritium at various distances in each compass direction – 100 m stack height
(χ/Q in s/m^3)

Distance		N	NNW	NW	WNW	W	WSW	SW	SSW
Miles	(km)								
0.75	1.2	0.524×10^{-7}	0.148×10^{-7}	0.220×10^{-7}	0.150×10^{-7}	0.650×10^{-7}	0.148×10^{-6}	0.211×10^{-6}	0.573×10^{-7}
1.5	2.4	0.310×10^{-7}	0.849×10^{-8}	0.128×10^{-7}	0.876×10^{-8}	0.379×10^{-7}	0.911×10^{-7}	0.131×10^{-6}	0.350×10^{-7}
2.5	4.0	0.220×10^{-7}	0.580×10^{-8}	0.876×10^{-8}	0.589×10^{-8}	0.266×10^{-7}	0.669×10^{-7}	0.976×10^{-7}	0.253×10^{-7}
3.5	5.6	0.172×10^{-7}	0.454×10^{-8}	0.665×10^{-8}	0.461×10^{-8}	0.208×10^{-7}	0.530×10^{-7}	0.777×10^{-7}	0.199×10^{-7}
4.5	7.2	0.141×10^{-7}	0.375×10^{-8}	0.567×10^{-8}	0.383×10^{-8}	0.172×10^{-7}	0.439×10^{-7}	0.646×10^{-7}	0.164×10^{-7}
7.5	12.1	0.902×10^{-8}	0.245×10^{-8}	0.371×10^{-8}	0.253×10^{-8}	0.111×10^{-7}	0.281×10^{-7}	0.414×10^{-7}	0.104×10^{-7}
15	24.1	0.406×10^{-8}	0.112×10^{-8}	0.169×10^{-8}	0.117×10^{-8}	0.500×10^{-8}	0.126×10^{-7}	0.186×10^{-7}	0.468×10^{-8}
25	40.2	0.220×10^{-8}	0.606×10^{-9}	0.916×10^{-9}	0.632×10^{-9}	0.271×10^{-8}	0.685×10^{-8}	0.101×10^{-7}	0.254×10^{-8}
35	56.3	0.143×10^{-8}	0.393×10^{-9}	0.593×10^{-9}	0.408×10^{-9}	0.175×10^{-8}	0.445×10^{-8}	0.654×10^{-8}	0.165×10^{-8}
45	72.4	0.103×10^{-8}	0.281×10^{-9}	0.424×10^{-9}	0.291×10^{-9}	0.126×10^{-8}	0.320×10^{-8}	0.468×10^{-8}	0.119×10^{-8}
		S	SSE	SE	ESE	E	ENE	NE	NNE
0.75	1.2	0.309×10^{-7}	0.259×10^{-7}	0.499×10^{-7}	0.631×10^{-7}	0.103×10^{-6}	0.121×10^{-6}	0.190×10^{-6}	0.107×10^{-6}
1.5	2.4	0.187×10^{-7}	0.152×10^{-7}	0.301×10^{-7}	0.393×10^{-7}	0.636×10^{-7}	0.752×10^{-7}	0.118×10^{-6}	0.658×10^{-7}
2.5	4.0	0.134×10^{-7}	0.108×10^{-7}	0.215×10^{-7}	0.287×10^{-7}	0.464×10^{-7}	0.550×10^{-7}	0.865×10^{-7}	0.479×10^{-7}
3.5	5.6	0.104×10^{-7}	0.840×10^{-8}	0.168×10^{-7}	0.224×10^{-7}	0.364×10^{-7}	0.433×10^{-7}	0.683×10^{-7}	0.376×10^{-7}
4.5	7.2	0.855×10^{-8}	0.691×10^{-8}	0.138×10^{-7}	0.183×10^{-7}	0.299×10^{-7}	0.357×10^{-7}	0.565×10^{-7}	0.310×10^{-7}
7.5	12.1	0.539×10^{-8}	0.441×10^{-8}	0.869×10^{-8}	0.114×10^{-7}	0.188×10^{-7}	0.227×10^{-7}	0.360×10^{-7}	0.197×10^{-7}
15	24.1	0.242×10^{-8}	0.199×10^{-8}	0.389×10^{-8}	0.512×10^{-8}	0.842×10^{-8}	0.102×10^{-7}	0.162×10^{-7}	0.883×10^{-8}
25	40.2	0.132×10^{-8}	0.108×10^{-8}	0.211×10^{-8}	0.279×10^{-8}	0.458×10^{-8}	0.554×10^{-8}	0.876×10^{-8}	0.480×10^{-8}
35	56.3	0.859×10^{-9}	0.702×10^{-9}	0.138×10^{-8}	0.183×10^{-8}	0.299×10^{-8}	0.362×10^{-8}	0.570×10^{-8}	0.313×10^{-8}
45	72.4	0.621×10^{-9}	0.505×10^{-9}	0.997×10^{-9}	0.133×10^{-8}	0.216×10^{-8}	0.261×10^{-8}	0.409×10^{-8}	0.226×10^{-8}

