Evaluation of Tritium Management Approaches on Tritium Waste Volumes in Reprocessing Plants

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SUMMARY

Four radionuclides have been identified as likely to be volatilized during the reprocessing of nuclear fuel in amounts such that their gaseous release needs to be controlled to meet US regulatory requirements. These radionuclides are ^{3}H, ^{14}C, ^{85}Kr, and ^{129}I. While iodine has been the focus of a number of studies due to the high decontamination factor (DF) requirements on multiple off-gas pathways, it is also clear that the control of tritium could also be a significant issue for future reprocessing plants, especially if processing relatively short-cooled fuels (e.g., <10-yr post reactor discharge). Like iodine, tritium contained in a used nuclear fuel (UNF) matrix will be released into the liquid and off-gas streams during fuel reprocessing. The amount released into the process streams is a function of the type and burnup of the fuel processed, and the chemical processes and processing conditions employed within the reprocessing plant. The required DF for ^{3}H could be as high as 200, depending on the age of the fuel processed.

This study examines three main cases with a number of subcases for comparative purposes. These include:

1. A single acid/water recovery system that handles the entire UNF reprocessing plant (with and without a “bleed” stream), and tritium abatement potentially applied to the off-gas streams. Other variants include the addition of tritium pretreatment, impact of tritium bound in the cladding, and the release of the tritium in a zirconium recycle process.
2. A split acid/water recovery approach in which the acid/water recycle is split into two systems: one for the head-end and high-level waste operations, and one for the balance of the plant. This will be examined with and without bleed streams. This case will also include tritium abatement applied to the off-gas streams.
3. The selective removal of tritium-contaminated water (i.e., HTO) from the recycle acid/water streams.

Using these three cases, steady-state material balances were performed to examine the equilibrium tritium concentrations that are reached within the acid/water recycle systems and the amount of tritium-contaminated water that must be removed from the off-gas streams or as a bleed stream from the plant to achieve an overall plant DF of 100.

To limit the losses of tritium-contaminated water from the plant, the off-gas dew point was controlled. Lowering the dew point increased the tritium concentration within the plant. In the case of the single acid/water recycle system, the tritium concentrations in the acid/water recycle streams ranged from 2.0×10^{-7} to 1.55×10^{-5} mol%, and in the case of the split system the tritium concentrations in the high-tritium acid/water recycle streams ranged from 7.9×10^{-7} to 1.57×10^{-3} mol%.

By adjusting the off-gas dew points, the amount of tritium-contaminated water recovery required could be changed, and in some cases not all off-gas streams required abatement. The amount of tritium-contaminated water removed from the off-gas streams that would require solidification ranged from 2000 to 50,000 mol/h.

In most cases examined, the evaluated tritium bleed stream from the acid/water recycle system was inadequate to eliminate the need for tritium abatement on the off-gas streams. A bounding case was created to achieve this by limiting the dew point of all of the off-gas streams to -40 °C. In this case the amount of tritium-contaminated water that was bled out and would need to be

solidified was about 9000 mol/h. The application of a tritium-selective membrane to remove and concentrate the tritium-contaminated water from the acid/water recycle system(s) to the extent needed to avoid the need for tritium abatement on of the off-gas streams resulted in slightly higher amounts of tritium-contaminated water to be managed. However, this conclusion is directly attributed to the assumed concentration factor of 1000. A higher factor could significantly reduce this amount.

The smallest amount of tritium-contaminated water to be managed is associated with the use of tritium pretreatment to remove the tritium prior to the aqueous portion of the UNF recycling plant. If 100% of the tritium is released into the tritium pretreatment off-gas (TPTOG) stream, ~0.5 mol/h of tritium-contaminated water must be recovered and solidified. This is four orders of magnitude less than for all other cases examined.

From this analysis, two aspects warranting further development and verification include:

1. Demonstration that TPT can effectively remove >99.5% of the tritium from the fuel meat. This has the potential to eliminate the need for tritium management on any of the other off-gas streams and to significantly reduce the amount of tritium-contaminated water that must be solidified.
2. Development of a tritium selective membrane capable of maintaining tritium concentration at $<1.0\times10^{-7}\%$ tritium concentration and capable of tritium concentration factors >10,000.

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ACRONYMS

DF	decontamination factor
DOG	dissolver off-gas
MTIHM	metric tonne initial heavy metal
TPT	tritium pretreatment
TPTOG	tritium pretreatment off-gas
UNF	used nuclear fuel
VOG	vessel off-gas
WOG	waste off-gas
ZrROG	zirconium recycle off-gas

EVALUATION OF TRITIUM MANAGEMENT APPROACHES ON TRITIUM WASTE VOLUMES IN REPROCESSING PLANTS

1. INTRODUCTION

Four radionuclides have been identified as likely to be volatilized during the reprocessing of nuclear fuel in amounts such that their gaseous release needs to be controlled to meet US regulatory requirements [Jubin et al., 2011; Jubin et al., 2012]. These radionuclides are ^{3}H, ^{14}C, ^{85}Kr, and ^{129}I.

Previous studies have discussed the need for treating off-gas streams to control ^{3}H, ^{14}C, ^{85}Kr, and ^{129}I releases at used nuclear fuel (UNF) facilities and the regulations that specify the limits for releases from such facilities [Jubin et al., 2012]. These regulations ultimately prescribe the decontamination factor (DF) that must be met in the facility. Jubin et al. [2013] examined the role of off-gas pathways since the gaseous radionuclides can partition to different reprocessing off-gas streams. Based on this report and subsequent analysis [Jubin et al., 2014], the required DF for ^{129}I could be 1000 to 3000. The required DF for ^{3}H could be as high as 200, depending on the age of the fuel processed. The DF for ^{85}Kr could be up to ~35, also depending on fuel age. The required DF for ^{14}C is 1 in many cases (no treatment required) but could be higher in specific cases.

While iodine has been the focus of a number of studies due to the high DF requirements on multiple off-gas pathways, it is clear from the studies noted above that the control of tritium will also be a significant issue for future reprocessing plants, especially if processing relatively short-cooled fuels (<10-y post reactor discharge).

Tritium contained in a UNF matrix will be released into the liquid and off-gas streams during fuel reprocessing. The amount released into the process streams is a function of the type and burnup of the fuel processed and the chemical processes employed within the reprocessing plant.

2. Cases Evaluated

A previous study by Grimes et al. [1982] examined the tritium control and disposal options for tritium and compared the use of either tritium pretreatment (TPT) or an aqueous tritium-containing “bleed” stream that was subsequently processed. This study will examine three main cases with a number of subcases for comparative purposes. These include:

1. Case 1 uses a single acid/water recycle system, tritium abatement is applied to the off-gas streams in most subcases (one subcase will examine no tritium abatement), no TPT of the incoming fuel in most subcases (one subcase will examine the impact of TPT). In this case, the tritium is assumed to accumulate within the plant until the tritium discharge rate to the off-gas streams equals the feed rate of tritium entering the plant in the UNF stream. Figure 1 is a schematic diagram showing the reprocessing facility as a block diagram with the main off-gas treatment streams noted.

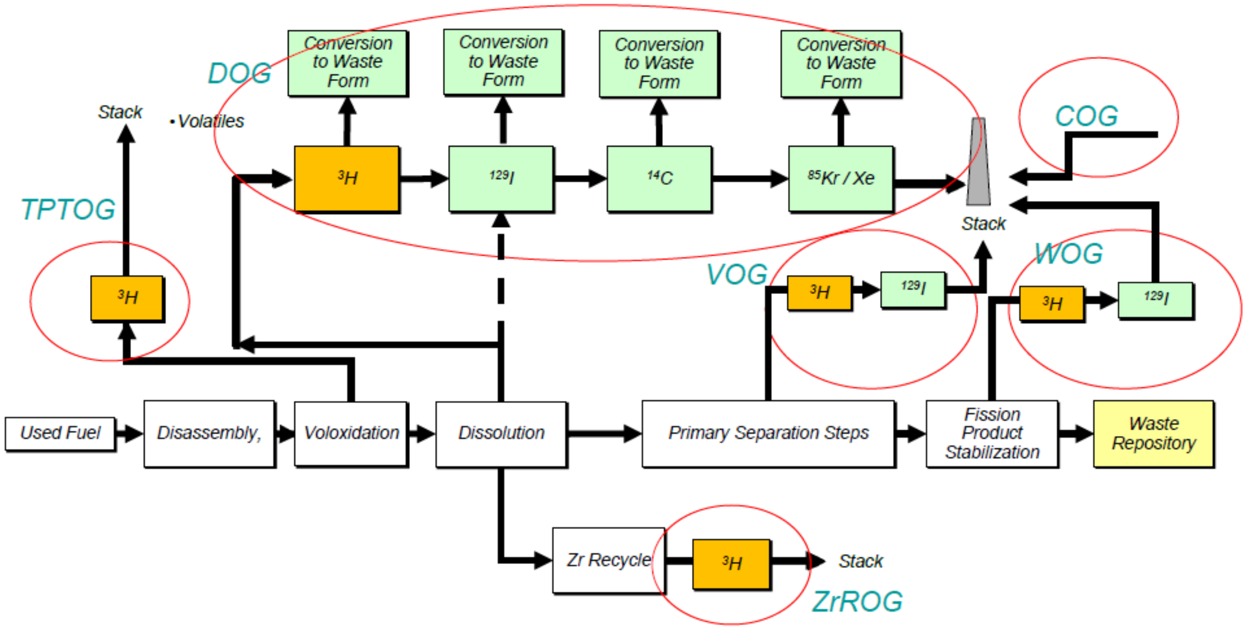

Figure 1. Block diagram of tritium material balance problem.

a. This subcase will be evaluated using a number of off-gas conditions to examine the effects of simply varying the use of condensers on the off-gas streams as a means to control the release of tritium from the reprocessing plant. The volume of tritium-contaminated water/acid lost to the stack is assumed to be made up with fresh water and acid to maintain a constant total water/acid inventory within the plant.

b. Subcase 1b is a minor variant of subcase 1a and will be evaluated using the same conditions as case 1a, with the addition of tritium abatement systems to examine their impact on the resulting off-gas streams and on the volume of resulting tritium-contaminated water to be grouted for disposal. The volume of tritium-contaminated water/acid lost to the stack is assumed to be made up with fresh water and acid to maintain a constant total water/acid inventory within the plant. This subcase will be used as the basis for comparison for all other subcases.

c. Subcase 1c includes the use of a tritium bleed stream in which some fraction of the water/acid inventory within the plant is removed from the system and replaced with fresh acid and water. The removal of some fraction of the tritium-contaminated water/acid from the recycled nitric acid is expected to reduce or eliminate the requirement to treat the off-gas streams. The benefit in terms of the water removal will be compared to subcase 1b.

d. Subcase 1d examines the application of standard tritium pretreatment using air or O_2. This variant also examines the impact of the efficacy of the process on the overall tritium balance. This case will be compared to subcase 1b.

e. Subcase 1e examines the impact of the tritium distribution between the cladding and the fuel meat. Also as part of this subcase, the recovery of Zr from the UNF cladding will be examined. In this case, the tritium contained within the cladding is released into a separate stream. The total tritium introduced into the plant remains the same, but the quantity contained in the fuel meat is reduced. This case will be compared to subcase 1b.

