MLM-2262

MLM-2262

Monsanto/Mound Laboratory Tritium Waste Control Technology Development Program

John C. Bixel, Carl J. Kershner and T. Ben Rhinehammer

December 30, 1975

Monsanto

MOUND LABORATORY
Miamisburg, Ohio
operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company
for the
UNITED STATES ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
U. S. Government Contract No. E-33-1-GEN-53

MLM-2262
TID-4500
Distribution Category UC-70

Monsanto/Mound Laboratory Tritium Waste Control Technology Development Program*

John C. Bixel, Carl J. Kershner, T. Ben Rhinehammer

Issued: December 30, 1975

*Presented at the Third ERDA Environmental Protection Conference, September 23-26, 1975, Chicago, Illinois

PRINTED IN THE UNITED STATES OF AMERICA

Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161
Price: Printed Copy $4.00; Microfiche $2.25

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

MONSANTO RESEARCH CORPORATION
A Subsidiary of Monsanto Company

MOUND LABORATORY
Miamisburg, Ohio 45342
operated for

UNITED STATES ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

U. S. Government Contract No. E-33-1-GEN-53

Table of Contents

Abstract

Over the past four years, implementation of tritium waste control programs has resulted in a 30-fold reduction in the gaseous tritium effluents from Mound Laboratory. However, to reduce tritium waste levels to the "as low as practicable" guideline poses problems that are beyond ready solution with state-of-the-art tritium control technology.

To meet this advanced technology need, a tritium waste control technology program was initiated. Although the initial thrust of the work under this program was oriented toward development of gaseous effluent treatment systems, its natural evolution has been toward the liquid waste problem. We contend that, of all the possible approaches to disposal of tritiated liquid wastes, recovery offers the greatest advantages. End products of the recovery processes would be: 1) water detritiated to a level below the Radioactivity Concentration Guide (RCG) or detritiated to a level that would permit safe recycle in a closed loop operation and, 2) enriched tritium. The detritiated water effluent could be either recycled in a closed loop operation such as in a fuel reprocessing plant or safely released to the biosphere, and the recovered tritium could be recycled for use in fusion reactor studies or other applications.

Introduction and Background

A December 1970 Commission communique asked that contractors seek to limit their tritium and other radioactive effluents to levels that are "as low as practicable." Subsequent communiques suggest control of radiological release to less than 10% of the Radioactivity Concentration Guide (RCG) for uncontrolled areas and also suggest moving the point of concentration measurement from plant boundary limits to within the effluent stacks.

Beginning in 1970, an intensive tritium emission control effort was put into effect at Mound Laboratory. This effort has as its goals an ultimate objective of approaching zero emissions and an engineering objective quantified in terms of maintaining stack emission levels at or below 10% of the present RCG values (40 $\mu Ci/m^3$ for HT and 0.2 $\mu Ci/m^3$ for HTO). To accomplish these goals facility design and operating philosophies were revised to those of containment and recycle as opposed to the past practice of high dilution and release. Over the past 4 yr, implementation of this philosophy through facility modifications and additions and changes in operating procedures has resulted in a 30-fold reduction in the gaseous tritium effluents from the laboratory. However, reduction of tritium effluent levels to 10% of RCG values at the point of emission and nearly complete recycle pose problems that are beyond ready solution with state-of-the-art tritium control technology.

To meet this advanced technology need, a tritium effluent control project was initiated in January 1972. The experimental direction of this project was predicated on the results of an initial source and facility evaluation which revealed that as much as 80% of the total annual release to the atmosphere could be attributed to "background" diffusion from gloveboxes and other containments to the room ventilation systems. To treat the voluminous quantity of high humidity room air was deemed to be both economically and technically impractical. Therefore, emphasis has been placed on confining the tritium at the source through use of glovebox atmosphere detritiation and recovery systems and applying room air treatment only for emergency conditions in the event of accidental releases. A test laboratory, embodying many of the results of the past research phase of the work, has been designed and its construction has been completed.[1]

As the program has matured, scope of the development effort has expanded to include liquid and solid as well as gaseous tritium wastes. Over the past several years it has become increasingly evident that the currently acceptable practices for disposing of tritiated liquid waste will not

be adequate for the future because of the expected increases in quantities and the growing public concern with radioisotopic releases to the environment.

At present, the primary sources of tritiated liquid wastes are the ERDA contractors. Because of increased emphasis on the control of effluents, glovebox detritiation and other effluent removal systems have been and are being installed which result in increased production of high level waste (>1000 Ci/liter). Although modest increases are expected in the high level category when Los Alamos and Sandia Livermore go on-stream with their new effluent removal and glovebox atmosphere detritiation systems, the intermediate level liquid wastes (between 1000-0.01 Ci/liter) from fuel reprocessing plant operations are expected to dominate by 1985 (Tables 1 and 2). Moreover, in the period from 1985 to the year 2000 the quantities of tritium being produced from the reprocessing of fission reactor fuels are expected to more than triple. Added to this will be a yet unknown quantity of tritiated liquid wastes generated by fusion experiments and reactors which could significantly contribute to the quantities to be disposed of in the latter part of this century.

