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MODERATOR DETRITIATION AT THE SAVANNAH RIVER PLANT

by

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ABSTRACT

A study has been made of the technical and economic aspects of reducing tritium concentration in Savannah River Plant heavy-water moderator by 90%. A single detritiation plant would serve four operating reactors and the desired tritium reduction would be achieved in less than ten years. The process choice has narrowed to three processes. These involve a front-end extraction or preparation of molecular DT in a D_2 stream, and a back-end fractional distillation of this stream followed by catalytic conversion to make 98% tritium T_2.

INTRODUCTION

Neutron absorption in heavy-water moderator has resulted in appreciable tritium levels in Savannah River Plant (SRP) production reactors. These tritium levels contribute to onsite personnel radiological exposures and offsite population dosage. There has been a continuing SRP effort to limit and reduce radiological health effects, and tritium concentrations in atmospheric or liquid discharges exceed no official guidelines. However, to explore ways to reduce unnecessary dosages, a study was made of processes and their implementation to reduce reactor tritium levels. A 90% tritium level reduction was chosen as basis for planning and estimating purposes. Actual implementation of an SRP moderator detritiation plant will depend on U.S. Department of Energy and Congressional approvals.

SRP TRITIUM GENERATION AND RELEASES

Tritium accumulated in heavy water moderator in the three operating SRP nuclear reactors is in the form of the compound DTO. The tritium has reached an activity level of 15 curies per kilogram of heavy water; the total amount is 11 megacuries. The moderator releases tritium to the environment by evaporation to the atmosphere, leakage to the river, and intentional purges to seepage basins. Reactor tritium releases cause 45% of the calculated offsite plant radioactive personnel dosage, including all radioisotopes. Proposed startup of a fourth SRP reactor will add to the problem as its tritium level slowly increases.

SRP tritiated liquid discharges ultimately appear in the Savannah River. Concentrations and dosage to maximum individuals can be calculated using verified mathematical models for transport and exposure to people (2). Similarly, atmospheric stack releases and their environmental impact can be calculated. The 1983 results for all radioisotopes are summarized in Table 1. Ninety percent of the atmospheric impact, and 87 to 99% of the liquid impact were due to tritium. Doses to maximum individuals were only a fraction of the most stringent exposure guides and natural background radiation as shown in Table 2. Moderator detritiation would reduce the indicated doses by about 40%, assuming no other tritium mitigation efforts were effected at SRP. However, such efforts are now being considered, and if the initiatives are successful, annual doses could be reduced further.

TABLE 1

1983 INDIVIDUAL WHOLE BODY DOSE AT THE SRP PLANT BOUNDARY (2)

	Millirem/Year			
	Adult	Teen	Child	Infant
Atm Releases	1.30	1.42	1.88	0.90
Liquid Releases				
Due to:				
Drinking Water	0.13	0.10	0.18	0.27
Eating Fish	0.13	0.08	0.04	-
Total	1.56	1.59	2.10	1.17

TABLE 2

OFFICIAL RADIATION PROTECTION GUIDES
Whole Body Doses, Millirem/Year

National Committee on Radiation Protection Guides - Above Natural Background	
Population Average	170
Maximum Individual	500
(Natural Background near SRP	93)
Nuclear Regulatory Commission Design Criteria for Commercial Reactors, Dose to Maximum Individual	
Atmospheric Releases/Site	5
Liquid Releases/Reactor	3
Liquid Releases/Site	5
Environmental Protection Agency Radiation Protection Standards, Doses to Maximum Individual	
Atmospheric standard	
For commercial site	25
Proposed for DOE site	25
Drinking water standard, Intake at drinking water plant	4

TRITIUM BUILDUP AND CONTROL

Tritium buildup in heavy water moderator follows the equation

$$dx/dt = A - (\lambda + g)x \qquad (1)$$

where
x = tritium concentration, Ci/kg
t = time, years
A = annual specific tritium generation rate, Ci/kg/yr
λ = fractional decay rate, 0.0565/yr
g = fractional physical loss rate, yr^{-1}

g was determined to be 0.017/yr from measured atmospheric and liquid tritium losses. A was determined to be 1.22 Ci/kg/yr or 1.35 Ci/L/yr, assuming that the present 15 Ci/kg concentration was still increasing and corresponded to 28 years of reactor operation.

