CONF-8311109--2

DP-MS-83-114

TRITIUM WASTE DISPOSAL TECHNOLOGY IN THE U.S.

DP-MS--83-114

by

DE84 002620

E. L. Albenesius and O. A. Towler

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, South Carolina 29808

A paper proposed for presentation at the
Joint U. S./French Meeting on Waste Management
Knoxville, Tennessee
November 9 - 11, 1983
and for publication in the Proceedings of the Meeting

This paper was prepared in connection with work done under Contract No. DE-AC09-76SR00001 with the U. S. Department of Energy. By acceptance of this paper, the publisher and/or recipient acknowledges the U. S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper, along with the right to reproduce and to authorize others to reproduce all or part of the copyrighted paper.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TRITIUM WASTE DISPOSAL TECHNOLOGY IN THE U.S.*

E. L. Albenesius and O. A. Towler
E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, SC 29808

ABSTRACT

Tritium waste disposal methods in the U. S. range from disposal of low specific activity waste along with other low-level waste in shallow land burial facilities, to disposal of kilocurie amounts in specially designed triple containers in 65' deep augered holes located in an arid region of the U. S. Total estimated curies disposed of are 500,000 in commercial burial sites and 10 million curies in defense related sites. At three disposal sites in humid areas, tritium has migrated into the ground water, and at one arid site tritium vapor has been detected emerging from the soil above the disposal area. Leaching tests on tritium containing waste show that tritium in the form of HTO leaches readily from most waste forms, but that leaching rates of tritiated water in polymer impregnated concrete are reduced by as much as a factor of ten. Tests on improved tritium containment are ongoing.

Disposal costs for tritium waste are 7 to 10 dollars per cubic foot for shallow land burial of low specific activity tritium waste, and 10 to 20 dollars per cubic foot for disposal of high specific activity waste. The cost of packaging the high specific activity waste is 150 to 300 dollars per cubic foot.

BACKGROUND

Tritium containing waste has not been treated as a separate waste form. Whereas transuranic waste has long been segregated from other low-level wastes and disposed of in segregated trenches or in retrievable storage areas, tritium containing waste has been mixed with and disposed of along with large amounts of other low-level radioactive waste. Over a period of about 40 years, three methods have been used for low-level radioactive waste disposal: dilution and dispersion, sea disposal, and shallow land burial.⁽¹⁾

Dilution and dispersion of gaseous wastes is still permitted under existing regulations, but with increasing emphasis on making the releases as low as reasonably achievable (the ALARA principle). Sea disposal⁽¹⁾ of radioactive wastes was discontinued in the U. S. in June of 1970. The disposal methods that will be discussed here are the

* The information contained in this article was developed during the course of work under Contract No. DE-AC09-76SR00001 with the U. S. Department of Energy.

shallow land burial of low specific activity tritium wastes (i. e., in trenches), and the disposal of high specific activity tritium waste in triply-contained waste forms in augered holes or in trenches.

A major review of tritium control technology was conducted by Rhinehammer and Lamberger in 1973⁽²⁾, which included reviews of monitoring, handling, containment, packaging, storage and disposal. No major changes have been made in tritium waste disposal technology since that review was made.

TRITIUM WASTE INVENTORY

The activity in the tritium waste disposed of by shallow land burial in the United States over the last 40 years totals about 10.4 million curies.⁽³⁾ Early inventory records are incomplete, and this figure, which is the undecayed value, must be taken only as an approximate value. The waste inventory is small, about 1%, *compared with* of the global inventory of tritium in the hydrosphere and stratosphere of 1000 million curies produced by natural cosmic radiation and past weapons tests⁽⁴⁾.

About 95% of the tritium waste has been disposed of at defense related sites such as the Nevada Test Station and the Savannah River Plant (Table 1), and 5% of the waste has been disposed of at six commercially operated disposal sites (Table 2). The major portion of the tritium waste disposed of at the commercial sites is waste from electric power reactors, research and medical facilities, and manufacturers of luminous materials. Isotopic inventories at commercial sites are not well known; the values in Table 2 must be considered to be only approximate values, based on estimated average isotopic content of the waste and the total waste volume.

DISPOSAL SITES

There are six commercial sites at which some tritium waste is buried, and four defense related sites.⁽⁵⁾ *(Table 3)* The six commercial sites are:

- o West Valley, New York

The disposal site is 25 acres in a total site area of 3345 acres.

The region is humid with a rainfall of about 40 inches per year.

Rainfall exceeds evaporation. The ground water levels are 0-10 feet below ground. The site operated between 1962 and 1975, when the

site was closed due to water management considerations, ⁽⁵⁾ Tritium was found in water collected at onsite monitoring stations, the source of which was water seeping from the interface between the original till and the caps of two trenches. The site is now closed; monitoring continues under state direction.