2. Case 2 contains several subcases that explore the impact of separating the acid/water recycle system into two smaller systems with little cross contamination, namely high- and low-tritium concentration acid/water recycle systems. In this case, the tritium is assumed to accumulate within the plant until the tritium discharge rate to the off-gas streams equals the feed rate of tritium entering the plant in the UNF stream. A separate aqueous system is used for head-end, first-cycle solvent extraction and high-level waste, and a second system is used for the balance of the plant. This limits the spread of tritium from head-end and first- cycle solvent extraction. The extent of the spread is impacted by the entrainment of aqueous streams between the first and second cycles of solvent extraction. This essential establishes a tritium barrier between the two acid/water recycle systems.
    a. Subcase 2a examines the implementation of tritium abatement on the off-gas stream to achieve the desired overall plant tritium DF. The volume of resulting tritium-contaminated water to be grouted for disposal is determined.
    b. Subcase 2b examines the implementation of a tritium bleed stream in which some fraction of the aqueous (water/acid) inventory is removed from the system and replaced with fresh acid and water. The removal of some fraction of the HTO-containing aqueous stream from the recycled nitric acid is expected to reduce or eliminate the requirement to treat the off-gas streams. The benefit of subcase 2b in terms of water removal will be compared with subcases 1b and 2a.
3. Case 3 involves the selective removal of tritium-contaminated water (i.e., HTO) from the recycle acid/water streams. This case will be compared to subcase 1b.

3. Assumptions

This analysis is based on the following assumptions:

- The example UNF reprocessing plant is sized to process 1000 MT/yr and operate 250 d/yr. The fuel feed rate is 0.1666 MT/h.
- The fuel to be processed has a burnup of 60 GWd/MTIHM cooled 5 years. Tritium content of the fuel is 790.3 Ci/MT. The resulting tritium feed rate is 131.7 Ci/hr (0.0136 g tritium/hr or 0.0045 mol tritium/hr).
- Dissolver off-gas (DOG) rate is 1000 m^3/h.
- Vessel off-gas (VOG) rate is 10,000 m^3/h.
- Waste off-gas (WOG) rate is 500 m^3/h.
- In most cases, 100% of the tritium is assumed to be found in the fuel meat. The actual values that can be found in the literature vary widely, from10% to 90% [Robinson and Jubin, 2013].
- For this discussion, the term “tritiated water” should include both T_2O and HTO. However, within this study only HTO is assumed. Tritium-contaminated water refers to a mix of HTO, T_2O, and H_2O.

- The UNF reprocessing plant has no liquid discharge streams. This implies that all liquid waste is solidified or is lost via evaporation. High-level waste is assumed to be vitrified. The liquid is evaporated, and water and acid are recovered from the off-gas stream for reuse within the plant. Tritium-contaminated water vapor not recovered during waste processing is discharged to the stack. Tritium-contaminated water/acid purges and low-level liquid waste are grouted.
- Water losses from the UNF reprocessing plant are determined based on cooling the off-gas to a specified temperature to condense and recycle the condensate within the plant.
- In most cases, it was assumed that tritium capture on gas effluent systems was sufficient to achieve a desired DF. In the cases where tritium capture from the off-gas streams was employed, impacts of an overall tritium DF for the plant of 100 was considered.
- Tritium is captured from off-gas streams after cooling to a desired dew point. The condensate produced by cooling the off-gas stream to a specified dew point is assumed to be recycled within the plant. A solid sorbent-based system is used to recover the tritium-contaminated water from the off-gas stream. The sorbent is assumed to be regenerated, and the tritium-contaminated water is recovered for immobilization most likely as grout. The grouted waste stream is an exit point from the overall material balance.
- Water and acid are introduced into the UNF reprocessing plant to compensate for water and acid losses to the stack.
- Once steady state with regard to tritium accumulation is reached, the ratio of tritiated water and normal water is the same throughout the plant. In the case of the tritium barrier the plant has two zones, each with a specific tritiated water-to- normal water ratio as described in the discussion of case 2.
- The tritium-contaminated liquid water and the tritium-contaminated water vapor in equilibrium over it have the same tritiated water-to-normal water ratio (i.e., no isotopic enrichment).

For the cases where TPT is employed, additional assumptions are made:

- The TPT off-gas stream is 270 m^3/hr or 350 kg dry air/hr.
- The water content of the off-gas can be reduced to a -60 °C dew point using solid sorbents. Humidity at -60 °C is 0.0000072 kg H_2O/kg dry air. At the -60 °C dew point, the TPT off-gas stream would contain 0.002515 kg/h H_2O.
- Hundredfold additional water based on the amount of hydrogen and tritium in the fuel is added to the TPT off-gas stream, and the dew point of the resulting moist off-gas stream is reduced back to -60 °C, facilitating a DF of 100 for water. This results in the removal of ~0.25 kg/hr of water removed from the TPT off-gas stream.

For the cases where zirconium (Zr) recycle is employed on the cladding, additional assumptions are made:

- In subcase 1e, 60% of the tritium is contained in the fuel meat, and 40% is contained in the cladding. This results in a tritium content of the fuel of 474.2 Ci/MT. The resulting tritium feed rate is 79.03 Ci/hr (0.0082 g tritium/hr or 0.0027 mol tritium/hr) from the fuel meat into the dissolver and balance of the plant. The remaining 40% of the tritium either remains in the cladding and is disposed of as part of the solid cladding waste or is released during the recovery of Zr from the cladding.

- The Zr Recycle off-gas (ZrROG) is a recycled Cl_2 stream. The tritium accumulates within the recycle stream as TCl, along with any HCl resulting from the chlorination of Zr hydrides in the cladding.
- The hydrogen and tritium from the HCl and TCl are assumed to be converted to HTO and H_2O by passing them through a metal oxide bed, and the resulting tritium-contaminated water stream is captured in a manner similar to that used for the TPT system.

4. Methodology

To facilitate an understanding of the distribution of the tritium within the reprocessing plant, a model was developed to track the accumulation of tritium within the plant. The model is a modification of an Excel® spreadsheet tool previously developed to evaluate the impacts of the treatment capabilities, employed on each off-gas stream within the reprocessing plant, on the overall plant DF [Jubin, 2015]. It also tracks the accumulation of tritium within the plant and may be used to evaluate the associated volatilization of the HTO.

This analysis requires the solution of simultaneous liquid/vapor equilibrium and tritium material balances. These are solved either over the entire reprocessing plant or over specific portions of the plant. To better understand the process, a simple example will be used. Consider the system shown in Figure 2.

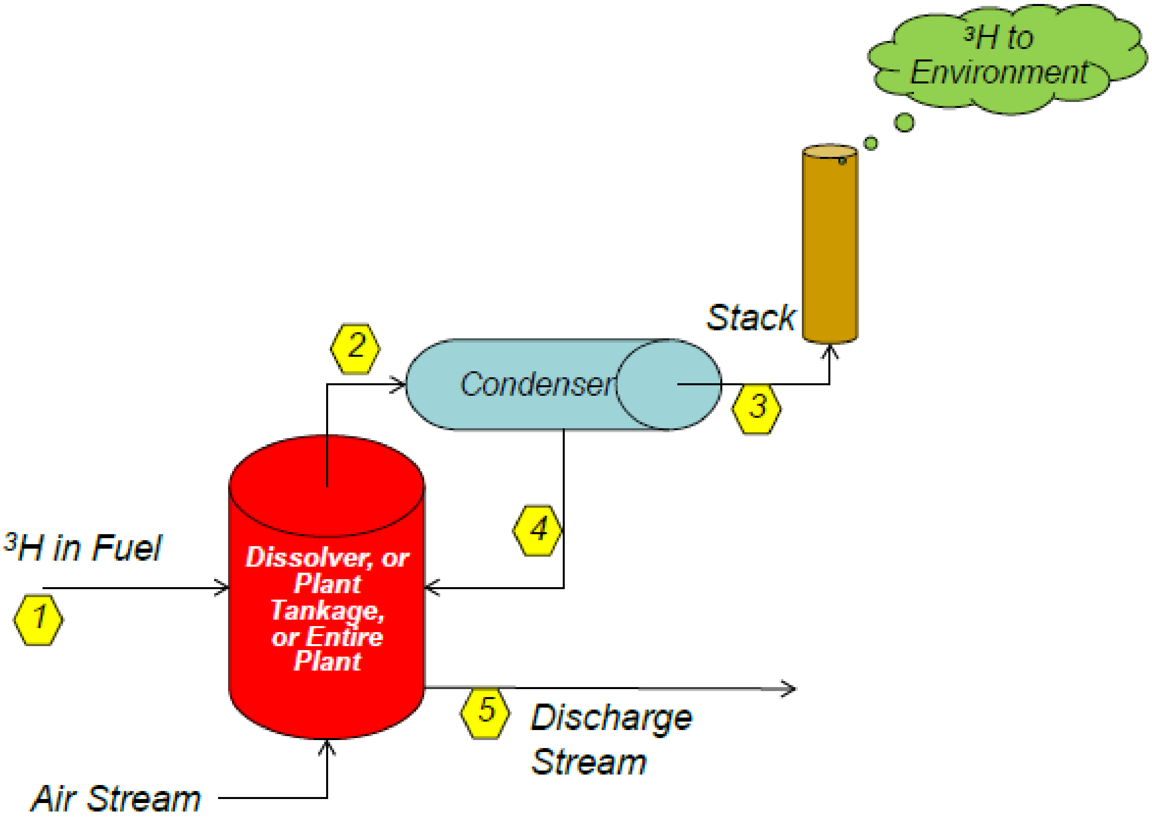

Figure 2. Block diagram of tritium material balance problem.

Approach to steady state is assumed with the accumulation of tritium in the recycle acid/water system until the losses to the stack equal the tritium fed into the plant in the fuel. The following logic is then used to determine the concentration of tritium in the liquid in the facility tanks/processes and the rate of tritium loss to the stack.

1. Assume initially that there is no tritium in stream 5. The only pathway for tritium out is the stack. This is based on the assumption of no liquid discharge from the plant and the vitrification of the high-level waste. Stream 5 can also be thought of as optional liquid removal stream that is initially zero flowrate. If it is used as a tritium bleed stream the resulting liquid is assumed to be solidified.

2. Assume that the ratio of tritiated water to normal water is the same throughout the system, as stated in the assumption section of this report. Thus, the vapor and liquid phases are assumed to have the same ratio.

3. To close the overall material balance, the flowrate of tritium in stream 3 must equal the flowrate in stream 1. This implies no accumulation once steady state is achieved.

4. Concentration of tritium in stream 2 is a function of temperature in the tank(s) and is assumed to reach the equilibrium humidity concentration. However, at this point the tritiated water-to-normal water ratio in the tank/process is unknown.

5. The flowrate of tritium in stream 2 is then the equilibrium concentration in the stream multiplied by the gas flowrate.
6. The flowrate of tritium in stream 3 plus stream 4 equals that in stream 2.
7. The water concentration in stream 3 is a function of the condenser temperature and is assumed to be the equilibrium humidity.
8. The flowrate of tritium in stream 3 is then the equilibrium humidity concentration in the condenser multiplied by the tritium-to-water ratio multiplied by the gas flowrate. Stream 4 contains the balance of the tritium.
9. The addition of stream 4 back into the tank changes the tritium concentration in the tank, which in turn changes the tritium concentration in stream 2, until the tritium flowrate in stream 3 equals that in stream 1, and the tritium flowrate in stream 2 equals the sum of the tritium flowrates in streams 1 and 4.