the assumption that drinking water has a specific activity that is 1% of that in the atmospheric moisture at the individual's location. If so, ingestion of 1512 g/d of drinking water is responsible for 5.70×10^{-8} rem/year (5.70×10^{-5} millirem/year) for each picocurie of tritium contained in a cubic meter of air.

The individual is assumed to breathe $22.0 \text{ m}^3/\text{d}$ of air, and this air is, of course, assumed to be at the tritium concentration peculiar to his location. Tritiated water absorbed through the skin is assumed to be 50% of that inhaled. Allowance for tritiated water absorbed through his skin is made by using as the dose conversion factor for inhalation 1.25×10^{-4} rem/ μCi inhaled (1.5 times the 8.3×10^{-5} rem/ μCi ingested). An individual breathing air at 1 picocurie of tritium per cubic meter receives an annual dose of 1.004×10^{-3} millirem via inhalation and skin absorption.

An individual exposed to air containing tritium at 1 pCi/ m^3 (0.125 pCi/g of water in air) would, accordingly, receive an annual dose of

$$6.18 \times 10^{-3} + 5.7 \times 10^{-5} + 1.004 \times 10^{-3} = 7.24 \times 10^{-3} \text{ millirem/year}$$

if his food were all produced (and in equilibrium) at that location and if his drinking water were at 1% of the specific activity of the atmospheric moisture.

The population distribution expected within 50 miles of the Oak Ridge site in the year 2020 has been estimated⁹ to be that shown in Tables D.7 and D.8. The combination of the plant exclusion area and the large Oak Ridge reservation would seem to ensure that no

Table D.7. Population distribution within ten miles of site in year 2020

Sector ^a	Miles from site					
	0-1	1-2	2-3	3-4	4-5	5-10
1	0	0	0	25	170	5 370
2	0	0	0	42	260	2 020
3	0	0	0	76	250	610
4	0	0	0	100	180	2 560
5	0	0	0	0	50	1 380
6	0	0	0	0	310	6 280
7	0	0	18	94	410	1 230
8	0	0	31	78	130	700
9	0	0	20	40	140	1 320
10	0	0	21	100	180	5 480
11	0	0	0	18	98	2 640
12	0	0	0	0	18	1 540
13	0	0	0	0	0	1 910
14	0	0	0	0	0	700
15	0	0	0	0	0	41 230
16	0	0	0	390	1 930	7080

^aSectors are numbered counterclockwise starting at 1 for due north.

Table D.8. Population distribution within 50 miles of site in year 2020

Sector ^a	Miles from site					
	0-5	0-10	10-20	20-30	30-40	40-50
1	190	5 370	880	1 020	4 970	4 730
2	310	2 020	2 260	740	8 210	4 870
3	330	610	3 740	3 610	1 430	7 920
4	280	2 560	4 070	620	1 150	5 010
5	50	1 380	16 600	2 160	21 500	5 060
6	310	6 280	16 900	4 400	6 700	2 480
7	510	1 230	3 310	2 540	4 600	16 500
8	240	700	1 790	8 900	46 200	18 600
9	200	1 320	12 600	12 100	8 890	4 040
10	300	5 480	12 000	7 310	1 080	320
11	120	2 640	6 130	18 000	850	1 150
12	18	1 540	9 030	82 800	8 410	12 900
13	0	1 910	57 700	246 900	18 900	29 800
14	0	700	26 900	97 700	28 000	23 500
15	0	41 200	30 100	9 610	8 470	8 810
16	2 320	7 080	3 580	15 300	20 400	10 900

^aSectors are numbered counterclockwise starting at 1 for due north.