- o Maxey Flats, Kentucky

The disposal site is on a 300 acre tract of land in a humid area, with an average rainfall of 46 inches. The ground water table is at a depth of 30 to 45 feet. The site operated from 1962 to 1977⁽⁵⁾. Tritium was detected in surface waters and in perimeter ground water wells.

- o Sheffield, Illinois

The disposal site is in a humid area with an average rainfall of 35 inches. The saturated zone occurs at depths of 15 to 60 feet. The site operated from 1967 to 1978.⁽⁶⁾ Tritium migration has not been detected, although other radionuclides have been detected outside the site perimeter.

- o Barnwell, South Carolina

This disposal site is located in a humid area with an average rainfall of about 46 inches. The site has operated as a burial ground since 1971. No tritium migration has been reported. The site is open and receiving waste.

- o Beatty, Nevada and Richland, Washington

These two sites are located in arid regions of the western United States. Annual rainfall is 8 to 10 inches, and evaporation greatly exceeds precipitation. Both sites are open and receiving waste. No tritium migration has been reported.

Of the four defense sites listed in Table 3, two are in arid regions with depths to water table of 800 to 1000 feet. The arid sites have operated since about 1943 -- a period of 40 years. Waste disposal has been in both trenches and augered shafts. At Los Alamos, tritium was found to be moving in the vapor phase from a solid waste storage shaft through open joints in the tuff⁽⁵⁾. The Oak Ridge and Savannah

River sites are both in humid regions in the Eastern part of the United States. Oak Ridge has disposed of waste since 1943 in six burial grounds, totaling 135 acres. The last disposal area began operation in 1973. Tritium disposed of in the fifth burial site has been observed to be migrating to the nearby creek (White Oak Creek).⁽⁵⁾

Tritium waste disposal at the Savannah River site began about 1954. The original disposal area of about 76 acres was filled in 1972; ^(labeled 643-6 in Figure 1) a second contiguous area of about 120 acres is still receiving waste. ^(labeled 644-76 in Figure 1)

Tritium migration has been observed outside the first disposal area^(7,8,9). The tritium plume in the saturated zone has been extensively mapped, ^{as shown by cross section of well drilling in Figure 2,} ~~The results are shown in Figures 2 through 5.~~ There are about 2000 curies in the upper plume, which is about 1200 feet long, 1700 feet wide, and 30 feet deep. Some 200 to 400 curies of tritium are estimated to be in ~~the~~^a lower plume. The tritium plume is still well within the boundary of the Savannah River Plant site. All but a small fraction of the tritium will decay before the plume reaches the nearest stream.

TRITIUM LEACHING TESTS

The Savannah River Laboratory (and others) have performed leaching tests on tritium waste forms, and other waste forms as well. The leaching tests are made to be as realistic as possible. The soil used is the burial ground soil; the test is conducted within the burial ground confines; the precipitation rate is the normal rainfall that occurs in the burial ground area. The test is conducted in a lysimeter -- a hollow tube that contains the soil, the waste form, and a percolate collection system. The lysimeter design is described in Reference 10. Figure ~~6~~² shows the results of tritium leaching from two waste forms^(7,11). The tests showed that over a seven year period essentially all of the tritium will leach from an untreated waste form, but that only 7 to 15% of the tritium will leach from a polymer impregnated tritiated cement waste form. (described in Reference 11).

WASTE PACKAGE DESIGN

While most tritium waste is disposed of with very low tritium concentrations, there are occasions where it is necessary to dispose of large quantities in relatively small packages. Mound Laboratory^(12,13,14) has performed a large amount of work on tritium package design and testing. Their package (Figure ~~7~~³) will hold up to 70,000 curies of tritium in water, oil, or octane. For tritium in water, for example, 35 liters of

water are added to 81 kg of a 3:1 mix of plaster and cement in a 27 gallon (102 liter) polyethylene drum, leaving a 35% void volume. The cover is sealed and placed in an asphalt-coated 30 gallon (114 liter) steel drum. The void spaces are filled with asphalt and vermiculite (an absorbent), and the drum lid is sealed and clamped. This container is placed inside a 55-gallon (208 liter) steel drum, and the void space is again filled with asphalt and vermiculite. The drum cover is sealed and clamped to form the final package.