For case 1 consider the systems shown in Figure 3. Three main portions of the plant and associated off-gas streams will be considered: (1) the head-end system containing the dissolver and DOG, (2) the waste treatment systems that solidify the waste and the WOG; and the separations portion of the plant and the VOG systems. It is assumed that there is a single acid/water recycle system and that condensers are located on each of the off-gas streams, such that water losses to the off-gas are controlled by the dew point of the individual off-gas streams. All off-gas streams exit the plant at the stack.

In this example, four key points apply to the evaluation of the tritium balance, denoted with the green triangles:

1. The fuel in;
2. The losses to the stack;
3. The tritium purge (if utilized); and
4. The tritium concentration of the recycle acid/water with in the plant.

By setting the losses to the stack equal to the feed to the plant and determining what fraction of the aqueous stream is to be purged, it is possible to calculate the tritium concentration at steady state in the aqueous recycle within the plant. The portion of the tritium sent to the stack in the off-gas streams is controlled by the dew point and off-gas flowrate. The conditions for subcase 1a (i.e, no tritium abatement) are shown in Figure 4. The dew point of each off-gas stream is set to 30 °C, and there is no tritium purge. The off-gas rates are constant for all of the cases used in this study and are noted in the Section 3 (Assumptions).

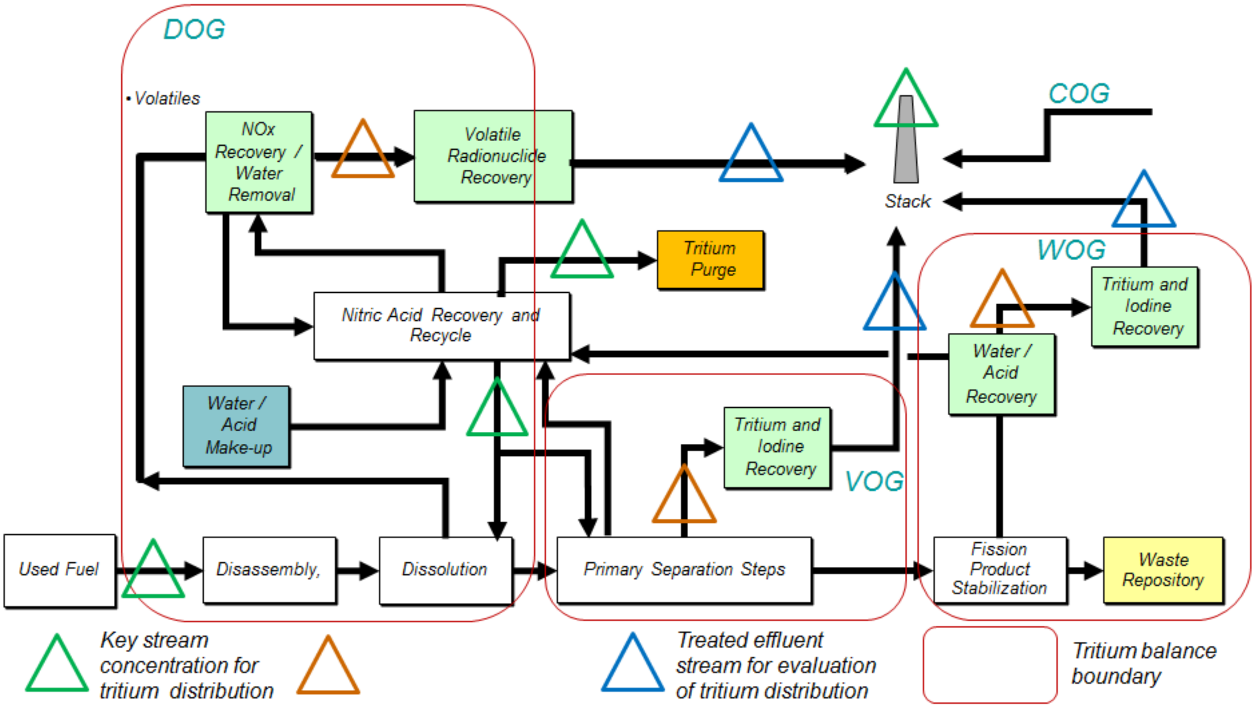

Figure 3. Block diagram of reprocessing plant for use in Case 1 material balance problem.

Figure 5 shows the tritium that would be lost to the off-gas stream assuming no tritium recovery.

If tritium recovery is employed, the amount of tritium and associated water that must be recovered from each off-gas stream is simply calculated by the application of the associated DF for the stream and the steady-state tritium concentration in the acid/waste recycle stream. For this example, the tritium concentration of the acid/water recycle stream is 2.03×10^{-5} mol%, and the tritium DF for each of the off-gas streams is set to 100. The total potential water loss to the stack is 22,420 mol/h, and applying the DF of 100 for tritium results in a total recovery requirement of 21,195 mol/h, of which 1930 mol/h is recovered from the DOG; 19,300 mol/h is recovered from the VOG; and 965 mol/h is recovered from the WOG. (This will be further discussed in Section 5.1.2.)

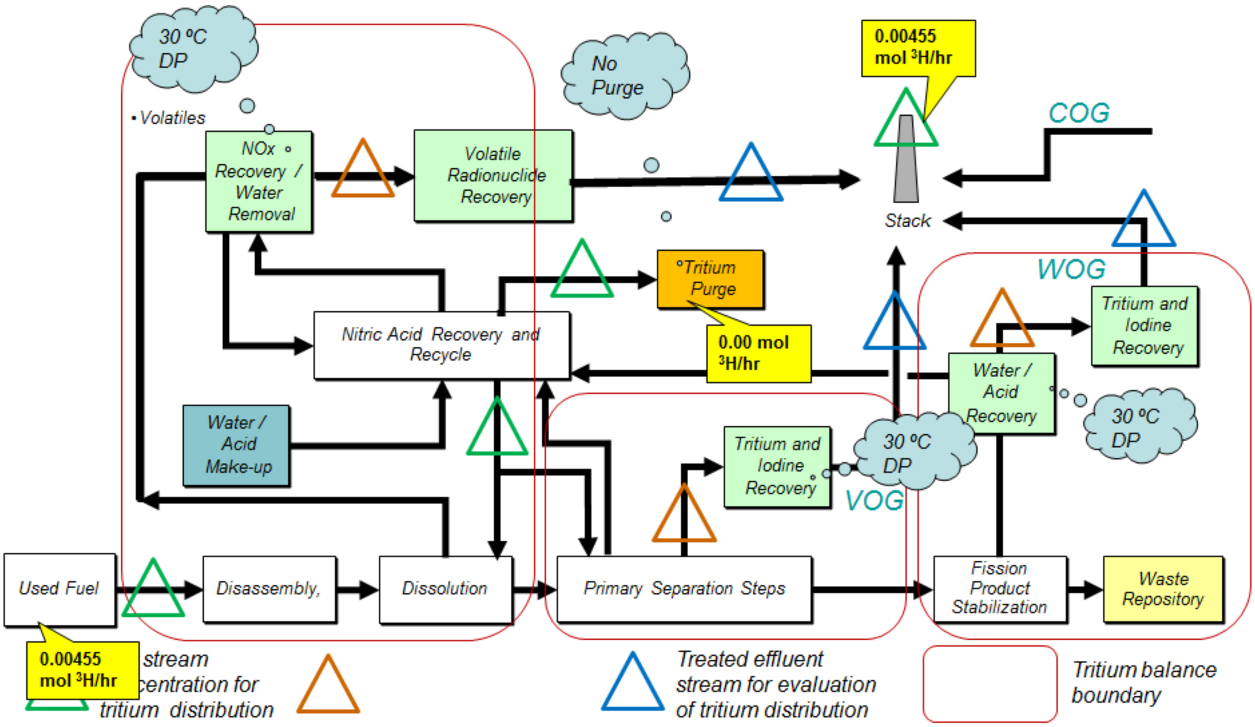

Figure 4. Block diagram of reprocessing plant for use in Case 1 material balance problem showing key operating assumptions.

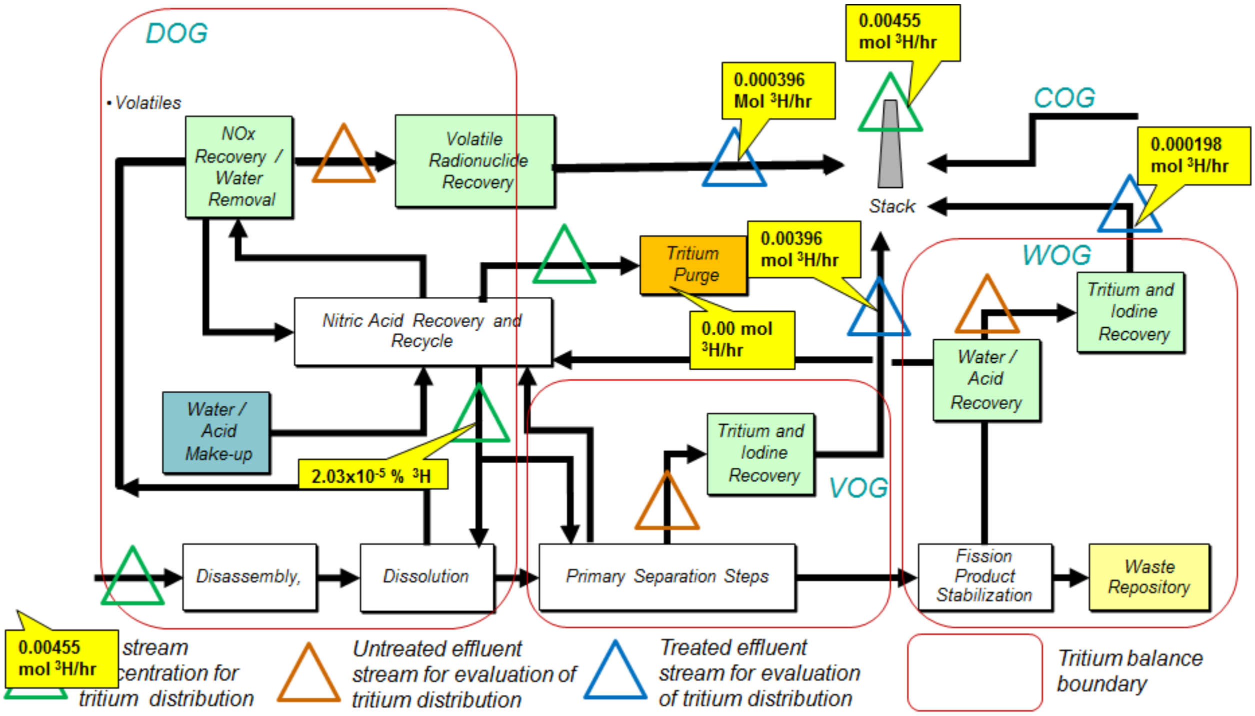

Figure 5. Block diagram of reprocessing plant for use in Case 1 material balance problem showing tritium distribution for conditions in Figure 4.

5. Results

5.1 Case 1 – Single Acid/Water Recycle System

This case will be divided into five subcases.