The addition of a tritium extraction plant adds a term to equation 1 involving the per-pass fractional extraction ϕ

$$dx/dt = A - (\lambda + g)x - \phi x(m/M) \qquad (2)$$

where
m = feedrate to detritiation plant, kg/yr
M = reactor heavy water inventory, kg

The solution to this equation is

$$x = A/(\lambda + g + \phi m/M) + [x_0 - A/(\lambda + g + \phi m/M)]*\exp[-(\lambda + g + \phi m/M)t] \qquad (3)$$

At steady state, the second term in equation (3) vanishes to zero; the first term gives the relationship between flowrate and steady-state tritium concentration.

Equations 2 and 3 assume that water is removed from and returned to the reactor inventory at a constant rate. The intention is to process water with the highest tritium concentration first. With 90% tritium removal per pass, steady-state tritium level of 1.55 Ci/kg, and a heavy-water inventory of 1,000,000 kg in four reactors, a feed rate of 800,000 kg/yr is required.

To limit the number of curies per shipment from the reactors to the detritiation plant, tank trucks of 7.6 cu m capacity are proposed. This would require 100 shipments of tritiated water to the plant per year.

Figure 1 shows the reduction of tritium concentrations in the four operating reactors, assuming the fourth reactor to have been operating for 7 years when the detritiation plant starts up.

A somewhat more efficient operation can result if an extra batch of heavy water and tankage sufficient to hold that batch are available. In that case, water from any one reactor can be detritiated completely before return to operation. With the same 800,000 kg/yr feedrate, the resulting tritium level is then reduced to 0.9 Ci/kg.

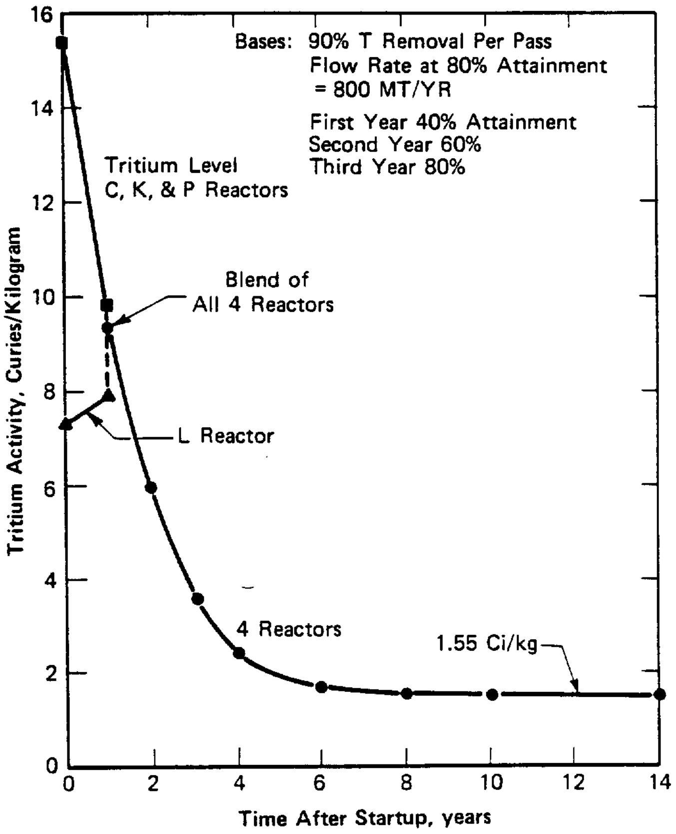

Fig. 1 Predicted Tritium Reduction Scenario

A single detritiation plant was chosen to treat heavy water from all reactors. Because the cost of cryogenic and tritium-qualified process equipment decreases only slowly as the equipment size is reduced (e.g., to one quarter the size), the cost of four separate plants would be substantially greater than that of a single plant.

PROCESS SELECTION

The selection of a process for the moderator detritiation plant is being done in two phases: 1) a preliminary survey of all proposed processes, and 2) an intensive comparision and costing of the prime candidate processes. The first phase has been completed and the three prime candidates from that phase are currently proceeding through the second phase evaluation.

The phase one survey considered nine different process steps or integrated processes (Table 3). Six of the process steps can be classified as "front-end" in that they transfer the tritium from the moderator into a carrier stream with little or no concentration of the tritium. Three of the processes are classified as "back-end" as they concentrate the tritium and regenerate the carrier stream. Each of the "front-end" and "back-end" steps utilizes deuterium gas (D_2) as the carrier, and thus a complete process could be formed out of a choice of any "front-end" step plus a "back-end" step. The laser isotope separation process does not fit into this classification scheme with the others, as it utilizes an entirely different working species ($CTCl_3$) and would achieve a high intial concentration of the tritium; therefore it was considered as a separate integrated process.