It is evident that now is the time to initiate development of disposal methods which can meet the needs of the ERDA contractors as well as the needs of the nuclear power industry in the 1980's and 1990's and which can be economical and acceptable to the public. There are a number of possible approaches for the disposal of tritiated liquid wastes. The characteristics of most of the approaches together with processing cost estimates are given in Table 3.

Evaluation of Advantages and Disadvantages

It can be seen from the information in Table 3 that in general the least costly disposal methods such as surface water discharge, atmospheric discharge, and deep-well injection, all have in common a high vulnerability to changing environmental and political constraints which could render them unacceptable. Moreover the tritium disposed of by any of these methods cannot be retrieved and represents an irrevocable environmental commitment. Deep ocean release is the next most attractive from a disposal cost basis. However, the technology would require development, and biological data are not available for the effects on sea life. Ocean release is also subject, although to a lesser degree, to the same legal and regulatory constraints as are the other release methods. Although tank storage is one of the lower-cost methods, it suffers from long-term maintenance and monitoring requirements, and it represents a potential local-exposure risk from leakage.

Land burial, either vessel containment or chemical fixation, offers greater protection against environmental release than any of the other disposal methods except recovery. However, both vessel containment and chemical fixation are quite costly for large volumes because of processing, packaging, and transportation costs. Burial grounds also require monitoring, and their operation and siting are vulnerable to changing legal and regulatory constraints. Recovery offers an even lower environmental risk than the burial methods at an estimated cost comparable to some of the release methods. Moreover, tritium will be a valuable fuel resource in the initial phases of the fusion power economy, and thus the value of the recovered tritium will either totally or partially defray the cost of the recovery operation.

Selection of the Best Alternate

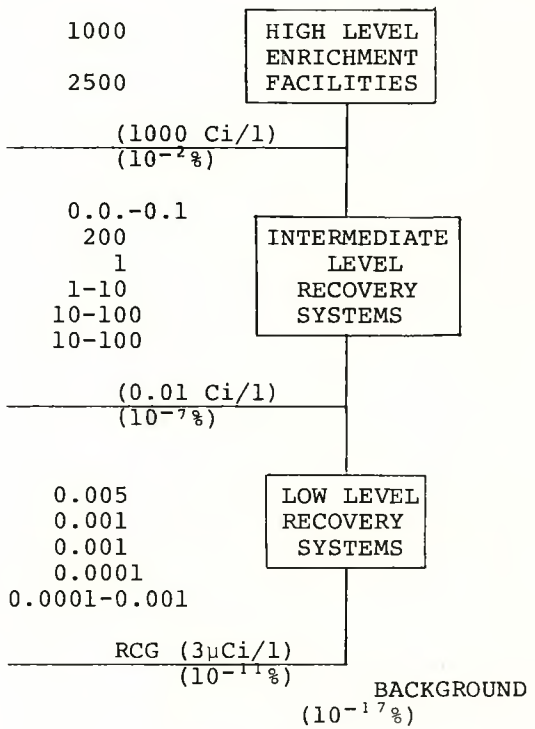
We contend that of all the possible approaches, recovery offers the greatest advantages for the high-level and intermediate-level categories.

Recent developments have altered the situation with respect to the economics and availability of recovery processes. Specifically, the tritium waste control project work at Mound Laboratory over the past 3 yr has led to the development of detritiation and recovery processes that can be directly applied to the tritiated liquid waste recovery problem. Although the initial thrust of this work was oriented toward development of gaseous effluent treatment systems, its natural evolution has been toward the liquid waste recovery problem, and the major development effort is currently in this area.

Table 1

SOURCE, QUANTITIES, AND CONCENTRATIONS OF TRITIATED WATER WASTES PROJECTED TO 1985

Tritium Sources	Costs of Biological Damage[c] ($K)	Tritium (g/yr) (1975)	Tritium (g/yr) 1985	Quantity of Water (liter/yr)	Average Concentration (Ci/liter)
Mound Laboratory	100		100	1000	1000
Other ERDA Contractors	25		25	100	2500
Fuel Reprocessing Plants[d]	100	(0)	100[a]	8×10^7	0.0.-0.1
	100	(0)	100[b]	3×10^3	200
Industry	2	(1)	2	2×10^4	1
ERDA Contractors	10	(2)	10	2×10^4	1-10
CTR	5	(0)	1-10	1×10^2	10-100
HTCR	3	(0)	1-6	600	10-100
Pressurized Water Reactors	7	(1)	7	2×10^7	0.005
LMFBR	1	(0)	0-2	1×10^7	0.001
CTR	5	(0)	1-10	5×10^7	0.001
Industry	2	(1)	1-3	2×10^8	0.0001
ERDA Contractors	5	(5)	5	1×10^8	0.0001-0.001



[a]Acid dissolution plants
[b]Voloxidation plants
[c]Based on Cohen-Higgins estimated cost of $0.10/Ci for biological damage[2]
[d]J. O. Blomake, C. W. Kee, and R. Salmon, "Shipment in the Nuclear Fuel Cycle Projected to the Year 2000," Nuclear News, 18:8, June 1975, pp. 62-65.