Mound Laboratory has performed permeation tests on the triply-contained waste, using tritium in water, oil, or octane. The permeation test arrangement is shown in Figure 8. The results are shown for tritium in ~~oil or water~~ ^{oil} in the test arrangement for 8.5 years (Figures 9 and 10), and for tritium in octane in the test arrangement for 11 years (Figure 11). The largest fraction^{al} release in this long term test is 5×10^{-5} after 8.5 years. (The fractional release is total curies leaving the triply contained package divided by the total curies contained initially and decay-corrected.)

COSTS

Costs for packaging low concentration tritium wastes are considered to be negligible -- considering only the cost of a cardboard box or a steel drum. Costs for packaging the triple container developed by Mound Laboratory are estimated to be \$2000 per 55-gallon drum⁽¹⁵⁾, or about \$270 per cubic meter of total waste volume. This cost is high because of the extra care and facilities required to contain the tritium while emplacing it in the primary container and to decontaminate the outer surfaces of the primary and secondary containers.

Disposal costs (Table 4) vary considerably^(16,17), depending on the size of the site -- large or small -- and the methods of estimating future costs. For the most part, unit disposal costs for different types of trenches or augered holes are \$200 to \$300 per cubic meter for a large site, and \$800 to \$1900 per cubic meter for a small site. Commercial disposal sites also add surcharges depending on the radiation level at the surface of the package, the curie content, and the weight of the container. The Barnewell site, for example, adds a surcharge of \$11,100 per cubic meter if the package registers a radiation reading of 1000 R/hour at the surface.⁽³⁾

CONCLUSIONS

In a brief discussion it is realized that one cannot discuss all of the options for tritium waste disposal. . Not discussed here are such tritium management options as immobilization of tritium on molecular sieves, the effect of different coatings on tritiated cement, deep well injection of tritium waste, disposal by Greater Confinement ^{rather} than ~~Shallow Land Burial~~, sea disposal, ^{gas generation} and even the simple concept of storing tritium waste in an engineered structure (for example, a double-walled tank) above ground for a sufficient time to allow for substantial decay. Also not discussed are the effects on waste disposal of proposed regulations and orders governing tritium to be issued by the Department of Energy, the Nuclear Regulatory Commission, and the Environmental Protection Agency.

We can, however, offer conclusions, based on the limited information discussed here, on several important characteristics of tritium waste disposal.

- o Cement alone is not a satisfactory waste form for tritium waste disposal.
- o Tritium in untreated waste forms exchanges rapidly with whatever water is available.
- o The only satisfactory tritium waste form demonstrated so far is the triple package developed by Mound Laboratory. ~~However, it does not provide for zero release.~~
- o Tritium waste disposal in the U. S. in shallow land burial sites has resulted in tritium migration to surface waters, seeps, and to the ground water.

^A ~~The~~ final statement is that while we have learned a lot about what happens to tritium waste, we must in the future expend considerable effort to produce cost effective criteria for better methods of tritium waste disposal.

An excellent summary of the tritium data base and waste management options, recently issued by the Argonne National Laboratory, ⁽¹⁸⁾ reaches the same conclusions listed above.

REFERENCES

1. Radiological Quality of the Environment in the United States, 1977.
U. S. Environmental Protection Agency, EPA 520/1-77-009, (1977).
2. T. B. Rhinehammer and P. H. Lamberger, Tritium Control Technology,
WASH-1269, (1973).
3. Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics. DOE/NE-0017/1. U. S. Department of Energy, Washington, D. C. (1982)
4. Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics. DOE/NE-0017/2. U. S. Department of Energy, Washington, D. C. (1983).
5. D. G. Jacobs, J. S. Epler, R. R. Rose. Identification of Technical Problems Encountered in the Shallow Land Burial of Low-Level Radioactive Wastes. ORNL/SUB-80/13619/1. Oak Ridge National Laboratory, Oak Ridge, Tennessee (1980).
6. The 1980 State-by-State Assessment of Low-Level Radioactive Wastes Shipped to Commercial Disposal Sites. LLWMP-11T. The National Low Level Waste Management Program. EG & G, Idaho (1982).
7. R. H. Hawkins. Migration of Tritium from a Nuclear Waste Burial Site. USDOE Report DP-MS-75-25, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC (1975),
8. J. A. Stone, S. B. Oblath, R. H. Hawkins, R. H. Emslie, J. P. Ryan, C. M. King. Migration Studies at the Savannah River Shallow Land Burial Site. USDOE Report DP-MS-83-89, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC (1983).
9. Technical Summary of Groundwater Quality Protection Program at the Savannah River Plant. Vol. II, Radioactive Waste. DPST-83-929. E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC. (To be issued).
10. R. L. Hooker and R. W. Root. Lysimeter Tests of SRB Waste Forms. DP-1591. E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC (1981).
11. P. R. Columbo, R. Neilson, W. Becker. Tritium Storage Development Program Progress Report No. 9, July - September 1976. BNL-50625. Brookhaven National Laboratory, Upton, NY (1977).