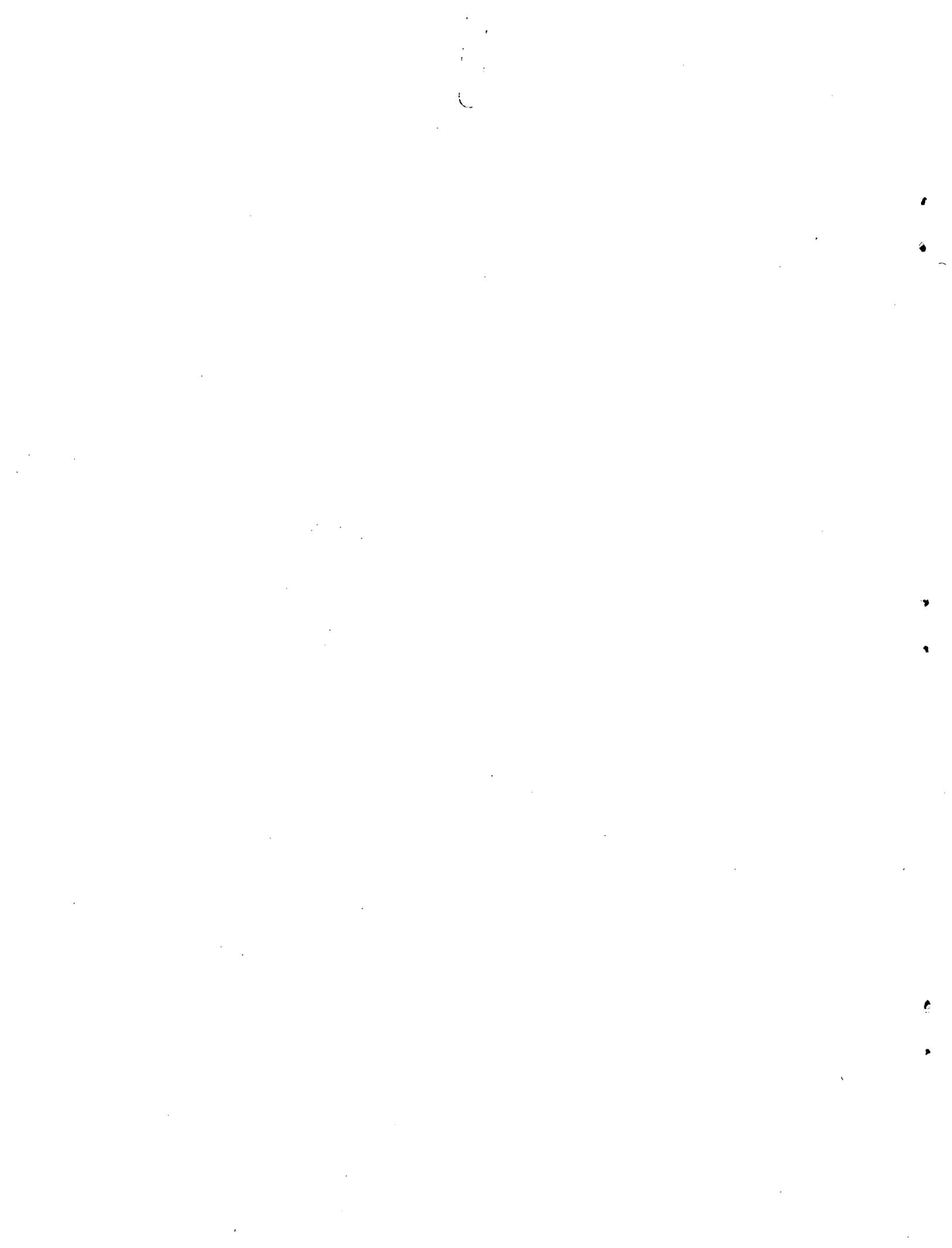
individuals live very close to the plant. A total of 1 155 000 people are expected to occupy the area in year 2020. That total and the distribution, shown by sectors in Tables D.7 and D.8, have been used in the pertinent dose estimates.

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