Modeling Tritium Lifecycle in Nuclear Plants

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ABSTRACT

The mathematical development of a tritium model for nuclear power plants is presented. The model requires that the water and tritium material balance be satisfied throughout normal operations and shutdown. The model results obtained at the time of publishing include the system definitions and comparison of the model predictions of tritium generations compared to the observed plant data of the Braidwood station. A scenario that models using ion exchange resin to remove coolant boron demonstrates the tritium concentration levels are manageable.

INTRODUCTION

The nuclear industry has requested research that develops a more detailed understanding of the tritium release pathways. EPRI has responded to the industry request to understand and define the tritium lifecycle from production to release by developing a tritium management model. The tritium lifecycle has been described in an EPRI report [1]; this paper presents the mathematical development for a practical tritium management model.

MODEL GOALS

The goals of the tritium model are below:

- Consider all significant tritium sources and their eventual in-plant retention and release paths.
- The tritium mass balance must be satisfied.

- Typical plant water management operations for both PWRs and BWRs will be considered including gaseous release, liquid effluents and zero liquid volume release strategies.
- Develop a predictive tool to model how in-plant and effluent tritium will respond to decisions related to water management or core design (production).

MODEL DEVELOPMENT

The foundation of the model is the standard chemical engineering definition of the mass balance. The mass balances are applied to both water and tritium.

 \sum Inputs – \sum Outputs ± Generation = Accumulation (Eq. 1)

The control volume is defined as the plant reactor coolant volume plus the static containment/drywell airspace. Each of these terms is developed as a rate with units in pCi/sec; however, inventory is stored as mass. Equation 1 is applied repeatedly to the total system volume and the individual unit operations for both liquid volume and tritium.

Total Coolant Tritium Balance

The tritium mass balance for the coolant is defined below in **Table I**.

Inputs	Outputs	Generation	Accumulation
Make-up	Monitored liquid effluent	Production	Total in-plant
water			inventory
	Monitored gaseous effluent	Tritium decay	
	Unidentified, unmonitored	Tritium	
	effluents	burnout	

Table I. Total Coolant Tritium Mass Balance Terms

Inputs

The input to the primary coolant is the make-up water, which is typically assumed to have no influent tritium. However, there are two known cases where the zero influent assumption is invalid: 1) plants that recycle liquid processing effluent - tritium is re-injected into the makeup volume, 2) rare situations where a plant is downstream of a river from another plant, and uses purified make-up water from that source.

Outputs

There are three effluent streams considered: the gaseous release, the liquid release, and unknown effluents.

• The liquid effluent is perhaps the best characterized because there are relatively accurate assessments of the volumes of liquid radwaste with known concentrations of tritium.

- The gaseous effluent, although measurable, is more difficult to quantify because of the variations in stack flow, sampling, and analytical techniques.
- Unidentified, unmonitored releases are quantified as liquid or gaseous releases to the atmosphere and/or groundwater or through other unmonitored pathways.

Generation

Tritium is assumed to be generated in the core. PWR and BWR generation term descriptions differ; however, they are similar in form.

Tritium has a half life of approximately 12 years. For this model, it is assumed that the decay of tritium is negligible.

For a PWR, the generation is divided into five different sources according to a Westinghouse source term reference [2] for a four-loop, 4100 MWt plant.

Primary chemistry data were provided by Exelon for Braidwood Unit 1, Cycle 11. Assuming concentration dependent generation terms for boron and lithium and constant generation for other sources (Fig. 1), the data were used to estimate the following generation rate constants, see Table II (deuterium was neglected).



Fig. 1. Tritium production by source —percentage basis

NOTE: These values do not necessarily apply generically to any PWR plant; they are a function of many parameters including core design, chemistry, and fuel performance.

Table II. Estimated Generation Rates for Tritium using Braidwood Unit 1 Cycle 11 Chemistry Data

Tritium Generation Rates	Rate Constant
Boron Rate (Ci/ppm/min)	1.45E-06
Lithium Rate (Ci/ppm/min)	1.30E-04
Fission Rate (Ci/min)	3.00E-04
IFBA Rate (Ci/min)	5.00E-05

Accumulation

The time dependent accumulation terms are the most complicated section of the model. The total system accumulation is defined as the sum of the individual unit operation accumulations. These terms are defined the following sections.

Unit Operations/Inventories

An example release path for tritium in a PWR is shown below in Fig. 2.

Not only must the total system mass balance be satisfied, but also the individual unit operation mass balance must be satisfied. This allows the development of a system of accumulation equations that may be solved simultaneously. The accumulation equations are derived below in Table III.

Note that the generation terms are not listed because they are zero except for the RCS system.

In the equations, the following variables are used. Subscripts will denote input, *i*, output, *o*, or the system name, e.g. RCS for reactor coolant system.