5.1.1 Subcase 1a results

This is the simplest of the subcases to be examined. In subcase 1a, there is no tritium pretreatment, no tritium abatement on off-gas streams, and a single acid/water recycle system. The tritium is assumed to accumulate within the plant until the tritium discharge rate to the off-gas streams equals the feed rate of tritium entering the plant in the UNF stream. This subcase will be evaluated using a number of off-gas conditions to examine the effects of simply varying the condenser operating condition for the off-gas streams as a means to control the loss of tritium from the reprocessing plant. The condensate from the condensers is assumed to be routed back to the acid/water recycle system either directly or, as in the DOG system, indirectly via the dissolver. The volume of tritium-contaminated water/acid lost to the stack is assumed to be made up with fresh water and acid to maintain a constant total water/acid inventory within the plant. Table 1 presents the results from this analysis. In subcase 1a-1, all the condensers are operated to result in a dew point of 30 °C for all of the off-gas streams. This results in the tritium distribution to the off-gas streams of 8.7% to the DOG, 87.0% to the VOG, and 4.3% to the WOG, and a total tritium-contaminated water loss to the stack of 22,420 mol/h. The equilibrium tritium concentration in the recycle acid/water stream is 2.0×10^{-5} mol%. Subcases 1a-2 and 1a-3 decrease the dew point of the DOG to 10 and -40 °C, respectively, in an attempt to decrease the tritium-contaminated water loss to the point that it might not require treatment. This clearly reduces the tritium losses in the DOG (2.6% and 0.04%, respectively), but it also results in a corresponding increase in the tritium losses to the VOG and the WOG as well as increases to the tritium content in the recycle water/acid systems. The total tritium-contaminated water losses to the stack are also decreased as a result of the lowering of the dew point in the DOG stream.

Subcase 1a-4 decrease the dew point of the VOG and WOG to 10 °C, while holding the DOG dew point at -40 °C in an attempt to decrease the tritium-contaminated water loss to the stack. This results in the tritium distribution to the off-gas streams of 0.14% to the DOG, 95.1% to the VOG, and 2.8% to the WOG, and a total tritium-contaminated water loss to the stack of 5752 mol/h. The equilibrium tritium concentration in the recycle acid/water stream is 7.9×10^{-5} mol%. This is a 75% reduction in the amount of tritium-contaminated water that reaches the stack, but the total amount of tritium in the stream is the same. The tritium content in the recycle acid/water stream is increase by about a factor of 4.

In summary, for subcase 1a, the primary tritium pathway to the stack is the VOG. This is directly tied to the relative off-gas flowrates where the VOG flow is 10 times the DOG flow. Lowering the dew point reduces the total water losses and can shift the relative distribution. Lowering the dew point also increases the tritium concentration within the recycle acid/water system.

Table 1. Subcase 1a: Tritium distribution to the plant off-gas systems and total tritium-contaminated water/acid losses to the stack

Case number	DOG dew point (ºC)	VOG dew point (ºC)	WOG dew point (ºC)	Tritium purge rate (%/h)	% ^{3}H to DOG (%)	% ^{3}H to VOG (%)	% ^{3}H to WOG (%)	[^{3}H] in recycle (%)	Total tritium-contaminated water loss to stack (mol/h)	Effective tritium DF for plant
1a-1	30	30	30	0.00%	8.70%	87.0%	4.3%	2.0E-05	22,420	1
1a-2	10	30	30	0.00%	2.60%	92.8%	4.6%	2.2E-05	21,017	1
1a-3	-40	30	30	0.00%	0.04%	95.2%	4.8%	2.2E-05	20,478	1
1a-4	-40	10	10	0.00%	0.14%	95.1%	4.8%	7.9E-05	5752	1

5.1.2 Subcase 1b results

Subcase 1b is virtually the same as subcase 1a, with the addition of tritium abatement on the three off-gas streams. This is the simplest of the subcases to be examined that actually limits the discharge of tritium from the reprocessing plant and will be used as a basis of comparison for all other cases. In subcase 1b there is no tritium pretreatment. Tritium abatement to achieve the desired tritium DF of 100 is applied to all three off-gas streams, and a single acid/water recycle system is employed. As in subcase 1a, the tritium is assumed to accumulate within the plant until the tritium discharge rate in the off-gas streams leading to the tritium abatement systems equals the feed rate of tritium entering the plant in the UNF stream. The tritium abatement systems are located downstream of the off-gas condensers. The tritium- contaminated water recovered in the tritium abatement systems is assumed to be grouted. Subcase 1b-1 was described in the example discussed in Section 4. For subcase 1b-1, the tritium concentration of the recycle acid/water stream is $2.03\times10\%^{-5}$. The total potential tritium contaminated water loss to the stack is22,420 mol/h, which is exactly the same values as for subcase 1a-1. Applying the DF of 100 for tritium results in a total recovery requirement of 21,195 mol/h, of which 1930 mol/h is recovered from the DOG; 19,300 mol/h is recovered from the VOG; and 965 mol/h is recovered from the WOG.

Subcases 1b-2 through 1b-4 parallel subcases 1a-2 through 1a-4. The results for subcase 1b are presented in Table 2. Note that as the aqueous concentration is driven up, the volume of tritium-contaminated water that must be captured and solidified decreases to achieve the same DF. The increase in concentrations and the decrease in volumes are inversely related and linear. It is also worth noting that in the case where the dew point of the DOG is reduced to -40 °C, the potential losses via the DOG are significantly less than 1%. As a result, it may be possible to avoid the application of tritium abatement to that stream and still achieve the overall desired DF.

5.1.3 Subcase 1c results

This subcase included the use of a tritium bleed stream in which some fraction of the aqueous (water/acid) stream is removed from the system and replaced with fresh acid and water to make up for the water removed. The objective of this subcase was to explore the possibility of significantly reducing or eliminating the need to utilize tritium abatement on the off-gas streams by directly removing some fraction of the recycle acid/water stream and solidifying it directly as a waste form. The benefit in terms of the water removal will be compared to base case 1b.

In subcase 1c-1, the same off-gas condenser conditions found in subcase 1b-4 are used with the addition of a bleed stream of 1%/h of the 100,000-l aqueous inventory. This bleed stream removes 87.8% of the tritium from the plant in a 41,444 mol/h stream yielding an effective tritium DF of 8.2. The use of the tritium bleed reduces the tritium concentration in the aqueous system from $7.9\times10^{-5}\%$ to $9.6\times10^{-6}\%$. However, this still means that 12.2% of the tritium reports to the off-gas streams, with 0.02% of the tritium reporting to the DOG, 11.6% reporting to the VOG, and 0.6% reporting to the WOG. A tritium DF for the off-gas streams of 12.2 is still required. This results in the need to remove 5280 mol of tritium-contaminated water/h from the 5752 mol/h of tritium-contaminated water exiting the condensers. This would mean that 46,714 mol of tritium- contaminated water/h must be managed as a tritium waste stream. The DFs for this case are shown in Table 3.

Table 1. Subcase 1b: Tritium distribution to the plant off-gas systems and total tritium-contaminated water/acid losses to the stack

Case number	DOG dew point (ºC)	VOG dew point(ºC)	WOG dew point (ºC)	Tritium purge rate (%/h)	Desired overall DF	% ^{3}H to DOG (%)	% ^{3}H to VOG (%)	% ^{3}H to WOG (%)	Tritium concentration in recycle (%)	Total tritium-contaminated water loss to stack (mol/h)	Tritium-contaminated water recovered from stack (mol/h)
1b-1	30	30	30	0.00%	100	8.70%	87.0%	4.3%	2.0E-05	22,420	22,195.32
1b-2	10	30	30	0.00%	100	2.60%	92.8%	4.6%	2.2E-05	21,017	20,806.86
1b-3	-40	30	30	0.00%	100	0.04%	95.2%	4.8%	2.2E-05	20,478	20,273.04
1b-4	-40	10	10	0.00%	100	0.14%	95.1%	4.8%	7.9E-05	5752	5694.18

Sub case 1c-2 reduces the larger-than-practical fractional bleed stream to 0.1%/h of the 100,000-l inventory. This obviously cuts the bleed stream by a factor of 10 to 4144 mol/h. This decreases the tritium removal to 41.9%, or an effective DF of 1.7. The tritium concentration in the water/acid recycle stream also increases to 4.6×10^{-5}%. An effective off-gas DF of 58.1 is required on the stack gas, which in turn requires the additional recovery of 5653 mol/h of tritium-contaminated water, for a total of 9797 mol/hr to be managed.

Subcase 1c-3 assumes a higher VOG and WOG dew point. As noted in previous sections, this reduces the tritium concentration in the recycle acid/water but allows more tritium- contaminated water to reach the tritium abatement systems. In this subcase the required DF for the off-gas systems is 83.2, and the total tritium-contaminated water to be managed is 24,376 mol/h.

Subcases 1c-4 and 1c-5 examine the impact of increasing the aqueous inventory of the facility. These subcases increase the inventory from 100,000 l to 1,000,000 l. Note that in subcase 1c-5 all conditions are exactly the same as in subcase 1c-3—except for the removal fraction, which is 0.01% vs 0.1%—but due to the increase in inventory, the quantity of tritium-contaminated water removed is exactly the same as its tritium concentration. As a result, the effective DFs and quantity of tritium-contaminated water to be managed are exactly the same. But plant *inventory* of tritium increased by tenfold.

Subcase 1c-6 examines the impact of higher off-gas dew points. This case allows comparison with subcases 1b-1 and 1c-3. Increasing the dew point of the DOG from -40 °C to 30 °C results in an increased portion of the tritium-contaminated water reaching the DOG tritium abatement system (7.34% vs 0.03%). The increase in tritium-contaminated water vapor in the DOG increases the total tritium-contaminated water recovery requirement from 24,376.04 to 26,298.32 mol/hr. This is associated with a slight decrease in the tritiated water concentration in the recycle acid/water from 1.8×10^{-5} to 1.7×10^{-5} %.

Subcase 1c-7 examines the situation where the dew point of the off-gas streams is lowered to the extent that the losses to the stack without tritium abatement are limited to 1% by the dew point and by the tritium bleed. The dew point of the DOG, VOG, and WOG are set to -40 °C, and the percent tritium bleed is varied to control the tritium concentration in the recycle acid/water system. The -40 °C dew point represents a case of near-complete capture of the tritium-contaminated water from the off-gas streams with the captured tritium-contaminated water being recycled to the acid/water recovery systems. This represents a bounding case where 99% of the tritium is pushed to the tritium bleed stream. An overall tritium DF of 100 is achieved with a tritium bleed of 0.21% per hour or 8703 mol/h from the 100,000-L inventory. This represents only a slightly higher amount of tritium-contaminated water than must be managed in subcase 1b-4. The associated tritiated water concentration in the recycle acid/water is 5.2×10^{-5} percent.

Even a 1%/hr tritium bleed stream resulted in an effective tritium DF of <10. Thus off-gas treatment to recover tritium is still required, and the total volume of tritium-contaminated water to be treated is greater than using off-gas treatment alone. This mode of operation did result in very low release fractions to the DOG.