Table 2

TRITIUM GENERATION SUMMARY FROM FUEL REPROCESSING PLANTS

Year	Nuclear Generating Capacity (GWe)	No. of Reactors	No.of LWR's	No. of HTGR's	No. of LMFBR's	Fuel Reprocessing Loads	Tritium Generation (g/yr)	Accumulated Tritium (g)	Volume of Water (liter/yr) Acid-Leach	Volume of Water (liter/yr) Voloxidation[f]
1975	35[a]	51[a]	51[a]	0	0	None				
1980	78[b] 85[c]	95[b]	94[b]	1[b]	0	2180[d]	130[d]	300[d]	8×10^7[g] 19×10^7[g]	
1985	180[b] 231[c]	186[b]	182[b]	4[b]	1[i]	4110[d]	200[d]	890[d]	8×10^7[g] 19×10^7[h]	5×10^3
1990	410[c]	395[e]	363[e]	31[e]	4[e]	7810[d]	440[d]	2200[d]	8×10^7[g] 19×10^7[h]	1.7×10^4
2000	850[c]	795[e]	671[e]	85[e]	38[e]	16680[d]	1000[d]	7500[d]		5×10^4

[a] In operation 8/1/75, Nuclear News, 18:10, August 1975, pp. 69-74.

[b] Nuclear News, 18:10, August 1975, pp. 69-74.

[c] Nuclear Power Growth 1974-2000, WASH-1139 (74), Office of Planning and Analysis, Superintendent of Documents, Government Printing Office, Washington, D. C., February 1974.

[d] J. O. Blomeke, C. W. Kee and R. Salmon, "Shipment in the Nuclear Fuel Cycle Projected to the Year 2000," Nuclear News, 18:8, June 1975, pp. 62-65.

[e] Calculations based on ref (b) and (c) assuming new reactors will average 1.1 GWe each.

[f] Calculations based on tritium concentration of 200 Ci/liter for voloxidation-type reprocessing plants as assumed in ref (d).

[g] Calculations based on acid-leaching plants (AGNS and NFS) in operation at combined capacity of 2150 MT/yr.

[h] B. I. Kullen, L. E. Trevorrow and M. J. Steindler, Tritium and Noble Gas Fission Products in the Nuclear Fuel Cycle, Fuel Reprocessing Plants, ANL-8135, Argonne National Laboratory, Argonne, IL, March 1975, p. 21.

[i] Clinch River Breeder Reactor Development.

Table 3

METHODS FOR TRITIUM WASTE DISPOSAL

Method	Costs/Gallon ($) Pack. & Dis.	Trans.	Advantages	Disadvantages
Storage (tank)	0.10-0.20[a]	$0.01-0.04	Relatively moderate cost. Low transportation cost. Retrievable. Simple monitoring required. Minimal pretreatment required.	High risk atmospheric release. Long-term maintenance required. Subject to physical damage. Subject to changing environmental and political constraints.
Surface Water Discharge	0.01[a]	0	Lowest cost of all methods. No transportation costs. Easily monitored.	Irrevocable release to environment. Some pretreatment of waste required. Requires large quantities of water for dilution. Not universal method of disposal - only selected sites qualify. Subject to changing political and environmental regulations.
Atmospheric Discharge	0.01-0.04[a]	0	Very low cost. No transportation cost. Easily monitored. Minimal pretreatment required. Energy for evaporation usually readily available at minimal cost.	Not amenable to populated area. Subject to shutdown because of weather conditions. Subject to great environmental and political pressures. Irrevocable release to environment.
Deep Well Injection	0.01-0.10[a,b]	0.04-0.20	Cheapest next to discharge to atmosphere or surface water. Extensive related technology available. Minimal pretreatment of liquid necessary. Moderate transportation costs.	Irrevocable release to environment. Extensive monitoring required. Increasing legal and regulatory constraints. Industrial trend to terminate this type of disposal for chemicals. Requires area having specific geohydrological characteristics.
Land Burial (vessel containment)	20[c]	0.40	Extensive technology available. Pretreatment unnecessary. Can be made retrievable. Excellent for low volume waste. Locations more readily available.	Moderate monitoring required. High transportation costs. Costly for large volume waste. Increasing legal and regulatory restraints. Requires specific geological properties for burial ground. Extremely high costs if made retrievable.
Land Burial (chemical fixation)	3[d]	0.40	Low environmental release risk. Not vulnerable to physical damage. Can be made retrievable.	High cost method. Requires moderate monitoring. Difficult to recover unless specifically provided for. Special facilities required for fixation. High transportation cost. Requires specific geological properties for burial ground.