F = Water flow rate (L/s) C = Concentration (μ Ci/L) V = Volume (L)

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Fig. 2. Example schematic of tritium flow path for a PWR

Volume is calculated using the water mass and density at the given temperature. The specific gravity (SG) is found from the following correlation.

$$SG = \left[\frac{1 + 0.134248SS_2 - 3946263x10^{-3}SS_1}{3.1975 - 0.3151548SS_2 - 1.203374x10^{-3}SS_1 + 7.48908x10^{-13}SS_1^4}\right]^{1/2}$$
(Eq. 2)

Where

 $SS_1=374.11$ -T $SS_2=SS_1^{1/3}$ T = temperature in Degrees Celsius

Unit Operation	Inputs	Outputs	Accumulation Equation
Reactor Coolant System (RCS)	Primary Water Storage Tank (PWST) Refuel Water Storage Tank (RWST) Boric Acid Storage Tank (BAST)	Letdown/Recycle Hold-up Tank (RHUT) Spent Fuel Pool (SFP) Refuel Water Storage Tank (RWST)	$\frac{\Delta (V_{RCS}C_{RCS})}{\Delta t} = F_{PWST}C_{PWST} + F_{RWST,i}C_{RWST} + F_{BAST}C_{BAST} - F_{RWST,o}C_{RCS} - F_{SFP,o}C_{RCS} - F_{RHUT}C_{RCS} + R_{H_{3}Gen}$
Primary Water Storage Tank (PWST)	Make-up System (MU) Recycle Monitor Tank	RCS	$\frac{\Delta (V_{PWST}C_{PWST})}{\Delta t} = F_{MU}C_{MU} + F_{RHUT}C_{RHUT} - F_{RCS}C_{PWST}$
Recycle Hold- up Tank (RHUT)	RCS	Liquid Radwaste (LRW), PWST (after processing),	$\frac{\Delta (V_{RHUT}C_{RHUT})}{\Delta t} = F_{RCS}C_{RCS} - F_{LRW}C_{RHUT} - F_{PWST}C_{RHUT}$
Spent Fuel Pool (SFP)	RCS (outages) PWST Manual water transfers	SFP HVAC, RWST (outages) Manual transfers to RHUT	$\frac{\Delta (V_{SFP}C_{SFP})}{\Delta t} = F_{RCS}C_{RCS} - F_{HVAC}C_{SFP} - F_{PWST}C_{SFP}$
Refuel Water Storage Tank (RWST)	SFP (outages) Blender	RCS, SFP (outages)	$\frac{\Delta (V_{RWST}C_{RWST})}{\Delta t} = F_{SFP}C_{SFP} - F_{RCS}C_{RWST}$
Spent Fuel Pool HVAC	SFP	Monitored Gas Release (MGR)	$\frac{\Delta (V_{HVAC}C_{HVAC})}{\Delta t} = F_{SFP}C_{SFP} - F_{MGR}C_{HVAC}$
Liquid Radwaste (LRW)	RHUT Floor Drains Equipment Drains	Recycle Monitor Tank Monitored Liquid Release (MLR)	$\frac{\Delta (V_{LRW}C_{LRW})}{\Delta t} = F_{RHUT}C_{RHUT} - F_{MLR}C_{LRW}$
Containment	Leaks and evaporation inside containment	Containment Purge Monitored unit vent pathway	Steam leaks and evaporation in containment – purge releases

 Table III. Unit Operation Mass Balances for Tritium Model

The mass balances are written generally. During various phases of the cycle, there are terms that will be zero, or flow rates and concentrations that will remain constant. In most situations, the

exit stream concentrations (those with a negative sign) will equal the concentration of the inventory volume. This assumes the inventory is perfectly mixed, which is consistent to Constant Stirred-Tank Reactor material balance definitions.

The expansion of the equations for application in a spreadsheet is relatively straightforward. First, the water balance is solved for the unit operation. Using the primary water storage tank as an example, let $V_{PWST,1}$ be the initial volume of the PWST before an operation and $V_{PWST,2}$ be the final volume after an operation. The final volume is solved by applying the water balance

$$\frac{\Delta(V_{PWST})}{\Delta t} = \frac{V_{PWST,2} - V_{PWST,1}}{\Delta t} = F_{MU} + F_{RHUT} - F_{RCS}$$
(Eq. 3)

$$V_{PWST,2} = V_{PWST,1} + \Delta t \left(F_{MU} + F_{RHUT} - F_{RCS} \right)$$
(Eq. 4)

Next, the concentration is solved by using the final volume calculated above.

$$\frac{\Delta(V_{PWST}C_{PWST})}{\Delta t} = (Eq. 5)$$

$$\frac{V_{PWST,2}C_{PWST,2} - V_{PWST,1}C_{PWST,1}}{\Delta t} = F_{MU}C_{MU} + F_{RHUT}C_{RHUT} - F_{RCS}C_{PWST,1}$$

$$C_{PWST,2} = \frac{V_{PWST,1}C_{PWST,1} + \Delta t (F_{MU}C_{MU} + F_{RHUT}C_{RHUT} - F_{RCS}C_{PWST,1})}{V_{PWST,2}} (Eq. 6)$$

Spent Fuel Pool Evaporation

Several methods for calculating the evaporation rate based on the dry-bulb temperature, wet-bulb temperature, pool temperature and air velocity are available. The following equations are taken from several references; most notably a summary website [3]. These equations are used for the model's gaseous effluent determination. An option is provided for bulk SFP gaseous contributions in lieu of data collection for evaporation calculations.