Table 2. Subcase 1c: Tritium distribution to the plant off-gas systems with a tritium bleed stream and total tritium-contaminated water/acid losses to the stack

Case number	DOG dew point (ºC)	VOG dew point (ºC)	WOG dew point (ºC)	Tritium purge rate (%/h)	% ^{3}H to DOG (%)	% ^{3}H to VOG (%)	% ^{3}H to WOG (%)	Tritium concentration in recycle (%)	Tritium removed in bleed stream (%)	Tritium-contam-inated water removed in bleed (mol/h)	Tritium-contam-inated water recovered from stack (mol/h)	Total tritium-contam-inated water recovery required (mol/h)	Effective DF of bleed/off-gas DF required	Notes
1b-1	30	30	30	0.00%	8.70%	87.0%	4.3%	2.0E-05	0.0%	–	22,195.32	22,195.32	1/100	
1b-4	-40	10	10	0.00%	0.14%	95.1%	4.8%	7.9E-05	0.0%	–	5694.18	5694.18	1/100	
1c-1	-40	10	10	1.00%	0.02%	11.6%	0.6%	9.6E-06	87.8%	41,444	5279.73	46,724.18	8.2/12.2	1
1c-2	-40	10	10	0.10%	0.08%	55.3%	2.8%	4.6E-05	41.9%	4144	5652.73	9797.18	1.7/ 58.1	1
1c-3	-40	30	30	0.10%	0.03%	79.2%	4.0%	1.8E-05	16.8%	4144	20,231.59	24,376.04	1.2/83.2	1
1c-4	-40	30	30	0.10%	0.01%	31.5%	1.6%	7.3E-06	66.9%	41,444	19,858.59	61,303.04	3.0/33.1	2
1c-5	-40	30	30	0.01%	0.03%	79.2%	4.0%	1.8E-05	16.8%	4144	20,231.59	24,376.04	1.2/83.2	2
1c-6	30	30	30	0.10%	7.34%	73.4%	3.7%	1.7E-05	15.60%	4144	22,153.88	26,298.32	1.18/84.4	1
1c-7	-40	-40	-40	0.21%	0.09%	0.9%	0.0%	5.2E-05	99.0%	8703	0	8703	100/1	1

Notes:

1. Recycle water/acid volume: 100,000 l
2. Recycle water/acid volume: 1,000,000 l

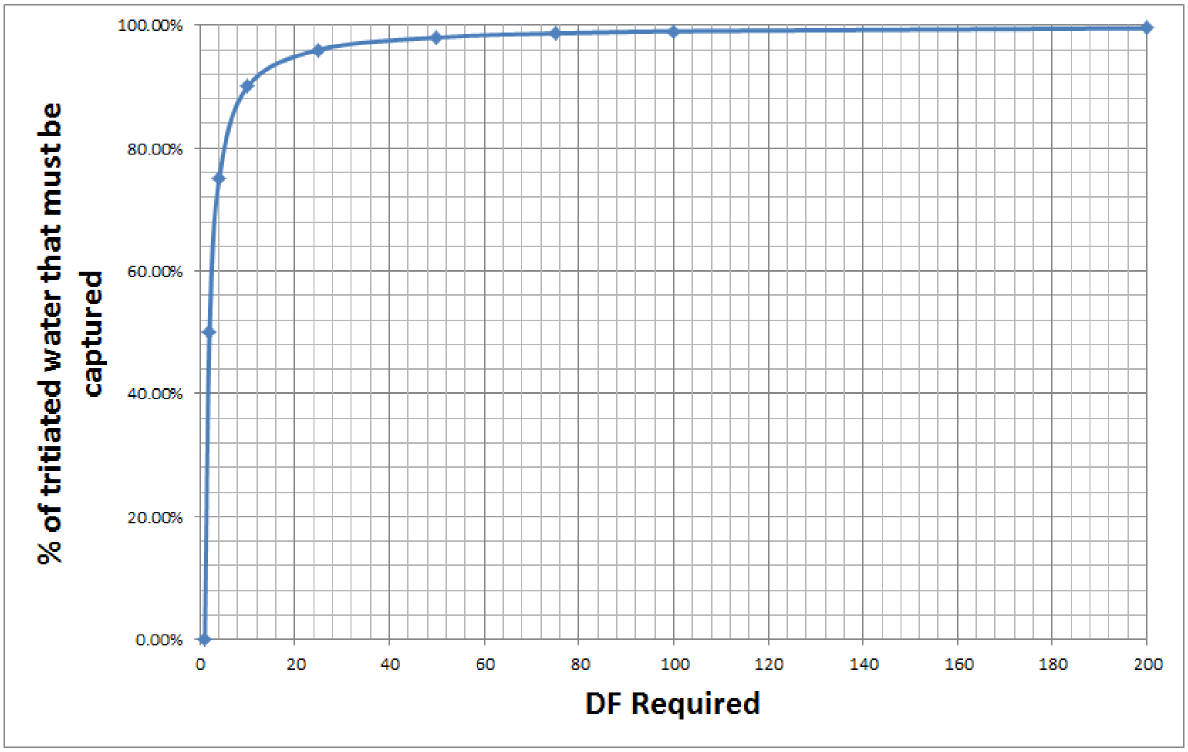

Figure 6. Percent capture requirement as a function of needed decontamination factor.

It is also worth pointing out the relationship of the required DF to the amount of tritium-contaminated water that must be recovered. Since DF is defined as the ratio of the amount of tritium leaving the tritium recovery process in the off-gas stream to the amount of tritium in the off-gas fed to the tritium recovery process, a DF of 100 requires the recovery of 99% of the tritium in the feed, and a DF of 10 requires the recovery of 90%. In the best case shown in Table 3 (subcase 1c-1) the required DF for the off-gas streams was reduced to ~12 and would require >90% recovery of the tritium-contaminated water contained in the off-gas stream to achieve this. Figure 6 shows the relationship of % recovery to required DF. For the sake of discussion the DF of a stream can be defined as DF=1/(1-x) where x is the fraction of contaminant removed from the stream.

5.1.4 Subcase 1d results

Subcase 1d examines the application of standard TPT using air or O_2. This variant also examines the impact of the extent of the efficacy of the TPT process on the overall tritium balance. This case will be compared to base case 1b as shown in Table 4. In all of the subcase 1d cases, it is assumed that the TPT off-gas (TPTOG) is treated in a separate off-gas system and that only after the desired tritium DF is achieved on the TPTOG stream would it be blended into the other off-gas streams going to the stack.

Subcase 1d-1 assumes that 100% of the tritium from the fuel meat is released into the TPTOG streams. This eliminates the concern of tritium accumulation in the aqueous portion of the plant. This stream is very dry, and hundredfold water is added to the stream to facilitate the recovery on a solid sorbent system with a DF of 100. The 0.45 mol/hr of tritium-contaminated water are removed from the TPTOG stream. The tritium concentration in the recycle acid/water system is 0.0.

Subcases 1d-2 and 1d-3 assume that the TPT does not achieve complete removal of tritium from the fuel meat. In both cases 1% of the tritium is not removed and reaches the aqueous portion of the plant. At steady state this results in a tritium concentration in the aqueous system of 2.0×10^{-7}%. If the tritium DF on the TPTOG stream is 100, a tritium DF of 100 is also required on the remaining off-gas streams to remove sufficient tritium such that the overall tritium DF is 100. In subcase 1d-2, this means an additional 22,195 mol/hr of tritium-contaminated water must be removed and managed as tritium-contaminated water waste although the level of tritium contamination is very much reduced. This is the same amount as required for subcase 1b-1 with no TPT. If, however, the tritium DF for the TPTOG stream can be increased to 200 as shown in subcase 1d-3 of Table 4, the required DF for the balance of the off-gas streams is only 2. However, it should also be noted that a DF of 2 only reduces the amount of water that must be recovered by half.

Subcase 1d-4 examines the impact of lowering the dew point of the DOG, VOG, and WOG streams to allow the comparison with subcase 1b-4 while holding the TPTOG DF at 200. As shown in subcase 1b-4, the lowering of the dew point on the streams associated with the aqueous systems concentrates the tritium in the acid/water recycle stream. As with subcase 1d-3 a tritium DF of 2 is needed for the DOG/VOG and WOG streams. But due to the lower dew points, virtually all of the tritium losses are in the VOG stream. This would be the only stream requiring additional tritium abatement.

For subcase 1d, it can be summarized that without TPT a significant amount of water must be captured and managed to control tritium release from the reprocessing plant. If greater than 1% of the tritium in the fuel meat reaches the aqueous portion of the plant, tritium abatement will be required on at least the VOG stream to achieve the overall plant DF of 100. Furthermore, if the TPTOG stream has a tritium DF less than or equal to the overall plant tritium DF goal and >1% reaches the aqueous portion, a significant amount of water must be captured and managed to control tritium release to achieve an overall plant DF of 100.

However, if the tritium DF on the TPTOG stream is greater than the overall tritium DF required for the plant, the higher the DF is, the lower the tritium release fraction in the TPT process can be. This is true provided that DF of TPT itself is greater than or equal to overall plant DF which is the limiting condition. Figure 7 is a simple block diagram that shows the relevant tritium splits associated with this analysis.

It can also be shown in this analysis that the capture of tritium from the aqueous portion of the plant can be avoided entirely if (a) the tritium release within the TPT process is greater than 99.5% and (b) a tritium DF of 200 can be achieved on the TPTOG stream (overall DF=100). If the tritium DF on the TPTOG stream is >100 and less than 200, the tritium release fractions must be greater. Table 5 shows the relationship of the achievable tritium DF for the TPTOG stream and the percent of tritium that must be released into the TPTOG stream so that the need to capture tritium from the DOG, VOG, and WOG streams can be avoided. The amount of tritium-contaminated water that must be recovered is orders of magnitude less than if tritium-contaminated water must be recovered from the DOG, VOG, and WOG streams or through the use of bleed streams.

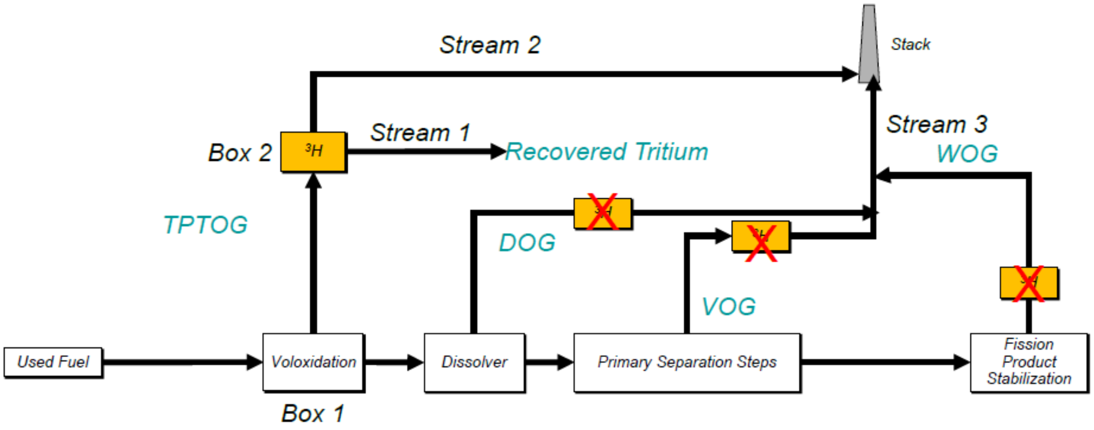

Figure 7. Simple block diagram showing relevant tritium splits (that are presented in Table 5) associated with the TPT processes needed to avoid tritium capture on DOG, VOG, and WOG streams.