TABLE 3

PROCESSES SURVEYED

CATALYTIC EXCHANGE/CRYOGENIC DISTILLATION
- Vapor Phase
- Liquid Phase
- Combined Liquid Phase/Electrolysis

ELECTROLYSIS/RECOMBINATION/CRYOGENIC DISTILLATION
- Direct
- Cascade
- Bipolar

PRESSURE OR TEMPERATURE SWING ABSORPTION
- Coupled with either catalytic exchange or electrolysis front end

LASER SEPARATION
- With $CTCl_3$ intermediate

Processes were evaluated based on two types of criteria. First, the process had to meet the basic tritium removal goals, maintain D_2O product quality, operate safely, and meet environmental performance standards. Second, the processes were compared and judged based on process maturity, safety, simplicity, and economics. The basic tritium removal goal was 90% reduction of the existing tritium level in operating reactors. D_2O quality refers to maintaining extremely low concentrations of ions, such as chloride, that could damage stainless steel equipment; mercury or copper that catalyze the corrosion of aluminum components; and ions such as gadolinium that have high neutron absorption cross sections.

The initial comparison revealed that only three of the front-end and one back-end process should be considered for the second phase evaluation. Figure 2 is a generic diagram of these three combinations. The front-end processes were vapor-phase catalytic exchange (VPCE)(4), liquid-phase catalytic exchange (LPCE)(3,5), and direct-electrolysis and recombination(1). The first two processes exchange the tritium atoms from the oxide molecule to the elemental molecule,

$$DTO + D_2 \longrightarrow D_2O + DT,$$

in the presence of a catalyst. The processes differ in whether the oxide molecule is in the vapor or liquid form when contacted with the catalyst and the gaseous D_2. VPCE proceeds at about 200°C, LPCE at 50-80°C. A single catalyst column can be used for LPCE, while VPCE requires discrete stages arranged countercurrently, each stage consisting of vaporizer, superheater, catalyst column, and condenser/separator. The third front-end process completely electrolyzes the moderator once to form D_2 + DT and oxygen. The DT is removed from the D_2 by the back-end and the purified deuterium is recombined with the oxygen.

Only cryogenic distillation is well enough developed as a back-end to be considered for the second phase evaluation. The catalytic front-ends require about the same size cryogenics, while the electrolytic front requires about half that size. This is because the D_2/D_2O flowsheet ratio in the catalytic front needs to be about 2 to obtain sufficient conversion, while that ratio of course equals one in electrolysis.

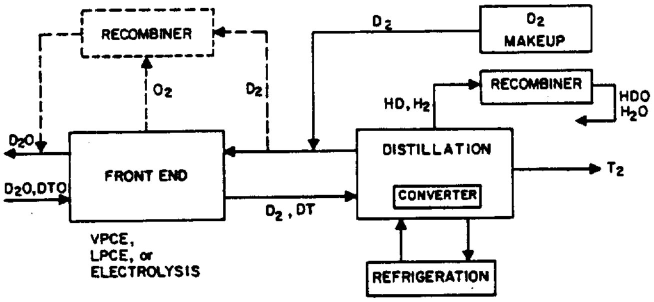

Fig. 2 Flowsheet for Three Preferred Processes

For the second phase (detailed comparison and costing), an in-depth conceptual design was prepared for each of the three prime candidate processes. Proposals were requested from vendors to provide engineering and design services for process facilities, and for ultimately furnishing part or all of such new facilities. The proposals were to include nonbinding "budget quality" estimates of the capital cost of process equipment and assess-assessment of anticipated operating costs. The Du Pont Engineering Department prepared estimates of the capital cost for the moderator detritiation plant including the cost of the vendor-supplied process, installation, overhead, auxiliaries, site preparation, and services.

The detailed comparison of the three processes is still in progress, but some preliminary indications are already apparent. Operating costs, when converted to present value (15 year life, 10% discount), are a relatively small fraction of the initial capital cost for all three processes; these operating costs are dominated by manpower costs. Estimated cost differences for the total moderator detritiation plant project are relatively small. The project would include, in addition to a vendor-supplied process, such items as the building, analytical and health protection facilities, and heavy water tankage and shipping facilities. The tentative conclusions imply that final process selection will probably be made on the basis of such issues as reliability, simplicity, safety, and process maturity.

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