REFERENCES

12. P. H. Lamberger, and M. L. Rogers. Tritium Waste Control: October-March 1983. MLM-3102, Mound Laboratory, Miamisburg, Ohio 45342 (1983). (and other Mound reports -- MLM-3048, MLM-2714, MLM-2844, MLM-2903, and MLM-2969).
13. E. A. Mershad, W. W. Thomasson, J. J. Dauby. Packaging of Tritium Contaminated Waste. Nuclear Science and Technology, Vol. 32, January 1977.
14. E. A. Mershad, W. W. Thomasson, and J. J. Dauby. Tritiated Liquid Waste Packaging. MLM-2440, Mound Laboratory, Miamisburg, Ohio (1977).
15. Personal Communication, R. Blauvelt, Montsanto Mound Facility October 1983.
16. P. T. Dickman and J. R. Boland. Cost Estimates for Greater Confinement Disposal of Radioactive Wastes. Nuclear and Chemical Waste Management, Vol. 4 (1983).
17. T. L. Gilbert and C. Luner. Analysis of Alternatives for Greater Confinement Disposal, Presentation at ANS San Francisco Meeting, November, 1983.
18. L. E. Trevorow, G. F. Vandegrift, V. M. Kolba, and M. J. Steindler. Comparability of Technologies with Regulations in the Waste Management of H-3, I-129, C-14, and Kr-85, Part I. Initial Information Base. ANL-83-57, Part I. Argonne National Laboratory, Argonne, Ill. (1983).

**TRITIUM WASTE INVENTORIES
IN DEFENSE SITES (1982)**

<u>SITE</u>	<u>PERCENT OF TOTAL</u>
LOS ALAMOS	5
IDAHO	0
NEVADA TEST STATION	54
OAK RIDGE	0.05
HANFORD	0
SAVANNAH RIVER	<u>41</u>
	100

TRITIUM WASTE INVENTORY IN COMMERCIAL SITES (1982)

<u>SITE</u>	<u>PERCENT OF TOTAL</u>
WEST VALLEY, NY	7
MAXEY FLATS, KY	15
SHEFFIELD, IL	10
BARNWELL, SC	43
BEATTY, NV	10
RICHLAND, WA	<u>15</u>
	100

DISPOSAL SUMMARY

<u>SITE</u>	<u>CLIMATE</u>	<u>STATUS</u>	<u>MIGRATION</u>
<u>COMMERCIAL</u>			
WEST VALLEY, NY	HUMID	CLOSED	³ H IN SURFACE WATER
MAXEY FLATS, KY	HUMID	CLOSED	³ H IN SURFACE WATER
SHEFFIELD, ILL	HUMID	CLOSED	NONE
BARNWELL, SC	HUMID	OPEN	NONE
BEATTY, NEV	ARID	CLOSED	NONE
RICHLAND, WA	ARID	OPEN	NONE

<u>DEFENSE</u>			
LOS ALAMOS	ARID	OPEN	³ H VAPOR IN SOIL
NEVADA TEST STATION	ARID	OPEN	NONE
OAK RIDGE	HUMID	OPEN	³ H MIGRATION TO CREEK
SAVANNAH RIVER	HUMID	OPEN	³ H MIGRATION FROM TRENCH

DISPOSAL COST ESTIMATES

<u>PACKAGE</u>	<u>COST</u>		<u>NOTES</u>
	<u>\$/m³</u>	<u>\$/ft³</u>	
LOW CONCENTRATION KILOCURIE PACKAGE	NEGLIGIBLE 9000	270	— FOR 55-GAL DRUM

<u>DISPOSAL</u>	<u>LARGE SITE</u>	<u>SMALL SITE</u>	
	<u>\$/m³</u>		
TRENCH, 8m DEEP	225	800	INCLUDES CAPITAL, OPERATING, CLOSURE, 100 YEAR INSTITUTIONAL CONTROL
TRENCH, 18m DEEP	240	1400	
AUGERED SHAFT, 40m DEEP	270	1900	
TRENCH, ENG. BARRIERS	250	1500	
TRENCH, 7m DEEP	—	175-350	

CROSS SECTIONS IN BURIAL GROUND FLOW PATH

TRITIUM LEACHING TESTS

TRITIATED WASTE PACKAGE

PERMEATION TEST ARRANGEMENT

PERMEATION TEST — — — WATER

PERMEATION TEST — — — OCTANE

PERMEATION TEST — — — OIL