5.1.5 Subcase 1e results

Subcase 1e examines the impact of tritium distribution between the cladding and the fuel meat. Also as part of this subcase, the recovery of Zr from the UNF cladding will also be examined. Here the tritium contained within the cladding is released into a separate stream as shown in Figure 1. The total tritium introduced into the plant remains the same, but the quantity contained in the fuel meat is reduced. This case will be compared to base case 1b-1 and 1b-4 as shown in Table 6.

Subcase 1e-1 assumes that 40% of the tritium is bound in the cladding. The remaining conditions are the same as for subcase 1b-1. Since 40% of the tritium is bound in the cladding and assumed to not be released to the gas streams, the effective tritium DF requirement is reduced to 60 to limit the tritium release to the stack to the same limit. However, as noted in Section 5.1.4, the reduction in DF from 100 to 60 translates to a very small difference in the amount of tritium-contaminated water that must be recovered: 22,195 mol/h for subcase 1b-1, and 22,046 mol/h for subcase 1e-1.

Subcase 1e-2 assumes the use of a Zr recovery process that also releases the tritium into a separate ZrROG stream. Assuming that a DF of 200 can be achieved on this stream, the effective DF for the remaining off-gas streams is increased to 75, and the total tritium-contaminated water recovery is increased slightly to 22,120 mol/h.

Subcase 1e-3 assumes the use of a Zr recovery process that also releases the tritium into a separate ZrROG stream but will use lower off-gas dew points for the DOG, VOG, and WOG. As with subcase 1e-2, assuming that a DF of 200 can be achieved on this stream, the effective DF for the remaining off-gas streams is increased to 75; due to the lowering of the dew points, the total tritium-contaminated water recovery is reduced to 5,675mol/h.

Table 3. Subcase 1d: Tritium distribution to the plant off-gas systems with tritium pretreatment

Case number	DOG dew point (ºC)	VOG dew point (ºC)	WOG dew point (ºC)	Tritium pretreatment efficiency (%)	% ^{3}H to DOG (%)	% ^{3}H to VOG (%)	% ^{3}H to WOG (%)	TPT off-gas DF	Tritium-contaminated water removed from TPT off-gas (mol/h)	Tritium-contaminated water recovered from stack (mol/h)	Total tritium-contaminated water recovery required (mol/h)	TPT off-gas tritium DF / off-gas DF required	Notes
1b-1	30	30	30	0.00%	8.70%	87.0%	4.3%		–	22,195.32	22,195.32	1/100	1,2
1b-4	-40	10	10	0.00%	0.14%	95.1%	4.8%		–	5,694.18	5,694.18	1/100	1,2
1d-1	30	30	30	100%	0.00%	0.0%	0.0%	100	0.4545		0.45	100/1	1,2
1d-2	30	30	30	99%	0.09%	0.9%	0.0%	100	0.4504	22,195.32	22,195.77	100/100	1,2
1d-3	30	30	30	99%	0.09%	0.9%	0.0%	200	0.9008	11,097.66	11,098.56	200/2	1,2
1d-4	-40	10	10	99%	0.00%	1.0%	0.0%	200	0.9008	2,847.09	2,847.99	200/2	1,2

Notes:

1. Recycle water/acid volume: 100,000 l
2. 100% of tritium is in fuel meat

Table 4. Tritium release fractions required to avoid the need for tritium capture from DOG, VOG, and WOG

Achievable tritium DF for TPTOG stream (Box 1)	Required tritium release fraction (%) (Box 2)	% of total tritium captured from TPTOG stream (%) (Stream 1)	% of total tritium released to stack from TPTOG stream (%) (Stream 2)	% of total tritium released to stack from combined DOG, VOG, and WOG streams (%) (Stream 3)
100	100.00%	99%	0.00%	0.00%
110	99.9083%	99%	0.9083%	0.0917%
125	99.7984%	99%	0.7984%	0.2016%
150	99.6664%	99%	0.6644%	0.3356%
200	99.4950%	99%	0.4975%	0.5025%

Notes:

1. Assumes overall tritium DF requirement of 100
2. Basis of percentage is total tritium in fuel
3. Assumes all tritium is in fuel meat
4. Box numbers or stream numbers tied to Figure 7

Table 5. Subcase 1e: Tritium distribution to the plant off-gas systems with zirconium recycle

Case number	DOG Dew Point (ºC)	VOG Dew Point (ºC)	WOG Dew Point (ºC)	Tritium in Cladding (%)	% H-3 to DOG (%)	% H-3 to VOG (%)	% H-3 to WOG (%)	ZrROG off-gas DF	Tritium-contaminated water removed from ZrROG (mol/h)	Tritium-contaminated water recovered from stack (mol/h)	Total tritium-contaminated water recovery required (mol/h)	Off-gas DF required	Notes
1b-1	30	30	30	0.00%	8.70%	87.0%	4.3%	NA	–	22,195.32	22,195.32	100	1,2
1b-4	-40	10	10	0.00%	0.14%	95.1%	4.8%	NA	–	5,694.18	5,694.18	100	1,2
1e-1	30	30	30	40%	5.22%	52.2%	2.6%	NA	NA	22,045.86	22,045.86	60	1,3
1e-2	30	30	30	40%	5.22%	52.2%	2.6%	200	0.3640	22,120.59	22,120.95	75	1,3,4
1e-3	-40	10	10	40%	0.08%	57.1%	2.9%	200	0.3640	5,675.00	5,675.37	75	1,3,4

Notes:

1. Recycle water/acid volume: 100,000 l
2. 100% of tritium is in fuel meat
3. 60% of tritium is in fuel meat
4. Zr Recycle releases 100% of tritium from cladding and has a tritium DF of 200 on off-gas stream

5.2 Case 2 – Split Acid/Water Recycle Systems

This case will be split into two subcases.

5.2.1 Subcase 2a results

Subcase 2a explores the impact of splitting the acid/water recycle into a high-tritium and low-tritium acid/water recycle system. As in previous cases, the tritium is assumed to accumulate within the plant until the tritium discharge rate to the off-gas streams equals the feed rate of tritium entering the plant in the UNF stream. A separate aqueous system is used for head-end, first-cycle solvent extraction and high-level waste solidification systems (i.e., fission product stabilization) as shown in Figure 8, and a second system is used for the balance of the plant. This limits the spread of tritium from head-end and first-cycle solvent extraction. The extent of the spread is impacted by the entrainment of aqueous streams between the first and second cycles of solvent extraction. In this evaluation the entrainment is nominally set at 0.1% between the first and second cycles. The overall tritium DF for the plant remains at 100. The volume of resulting tritium-contaminated water to grout for disposal is determined. It is assumed that the total volume of acid/water is split evenly between the two systems.

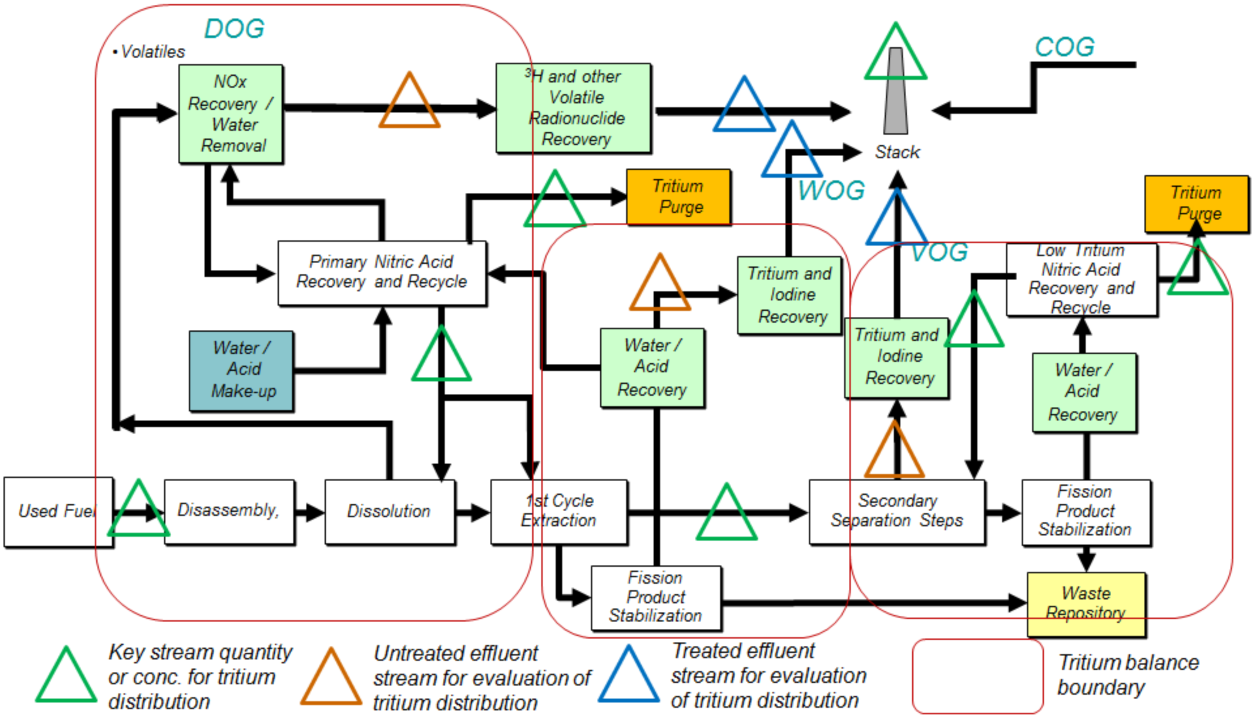

Figure 8. Block diagram showing implementation of tritium barrier and resulting high- and low-tritium acid/water recycle systems.

In subcase 2a-1, the tritium concentration in the high-tritium recycle system is significantly increased from that in the single recycle system case (1b-1) [1.55×10^{-4}% vs 2.00×10^{-5}%, respectively. There is also a significant shift in the tritium distribution to the off-gas streams due to the implementation of the tritium barrier, with the DOG and WOG accounting for 99.7% of the tritium compared with 13% in subcase 1b-1. With 99.7% of the tritium contained in the DOG and WOG, it is possible to achieve the desired overall tritium DF for the plant by only treating the DOG and WOG. To achieve the desired overall DF of 100, a DF of 146 would be required for the DOG and WOG. This reduces the total tritium-contaminated water recovery required from 22,195 mol/h if treating all off-gas streams to a DF of 100 down to 2904 mol/h for the DOG and WOG.

Subcase 2a-2 decreases the dew point on the DOG stream from 30 °C to 10 °C. As in subcase 1b, this increases the tritium concentration in the recycle acid/water streams and shifts the tritium discharge points from the DOG. The fraction of tritium in the DOG and WOG is still greater than 99% (99.4%), and an overall tritium DF of 100 can be achieved by increasing the tritium DF of the DOG and WOG to 251. This results in the need to recover and treat 1516 mol/h of tritium-contaminated water. Further reducing the DOG dew point to -40 °C (subcase 2a-3) continues this trend and forces 98.3% of the tritium into the WOG stream. If a tritium DF of 1491 can be achieved on the DOG and WOG, only 982 mol/h of tritium-contaminated water must be recovered and treated. This is a factor of 5 to 20 smaller than in base cases, 1b-1 and 1b-4. This is primarily the result of the much smaller volumetric flowrate of the DOG and WOG off-gases acting as carrier for water vapor having a higher concentration of tritiated water in it. The isolation of the VOG with its large volumetric flowrate carries water vapor with very tritium concentrations.