Table 3 (continued)

Method	Costs/Gallon ($) Pack. & Dis.	Trans.	Advantages	Disadvantages
Deep Ocean	0.10-0.50	0.04-0.20	Fewer environmental pressures. Moderate transportation costs. Stable disposal - no mixing when disposed at 30,000 ft. Nearly infinite 'sink.' Little monitoring required	Geopolitical ramifications may be difficult to solve. Technology needs development. Biological data for effects on sea life lacking. Irrevocable environmental release. Subject to transportation and transfer hazards. Surface disposal equivalent to surface water release on land.
Recovery	0.20[e]	$0.05-0.10	Lowest environment release risk. Moderate transportation costs. Conservation of valuable resource. No large disposal site required. Partial or total cost recovery from product utilization. High public acceptance.	High development costs. High facility costs. Greater occupational exposure risks.

[a] W. D. Arnold, R. Salmon, K. H. Lin and W. deLaguna, Preliminary Evaluation of Methods for the Disposal of Tritiated Water from Nuclear Stimulated Natural Gas Wells, ORNL-TM-4024, Oak Ridge National Laboratory, Oak Ridge, TN, April 1973.

[b] M. J. Steindler, et al., Chemical Engineering Division Waste Management Programs Quarterly Report: April-June 1974, ANL-8134, Argonne National Laboratory (November 1974), 54 pp.

[c] Mound Laboratory current 3-drum asphalt-covered burial package plus labor to package and burial in approved land burial facility.

[d] Includes only cost to produce polymer impregnated tritiated concrete, P. Columbo, M. Steinberg, and B. Manowitz, Tritium Storage Development: July-September 1974, BNL-19408, Brookhaven National Laboratory, October 1974, 18 pp.

[e] Cost calculated from data in this report based on annual 1985 intermediate-level quantities and 10-yr average recovery cost of $5,000/g excluding credit for value of tritium recovered.

Long Range Plan

The past accomplishments, the current effort, and the long range plan for tritium waste control technology development at Mound Laboratory are illustrated in Figure 1. The work has progressed from research and development toward implementation of water detritiation and tritium recovery on a production basis.

The present effort and its 8-10 yr projection are to develop and apply new processes for the detritiation of contaminated water from light water reactors, fuel reprocessing plants, and tritium handling facilities. The program emphasizes recycle of such waste water rather than disposal by burial or other means. End products of the processes will be: 1) water effluent detritiated to a level below RCG or to a suitable level for safe use in a closed loop process, and 2) enriched tritium. It is expected that the recovered tritium could be recycled for use in fusion reactor studies or other applications, thus long-term storage or disposal is not required.

A five-phase program is in progress to develop processes and to construct a facility to recover and process all contamination levels of tritiated liquid wastes. Modest recovery capabilities are expected to be realized as early as 1977 for high-level material and significant intermediate-level recovery capabilities are expected to be developed by 1980. The details of the various phases of this program are as follows:

PHASE I - HIGH LEVEL RECOVERY (HLR) DEVELOPMENT

Prototype systems are being built and tested to prove the concepts of recovery of tritium from and decontamination of high-level tritiated water wastes. This development work is based on cryogenic distillation, palladium column, and electrolysis technologies. After development tests are completed, the prototype high-level recovery system will be available for interim recovery operations. Data obtained in the test program are being used to estimate costs and forecast cost benefits of the process as well as for scale-up design of larger systems.

The Phase I program also includes studies on HTO waste generation quantities, waste shipping container development, and waste packaging development.

PHASE II - HLR OPERATION

A full-scale high-level recovery system will be designed, built, installed, tested, and placed in operation. This facility will be designed with capacity to process the quantities of high-level waste expected to the year 1985, as well as high-level waste quantities generated by future operations which will upgrade intermediate-level to high-level waste. During the design and construction stage of the full-scale system, capacity of the prototype high-level system will be increased by a factor of 10 to provide additional interim recovery capability. Figure 2 schematically illustrates the high-level recovery system as it is envisioned at this time.

PHASE III - RESEARCH AND DEVELOPMENT TOWARD INTERMEDIATE-LEVEL AND LOW-LEVEL RECOVERY (ILR AND LLR)

An advanced research and development effort on enrichment processes, such as H_2/H_2O catalytic exchange and selective photoexcitation (Figures 3 and 4, respectively), is directed toward solving the more difficult problems of tritium recovery from intermediate-level and low-level processes, as well as for scale-up design of larger systems. In addition, studies will be made to optimize recovery facility location with respect to waste water source, shipping costs, and other economic and environmental considerations. Finally the design of an intermediate-level tritiated waste water recovery facility will be initiated.

PHASE IV - ILR OPERATION

A full-scale, intermediate-level tritiated waste water facility or facilities would be designed, built, tested, and placed in operation to provide a tritium recovery capability for greater than 200 g/yr.