Subcase 2a-4 decreases the dew point of the VOG and WOG streams from 30 °C to 10 °C. In this case the tritium concentrations in the recycle acid/water streams are further increased, but >3% of the tritium is lost to the VOG stream, which will require treatment of all three off-gas streams. A total of 5694 mol/h of tritium-contaminated water would need to be recovered and treated in this subcase, which is the same as without the tritium barrier (subcase 1b-4). What is different is the tritium concentrations in the recycle acid/water and the distribution of the tritium between the various off-gas streams.

These results are summarized in Table 7.

5.2.2 Subcase 2b results

Subcase 2b adds the use of a tritium bleed stream to subcase 2a. In the bleed stream case, some fraction of the aqueous (water/acid) stream is removed from the system and replaced with fresh acid and water to make up for the water removed. The benefit in terms of the water removal will be compared with base cases 1b-1 and 1b-4. The results from this analysis are presented in Table 8.

Subcase 2b-1 is the same as subcase 2a-4, with the exception that a bleed stream of 1%/h of the 50,000 L in the tritium acid/water recycle system is added. The bleed stream removes 98.7% of the tritium. This lowers the DF required for the DOG+WOG to 1.35. The treatment of the DOG+WOG streams only recover 73.1 mol/h of tritium-contaminated water. This is added to the bleed stream for a total of 20,795 mol/h that must be treated.

Subcase 2b-2 reduces the tritium bleed from 1% to 0.1%. This reduces the total tritium recovery in the bleed to 87.8% and increases the required tritium DF for the DOG+WOG to 17. With that DF, a total of 264 mol/hr must be recovered from the off-gas stream, which is added to the 2072 mol/h in the bleed stream for a total of 2337 mol/h.

Subcase 2b-3 increases the dew point of the VOG and MOG from 10 °C to 30 °C. This decreases the tritium content in the recycle acid/water systems, reduces the amount of tritium removed in the tritium bleed to 67.7%, and increases the required tritium DF for the DOG+WOG to 44. With that DF, a total of 980 mol/hr must be recovered from the off-gas stream, which is added to the 2072 mol/h in the bleed stream for a total of 3033 mol/h.

Subcase 2b-4 increases the volume of the aqueous stream in each of the two acid/water recycle systems from 50,000 l to 500,000 l. Holding the bleed to 0.1% increases the volume removed as well as the fraction of tritium removed to 95.5%. The required tritium DF for the DOG+WOG is reduced to ~5. With that DF, a total of 770 mol/hr must be recovered from the off-gas stream, which is added to the 20,722 mol/h in the bleed stream for a total of 21,492 mol/h.

Subcase 2b-5 reduces the tritium bleed from 0.1% to 0.01%. This reduces the total tritium recovery in the bleed to 67.7% and increases the required tritium DF for the DOG+WOG to 44. Note that this is the same as for subcase 2b-3. With that DF, a total of 960 mol/hr must be recovered from the off-gas stream, which is added to the 2072 mol/h in the bleed stream for a total of 3033 mol/h.

Subcase 2b-6 examines the impact of increasing the entrainment between the high-tritium side of the tritium barrier to the low side from 0.1% to 1.0%. This increases the tritium concentration on the low side from 6.37×10^{-8}% to 6.21×10^{-7}% and only slight lowers the concentration on the high side. Again, assuming only a bleed from the high tritium side of the barrier, the bleed stream removes 66.1% of the tritium. But the increased tritium concentration in the low side results in more than 1% of the total tritium fed to the plant reporting to the VOG. A tritium DF of ~3 is required for the combined DOG+VOG+MOG streams. With that DF, a total of 19,873 mol/hr must be recovered from the off-gas stream, which is added to the 2072 mol/h in the bleed stream for a total of 21,946 mol/h.

Subcase 2b-7 examines the impact of higher off-gas dew points. This subcase allows comparison with subcases 1b-4 and 2b-5. Increasing the dew point of the DOG from -40 °C to 30 °C results in an increased portion of the tritium-contaminated water to reach the DOG tritium abatement system (38.9% vs 0.3%). The increase in tritium-contaminated water vapor in the DOG increases the total tritium-contaminated water recovery requirement from 960.31 mol/hr to 2882.20 mol/h. This is associated with a slight decrease in the tritiated water concentration in the recycle acid/water, from 1.48×10^{-4} to 9.09×10^{-5} percent.

Table 6. Subcase 2a: Tritium distribution to the plant off-gas systems with tritium barrier implemented and total water/acid losses to the stack

Case number	DOG dew point (ºC)	VOG dew point (ºC)	WOG dew point (ºC)	% ^{3}H to DOG (%)	% ^{3}H to VOG (%)	% ^{3}H to WOG (%)	Tritium concentration in high ^{3}H recycle (%)	Tritium Concentration in Low 3H recycle (%)	Off-gas DF required for DOG and MOG only	Total water recovery required (DOG+MOG) (mol/h)	Total water recovery required if all treated (mol/h)	Notes
1b-1	30	30	30	8.70%	87.0%	4.3%	2.0E-05	–	NA	NA	22,195.32	
1b-4	-40	10	10	0.14%	95.1%	4.8%	7.9E-05	–	NA	NA	5694.18	
2a-1	30	30	30	66.5%	0.3%	33.2%	1.55E-04	7.37E-08	146	2904.21	22,195.32	1,2
2a-2	10	30	30	35.7%	0.6%	63.7%	2.97E-04	1.41E-07	251	1515.75	20,806.86	1,2
2a-3	-40	30	30	0.8%	0.9%	98.3%	4.59E-04	2.18E-07	1491	981.92	20,273.04	1,2
2a-4	-40	10	10	2.7%	3.2%	94.1%	1.57E-03	2.65E-06	NA	NA	5694.18	1,2

Notes:

1. Recycle water/acid volume: 100,000 l—split 50/50 high- and low-tritium sides
2. Entrainment of 0.1% in first-cycle solvent extraction

5.3 Case 3 Results

Case 3 examines the selective removal of tritiated water (i.e., HTO) from the acid/water recycle streams assuming the use of a membrane. This case will be compared with subcases 1b-1 and 1b-4, and the results are shown in Table 9.

Subcases 3a-1 and 3a-2 use the same off-gas condenser conditions as subcases 1b-1 and 1b-4, respectively. It is assumed that the membrane reduces the tritium concentration in the acid/water recycle stream to a level such that tritium capture is not required for any of the off-gas streams. As shown in subcase 1b, lowering the dew point on the DOG, VOG, and WOG increases the tritium concentration in the acid/water recycle stream. This would imply that the tritium concentration in the recycle acid/water stream from $2.0\times10^{-5}\%$ to $2.0\times10^{-7}\%$ would be reduced for subcase 3a-1, and from $7.9\times10^{-5}\%$ to $7.9\times10^{-7}\%$ for subcase 3a-2. To achieve this, 0.004504 mol/h of tritium must be removed from the aqueous recycle streams per hour in both subcases. Assuming that the membrane can concentrate the effluent by a factor of 1000 over the concentration in the retentate, a total volume of tritium-contaminated water of 2220 l/h or 123,307 mol/h must be managed and solidified as tritium-contaminated waste for subcase 3a-1 and 31,634 mol/h for subcase 3a-2. This can be compared to the need to capture and manage 22,195 mol/h of tritium-contaminated water in subcase 1b-1 or 5694 mol/h of tritium-contaminated water in subcase 1b-4. See Table 9 for the comparison between this case and subcases 1b-1 and 1b-4. However, while these values are larger than those in some of the other subcases, the amount of tritium-contaminated water to be managed in the membrane case can be reduced if the concentration factor is increased; this may be possible through successive concentration stages. If a concentration factor of 100,000 were assumed, only 1233 mol/hr of tritium-contaminated water would require solidification.

Table 7. Subcase 2b: Tritium distribution to the plant off-gas systems with tritium barrier implemented with tritium bleed from high ^{3}H recycle and total water/acid losses to the stack

Case number	DOG dew point (ºC)	VOG dew point (ºC)	WOG dew point (ºC)	% ^{3}H removed in high ^{3}H bleed	% ^{3}H to DOG (%)	% ^{3}H to VOG (%)	% ^{3}H to WOG (%)	Tritium concentration in high ^{3}H recycle (%)	Tritium concentration in high ^{3}H recycle (%)	Off-gas DF required for only DOG and MOG	Total water recovery required (DOG + MOG) (mol/h)	Total water recovery required if all off-gas streams are treated (mol/h)	Total water recovery (off-gas + bleed) (mol/h)	Notes
1b-1	30	30	30		8.70%	87.0%	4.3%	2.0E-05	–	NA	NA	22,195.32	22,195.32	
1b-4	-40	10	10		0.14%	95.1%	4.8%	7.9E-05	–	NA	NA	5694.18	5694.18	
2b-1	-40	10	10	98.7%	0.0%	0.0%	1.3%	2.17E-05	7.65E-09	1.35	73.14	1485.137	20,795.00	1,2,3
2b-2	-40	10	10	87.8%	0.3%	0.3%	11.6%	1.93E-04	2.36E-07	17	264.42	5279.897	2337.00	1,2,4
2b-3	-40	30	30	67.7%	0.3%	0.3%	31.8%	1.48E-04	6.37E-08	44	960.31	19844.63	3033.00	1,2,4
2b-4	-40	30	30	95.5%	0.0%	0.0%	4.5%	2.10E-05	4.82E-09	5	769.93	15973.00	21,492.00	2,4,5
2b-5	-40	30	30	67.7%	0.3%	0.3%	31.8%	1.48E-04	6.37E-08	44	960.31	19844.63	3033.00	2,5,6
2b-6	-40	30	30	66.1%	0.2%	2.7%	31.0%	1.45E-04	6.21E-07	NA	NA	19873.36	21,946.00	5,6,7
2b-7	30	30	30	41.4%	38.9%	0.2%	19.5%	9.09E-05	3.90E-08	70	2,882.60	22036.84	4954.82	2,5,6

Notes:

1. Recycle water/acid volume: 100,000 l—split 50/50 high- and low-tritium sides
2. Entrainment of 0.1% in first-cycle solvent extraction
3. Tritium bleed from the high tritium content acid/water system of 1%
4. Tritium bleed from the high tritium content acid/water system of 0.1%
5. Recycle water/acid volume: 1,000,000 l—split 50/50 high- and low-tritium sides
6. Tritium bleed from the high tritium content acid/water system of 0.01%
7. Entrainment of 1.0% in first-cycle solvent extraction

Table 8. Subcase 3a: Tritium distribution to the plant off-gas systems and total tritium-contaminated water/acid losses to the stack with and without the use of a tritium selective membrane on the acid/water recycle system

Case number	DOG dew point (ºC)	VOG dew point (ºC)	WOG dew point (ºC)	Tritium purge rate (%/h)	Desired overall DF	% ^{3}H to DOG (%)	% ^{3}H to VOG (%)	% 3H to WOG (%)	Tritium concentration in recycle (%)	Total tritium-contaminated water loss to stack (mol/h)	Tritium-contaminated water recovered from stack (mol/h)	Tritium-contaminated water recovered via membrane (mol/h)	Total tritium-contaminated water recovery required (mol/h)
1b-1	30	30	30	0.00%	100	8.70%	87.0%	4.3%	2.0E-05	22,420	22,195.32	–	22,195.32
1b-4	-40	10	10	0.00%	100	0.14%	95.1%	4.8%	7.9E-05	5752	5694.18	–	5694.18
3a-1	30	30	30	0.00%	100	0.09%	0.9%	0.0%	2.0E-07	20,478	0	123,307.35	123,307.35
3a-2	-40	10	10	0.00%	100	0.00%	1.0%	0.0%	7.9E-07	5752	0	31,634.31	31,634.31

Reductions in the dew points of the off-gas streams will have the same effects on the amount of tritium-contaminated water that must be managed if a membrane system is used to control the tritium content of the acid/water recycle system.

6. Conclusions and Recommendations

Table 10 provides a comparison of the evaluation results for the subcases reported here; two different dew point conditions were evaluated for DOG, VOG, and WOG. At the 30/30/30 °C dew point, the lowest amount of tritium-contaminated water that must be managed is associated with the TPT subcase 1d-1 in which 100% of the tritium is released into the TPTOG stream. This resulted in ~0.5 mol/h of tritium-contaminated water that must be recovered and solidified. This is four orders of magnitude less than for all other cases examined. All other cases required the management/solidification of 5500 to 123,000 mol/h of tritium-contaminated water. Tritium concentrations in the acid/water recycle streams ranged from 2.0×10^{-7} to 1.55×10^{-5} mol%.

At the -40/10/10 ºC dew point, the lowest amount of tritium-contaminated water that must be managed would still be associated with the TPT subcase 1d-1 (a 30/30/30 ºC dew point case) in which 100% of the tritium is released into the TPTOG stream, as the dew points of the DOG, VOG, and WOG are irrelevant for this analysis since the tritium would not reach the aqueous systems. This again would result in ~0.5 mol/h of tritium-contaminated water that must be recovered and solidified. All other cases required the management/solidification of 2400 to 32,000 mol/h of tritium-contaminated water. The TPT case is again 3.5 to 4 orders of magnitude less. Tritium concentrations in the acid/water recycle streams ranged from 7.9×10^{-7} to 1.57×10^{-3} mol%.

The reader must keep in mind that these values are only reflective of the cases evaluated in this report and reflect the underlying assumptions that were used. One of the most significant assumptions is that of an overall plant DF requirement, in this case a DF of 100 was selected. The use of a lower overall DF should result in lower tritium-contaminated water recovery requirements and potentially fewer off-gas streams that would require tritium abatement.

In addition to that type of broad summary of the quantities of tritium-contaminated water that must be recovered and solidified, and the tritium concentrations within the plant, there are a number of specific trends and conclusions that can be drawn from the cases discussed in the Section 5.

In the case of no tritium barrier and no bleed streams:

- The bulk of the water losses to the stack are associated with the VOG stream, for which the volumetric flow rate is about 10 times larger than the sum of the other off-gas streams. This results in 87–95% of the tritium release occurring in the VOG stream. The VOG stream is the largest of the three off-gas streams and as a result would require the largest equipment.
- If the DF of 100 is required, treatment of the DOG, VOG, and WOG would be required.
    - DOG treatment could be avoided if the dew point was lowered to ~-40°C.
    - Up to ~400 kg/h of tritium-contaminated water would need to be removed from off-gas and solidified.

Table 9. Summary of total tritium-contaminated water/acid losses to the stack

Case number	DOG dew point (ºC)	VOG dew point (ºC)	MOG dew point (ºC)	Tritium purge rate (%/h)	Desired overall DF	% ^{3}H to DOG (%)	% ^{3}H to VOG (%)	% ^{3}H to WOG (%)	Tritium concentration in recycle (%)	Total tritium-contaminated water loss to stack (mol/h)	Tritium-contaminated water recovered from stack (mol/h)	Tritium-contaminated water recovered via purge, TPT, or membrane (mol/h)	Total tritium-contaminated water recovery required (mol/h)
1b-1	30	30	30	0.00%	100	8.70%	87.0%	4.3%	2.0E-05	22,420	22,195.32	–	22,195.32
1c-6	30	30	30	0.01%	100	7.34%	73.4%	3.7%	1.7E-05	22,420	22,153.88	4,144.00	26,298.32
1d-1	30	30	30	0.00%	100	0.00%	0.0%	0.0%	0.00	22,420	0.00	0.4545	0.45
1d-2	30	30	30	0.00%	100	0.09%	0.9%	0.0%	2.0E-07	22,420	22,195.32	0.4504	22,195.77
1d-3	30	30	30	0.00%	100	0.09%	0.9%	0.0%	2.0E-07	22,420	11,097.66	0.9008	11,098.56
1e-1	30	30	30	0.00%	100	5.22%	52.2%	2.6%	1.2E-05	22,420	22,045.86	NA	22,045.86
1e-2	30	30	30	0.00%	100	5.22%	52.2%	2.6%	1.2E-05	22,420	22,120.59	0.3640	22,120.95
2a-1	30	30	30	0.00%	100	66.5%	0.3%	33.2%	1.55E-04	22,420	22195.32	NA	22,195.32
2b-7	30	30	30	0.01%	100	38.9%	0.2%	19.5%	9.09E-05	22,420	2882.60	2072.22	4954.82
3a-1	30	30	30	0.00%	100	0.09%	0.9%	0.0%	2.0E-07	20,478	0.00	123,307.35	123,307.35
1b-4	-40	10	10	0.00%	100	0.14%	95.1%	4.8%	7.9E-05		5694.18	–	5694.18
1c-1	-40	10	10	1.00%	100	0.02%	11.6%	0.6%	9.6E-06		5279.73	41,444.00	46,724.18
1c-2	-40	10	10	0.10%	100	0.08%	55.3%	2.8%	4.6E-05		5652.73	4,144.00	9797.18
1d-4	-40	10	10	0.00%	100	0.00%	1.0%	0.0%	7.9E-07	5752	2847.09	0.9008	2847.99
1e-3	-40	10	10	0.00%	100	0.08%	57.1%	2.9%	4.7E-05		5675.00	0.3640	5675.37
2a-4	-40	10	10	0.00%	100	2.7%	3.2%	94.1%	1.57E-03	5752	5694.18	–	5694.18
2b-1	-40	10	10	1.00%	100	0.0%	0.0%	1.3%	2.17E-05	5752	73.14	20722.22	20,795.00
2b-2	-40	10	10	0.10%	100	0.3%	0.3%	11.6%	1.93E-04	5752	264.42	2072.222	2337.00
3a-2	-40	10	10	0.00%	100	0.00%	1.0%	0.0%	7.9E-07	5752	0	31,634.31	31,634.31

- Lowering the dew point of the VOG and WOG decreases water loss but increases aqueous tritium concentration in the acid/water recycle system.
    - Tritium concentration in the acid/water system ranges from ~2×10^{-5} to 8×10^{-5}mol%.
- If tritium pretreatment is implemented and 100% of the tritium is released this significantly reduces the tritium-contaminated water that must be managed (i.e., 0.01 kg/h).
    - TPT has the potential to reduce the tritium-contaminated water waste volume by 3 to 4 orders of magnitude.
    - If an overall tritium DF of 100 is needed and the TPT off-gas can achieve DFs > than 100, then fractional release of tritium from the fuel meat can be slightly less than 100%. For example, release fractions of 99.5 to 99.9% require TPTOG DFs of 200 and 112, respectively, to achieve an overall DF of 100. (This would then avoid the need for tritium management on the other gas streams.
    - Lower required overall plant DFs will also lower the required release accordingly. For example if the required overall DF is only 10 then the release fraction needed is only ~90.5% to 90.9%.
- If the tritium remains bound in the cladding and Zr recycle is not implemented the DF on the off-gas streams can be reduced, e.g., if 40% is assumed in the cladding then if an overall plant of 100 is needed, the DF for the off-gas streams can be reduced to 60.
- If Zr Recycle is implemented within the reprocessing facility and the DF on ZrROG is greater than the facility DF, the DF on other streams can be reduced slightly.
    - However, reducing the overall DF from 100 to 50 only changes the amount of water/% tritium that must be captured from 99% to 98%, and thus there is no significant reduction in overall tritium-contaminated water management issue.

If a tritium barrier is implemented with no bleed stream:

- Significant water losses still arise from the VOG stream.
    - Tritium losses are much lower (<0.3% to 4%), and in some cases tritium abatement can be avoided on the VOG stream.
    - Depending on the overall plant tritium DF required, the VOG may still require treatment.
- Depending on dew points, tritium losses in the DOG range from <1% to 67%, in with balance with the WOG.
- If a plant-wide DF of 100 is required, treatment of the DOG and WOG would be required.
    - Approximately 5.1 to 53 kg/hr of water would need to be removed from off-gas and solidified tritium-contaminated water, depending on the dew points involved.
- Lowering the VOG and WOG dew point decreases water loss but increases aqueous tritium concentration.

- Tritium concentrations in head-end acid/water recycle systems are higher than in the case of a single acid/water recycle system (Case 1).
    - Tritium concentration in head-end acid/water recycle systems is ~1.6×10^{-4} to 2×10^{-3} mol%.
    - Tritium concentration in back-end acid/water recycle systems is ~7.4×10^{-8} to 3×10^{-6} mol%.

If a tritium bleed system is implemented:

- One percent of the aqueous inventory per hour bleed stream is insufficient to reduce the tritium concentration in the VOG such that it would not require treatment for Case 1 conditions.
    - Significant water losses still arise from the VOG stream.
    - Total volume of water to be solidified (i.e., bleed plus VOG and WOG) is 150% of a non-bleed case.
    - Tritium concentrations in aqueous streams are reduced.
- For Case 2 (i.e., tritium barrier), the tritium concentration in the WOG was reduced significantly, but not enough to preclude the need for treatment of the WOG.
    - A significant increase in water to be solidified was observed in most cases.
    - Tritium concentrations in aqueous streams are reduced.
    - Higher aqueous entrainment in first-cycle solvent extraction results in high tritium concentrations in back-end aqueous systems.

Tritium abatement can be avoided by applying a selective tritium removal system on the acid/water recycle system. However, this system must reduce the steady- state tritium concentration by a factor of 100 to achieve a plant tritium DF of 100. If a concentration factor of 1000 is achieved, the total amount of recovered tritium-contaminated water that must be managed is higher by a factor of ~5 than for the case of tritium abatement on the off-gas streams under comparable condition. However, this can be significantly reduced if the membranes could achieve higher concentration factors.

It should also be mentioned that while a target tritium DF of 100 is used for planning purposes in this analysis, the exact DF value depends on site-specific reprocessing facility parameters and, of course, fuel age. Variations in these conditions could significantly change needed DFs for tritium and other gaseous radionuclides and could change the results of this study.

From this analysis, there are two aspects that warrant further development and verification.

1. Demonstration that TPT can effectively remove >99.5% of the tritium from the fuel meat. This has the potential to eliminate the need for tritium management on the other off-gas streams, significantly reducing the amount of tritium-contaminated water that must be solidified. This appears to be achievable based on data reported by Goode (1980) but needs to be verified.

2. Development of a tritium selective membrane capable of maintaining tritium concentration $<1.0\times10^{-7}\%$ tritium concentration and capable of tritium concentration factors >10,000.

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