PHASE V - LLR OPERATION

A full-scale, low-level, tritiated waste water facility or facilities would be designed, built, tested, and placed in operation to provide a recovery capability for the low-level waste quantities expected in 1985.

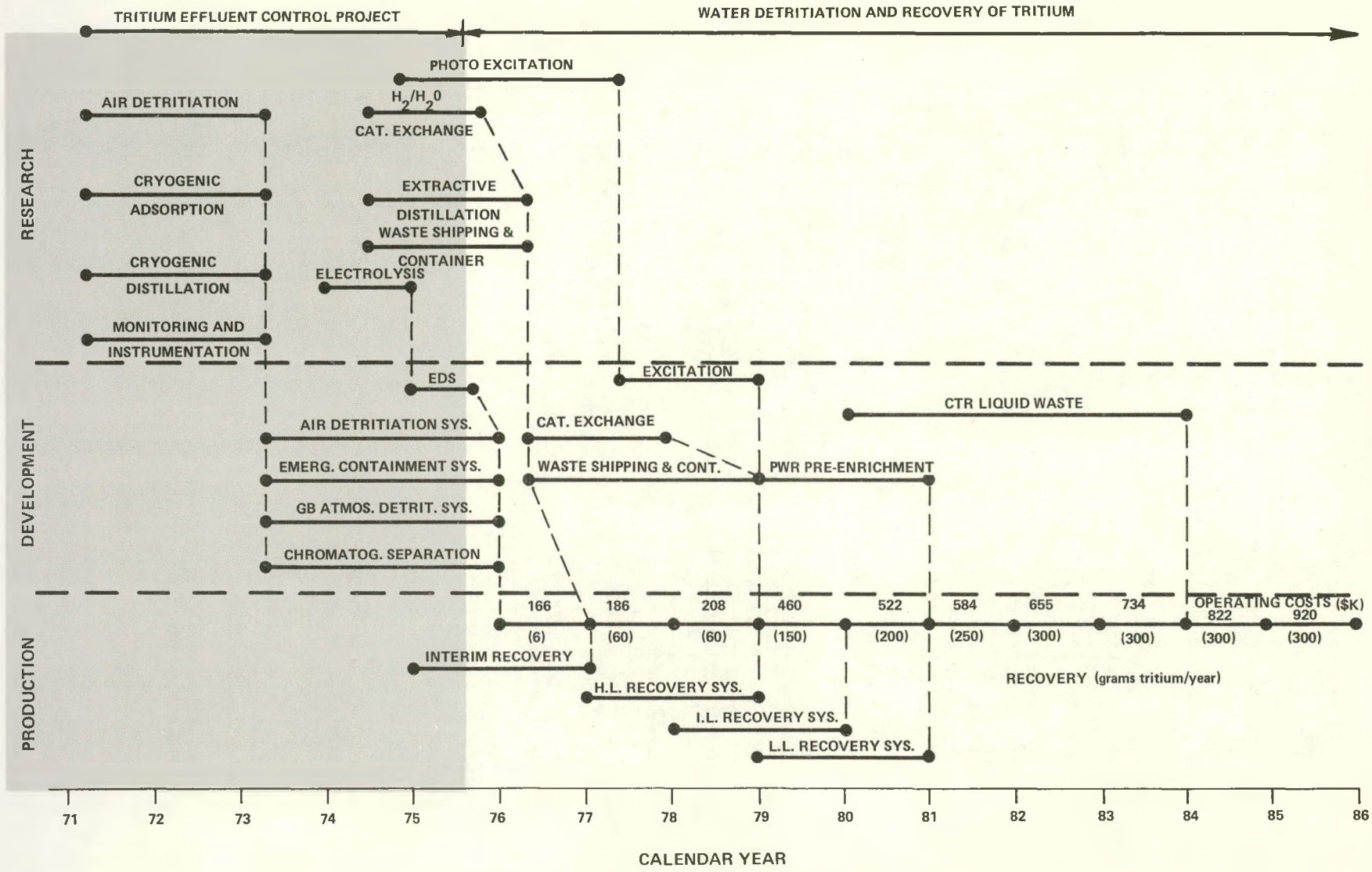


FIGURE 1 - Tritium effluent and recovery control plan.

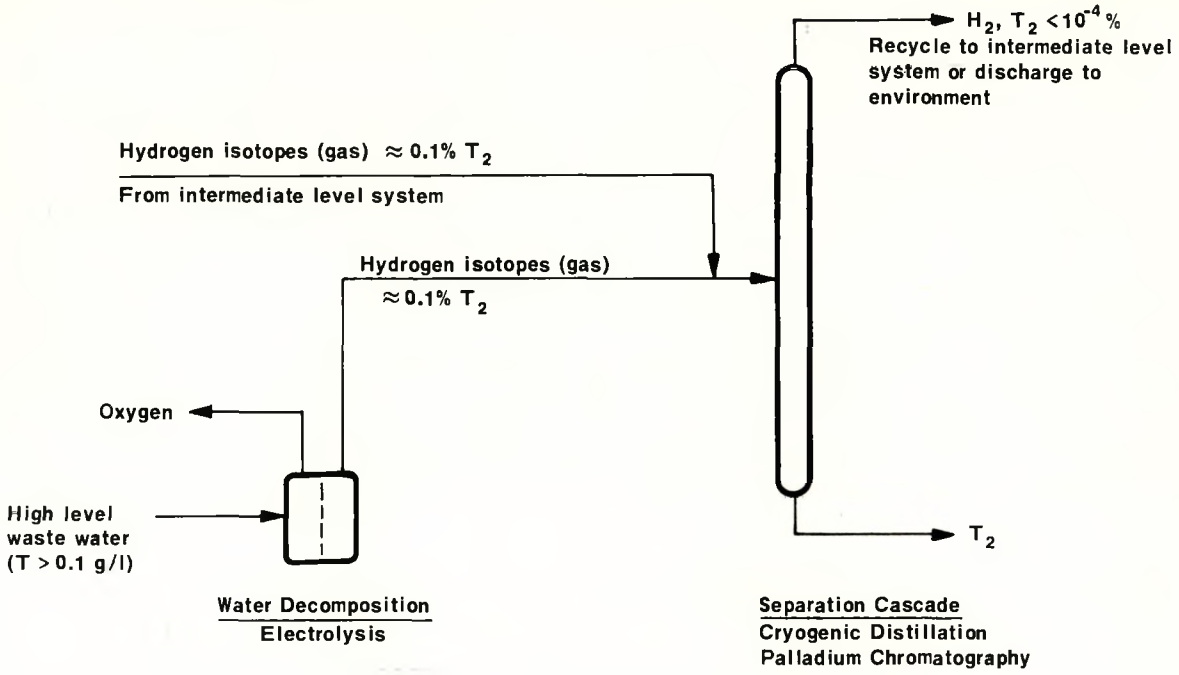


FIGURE 2 - High level recovery system schematic.

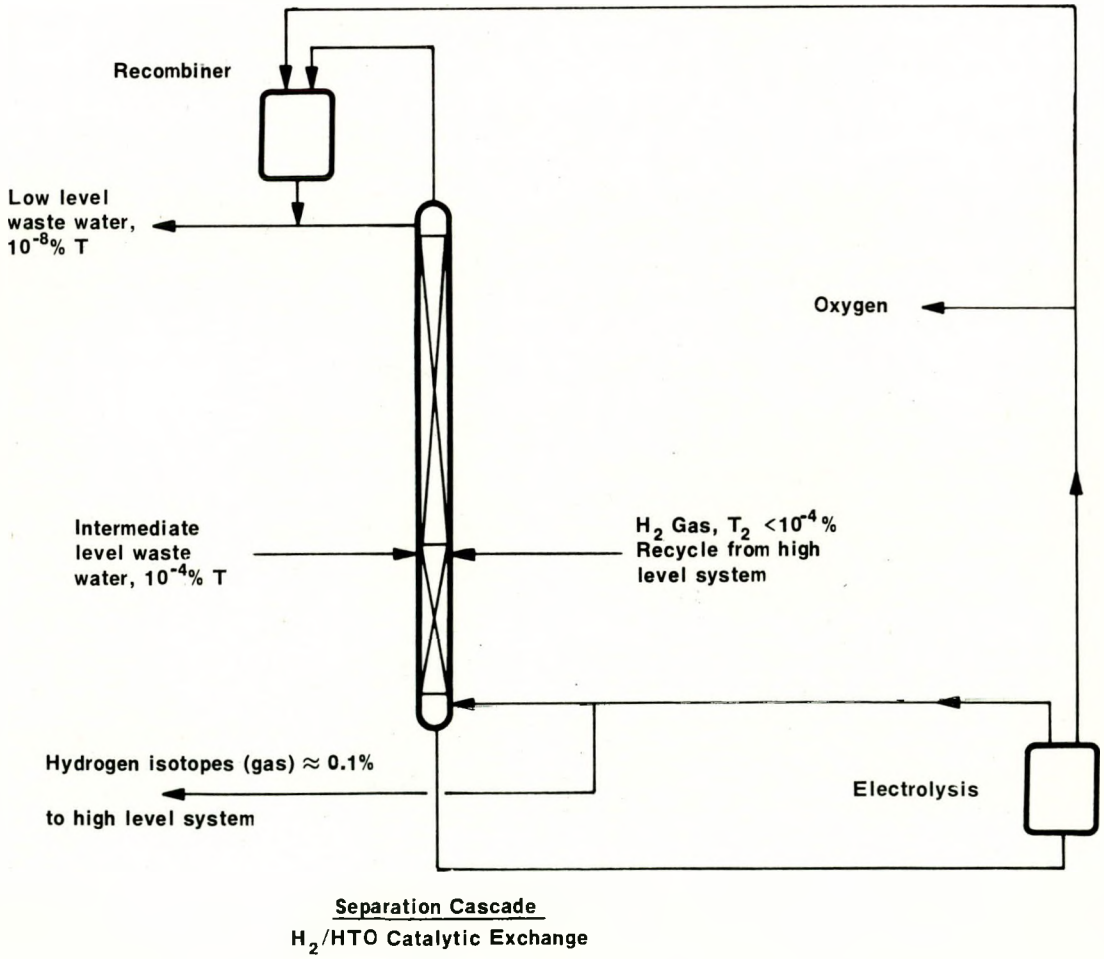


FIGURE 3 - intermediate level recovery system schematic.

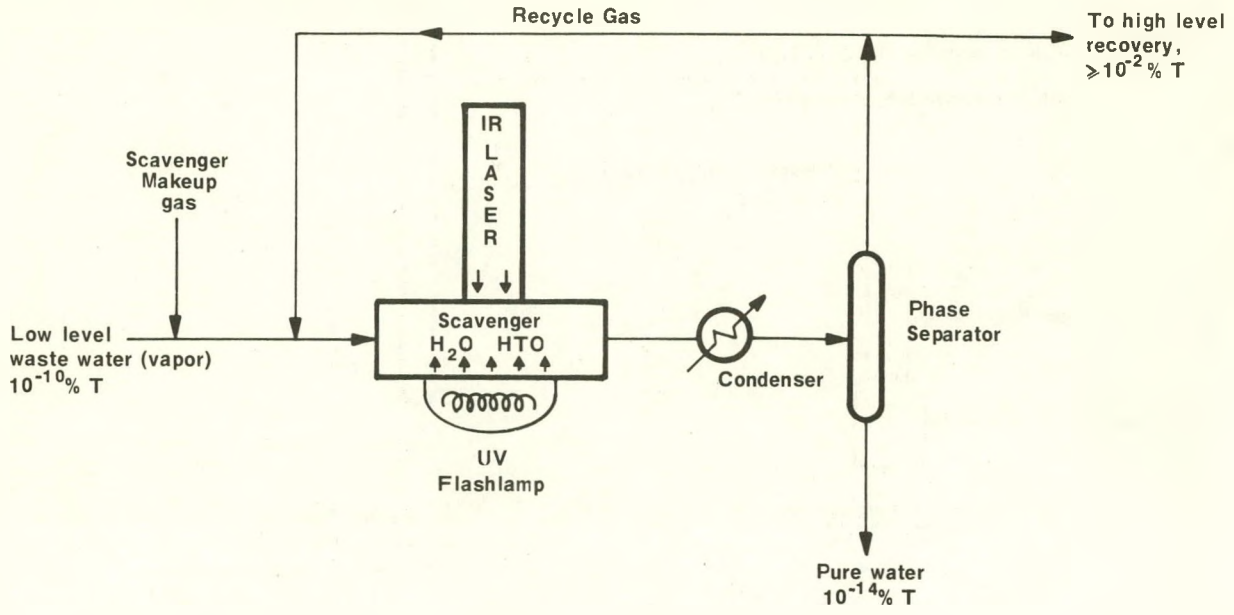


FIGURE 4 - Low level recovery system schematic.

Estimated Costs

The total capital cost of the recovery facility at Mound Laboratory is estimated to be $5 million. The annual operating costs are given in Figure 1. If all capital and operating costs are amortized against the total of 1926 g of recovered tritium for the 10-yr period, the average recovery cost is approximately $5,000/g. This value does not take any credit for the savings in disposal or biological damage costs estimated to be $400 and $1000/g of tritium, respectively.[2]

Discussion and Conclusion

Relatively large quantities of high-level and intermediate-level tritiated liquid wastes are currently being buried because, to date, these wastes have been considered economically unrecoverable, processing technology and facilities have not been available, and there has been no large market for the recovered tritium. However, a large market is developing in the CTR program, and by 1985 the inventory and burnup requirements for the EPR-1 (Experimental Power Reactor) alone are expected to exceed by several times the total quantities recoverable from the tritiated wastes shown in Table 1. Therefore, although waste tritium from fission reactors and ERDA sites will not represent a primary source of fuel to the initial nonbreeding fusion DT experiments and test power reactors, it will represent a valuable and useful resource.

As a consequence of the existing waste recovery and packaging facilities and the recovery technology developed over the past several years at Mound Laboratory, an integrated tritium waste recovery facility to serve all of ERDA and the nuclear power industry, as outlined in Figure 5, could be developed and made operable at Mound Laboratory by 1981.

References

1. C. J. Kershner and J. C. Bixel, "Tritium Effluent Control Laboratory," Proceedings of the Thirteenth AEC Air Cleaning Conference, San Francisco, CA, CONF-74087, August 12-15, 1974, pp. 948-970.

2. J. J. Cohen and C. H. Higgins, "The Socioeconomic Impact of Low-Level Tritium Releases to the Environment," in Tritium, A. A. Moghissi and M. W. Carter (ed.), Messenger Graphics, Phoenix, AZ, May 1973, pp. 14-22.

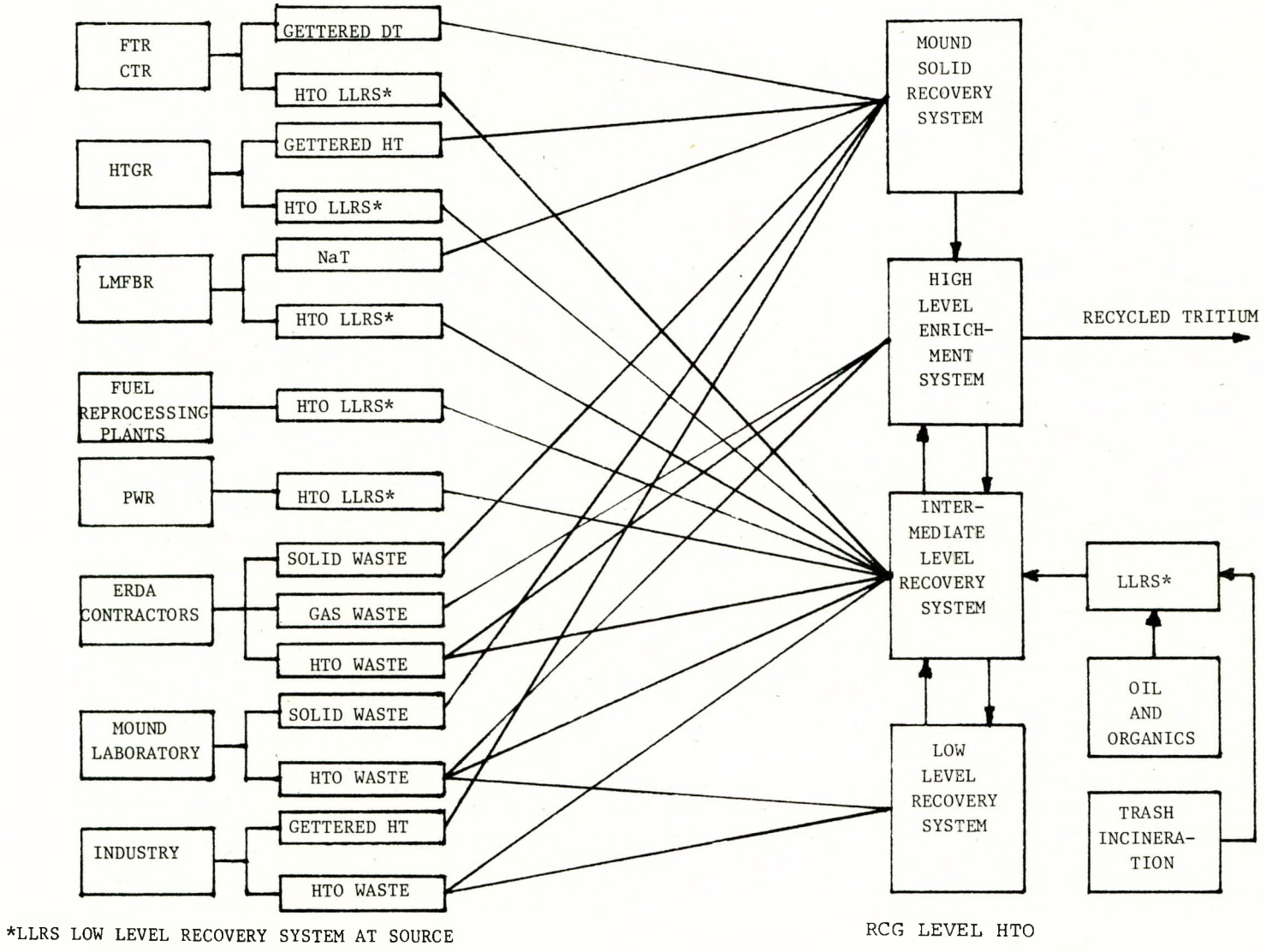


FIGURE 5 - Tritium waste recovery and management for ERDA and the Nuclear Power Industry.

Distribution

EXTERNAL

UC-70 (200)

CONSULTANTS

C. F. Curtiss
University of Wisconsin
C. F. Eck
Miamisburg, Ohio
H. W. Mattson
Monsanto Company
D. F. Griffing
Miami University
R. E. Miers
Ft. Wayne, Indiana
G. W. Powell
Ohio State University
A. Shapiro
University of Cincinnati
H. F. Swift
University of Dayton Research Institute
D. White
University of Pennsylvania

H. K. Nason, MRC

INTERNAL

J. C. Bixel (2)
W. T. Cave
R. K. Flitcraft
B. J. Hosfeld
L. V. Jones
D. P. Kelly
C. J. Kershner (2)
B. R. Kokenge
T. B. Rhinehammer (2)
H. L. Turner (20)
R. E. Vallee

Publications
Library (15)