The mass of water evaporated is found from the following equation;

$$\mathbf{Q}_{\mathrm{m}} = \mathbf{A} \left(\mathbf{x}_{1} - \mathbf{x}_{2} \right) \mathbf{a}_{\mathrm{e}} \tag{Eq. 7}$$

where,

 Q_m = mass of water evaporated (kg/s) A = the surface area (m²) x_1 = the specific humidity directly above the pool (kg H₂O/kg air) x_2 = the specific humidity of the room (kg H₂O/kg air) a_e = the evaporation constant (kg/m²s)

The evaporation constant is dependent on the velocity of the air above the pool.

$$a_{e} = \frac{(25 + 19_{V})}{3600}$$
(Eq. 8)

where,

v = air velocity (m/s)

The absolute humidity is typically found from a psychometric chart; however, these charts cannot be applied in a computer algorithm. Instead, the absolute humidity is calculated by using the relationships between saturation pressure and relative humidity.

The relative humidity, R_h , is defined as the ratio of the partial pressure of water (P_{steam}) and the saturation pressure of water (P_{sat}) at a given temperature.

$$R_{h} = \frac{P_{steam}}{P_{sat}}$$
(Eq. 9)

Both the partial pressure of water and the saturation pressure are related to the absolute humidity using the relations below.

$$P_{\text{steam}} = \left(\frac{P_{\text{atm}}}{P_{\text{sat}}}\right) \mathbf{x}$$
(Eq. 10)
$$P_{\text{sat}} = \left(\frac{R_{\text{air}}}{R_{\text{steam}}} + \mathbf{x}\right)$$
(Eq. 11)

 R_{air} and R_{steam} are the specific gas constants for air and steam; the ratio is a constant at 0.622. Inserting these definitions into the relative humidity expression yields,

$$R_{h} = \frac{P_{steam}}{P_{sat}} = \frac{\left(\frac{P_{atm}}{P_{sat}}\right)x}{\left(0.622 + x\right)}$$
(Eq. 12)

Solving for x gives an expression that requires the saturation pressure and relative humidity.

$$x = \frac{\frac{0.622R_{h}}{P_{atm}} - R_{h}}{P_{sat}}$$
(Eq. 13)

The saturation pressure can be found from the following correlation.

$$P_{\text{sat}} = 610.710701 + 44.4293573T + 1.41696846T^2 + 0.0274759545T^3 + 2.61145937e - 4T^4 + 2.85993708E - 6T^5$$
(Eq. 14)

where P_{sat} is in Pascals.

The relative humidity of the air directly above the pool is assumed to be 100%, and the ambient relative humidity from the local weather can be used for the relative humidity. The saturation pressure of the pool surface is found from the temperature of the surface water of the pool. The temperature of the surface water is slightly cooler than the bulk temperature because of energy loss due to evaporation. The following simple correlation can be used to estimate the pool surface temperature

 $T_{surface} = T_{pool} - \frac{\left(T_{pool} - T_{wetbulb}\right)}{8}$ (Eq. 15)

Other methods are quickly derived for using the wet-bulb temperature to calculate the absolute humidity if needed.

TRANSIENT MASS BALANCE MODELING

The flow rates and concentrations variables in the equations of Table I will be determined by the different operations that occur in a plant during the cycle. The operations are divided by their time in a cycle. For PWR's, the cycles are as follows:

- 1. Full power operations without boron removal (generation only).
- 2. Full power operations with boron removal via dilution (RCS to RHUT or LRW).
- 3. Shutdown drain to mid-loop (RCS to RHUT or LRW).
- 4. Cavity flood-up (RWST to Cavity to RCS).
- 5. Fuel transfer (RCS to Cavity to SFP)
- 6. Cavity drain down (Cavity (RCS) to RWST and LRW)
- 7. Refill and begin normal operations (RHUT and/or PWST to RCS.

A separate, but related, cycle plan will be developed for BWRs.

Each of these operations will have volumes of water with tritium concentrations determined by the previous step. A cumulative water mass/tritium mass balance will be maintained for each inventory. For recycle cases, the effluent tritium will be re-injected into the RCS.

MODEL VALIDATION AND VERIFICATION

The model is currently being developed in Microsoft Excel. After completion, the first task will be to model the tritium pathway of a plant. The proposed procedure for modeling the plant is as follows:

1. Determine the initial tritium concentration in all inventory locations at the start of a cycle.

- 2. Perform model calculations using only the estimated tritium production rates and the water management program.
- 3. Compare model results to effluent releases after the water management calculations and determine where the discrepancies occurred.
- 4. If justifiable, change water management actions or concentration data, otherwise, assign tritium to 'unaccounted' or 'lost.'
- 5. Repeat steps 1-4 until reasonable agreement is achieved.

'Justifiable' changes are those where it is possible for error to be introduced in the measurement of either concentration or flow. The purpose of the strategy is to avoid adjusting the mathematics to match the data in the middle of the calculation.

MODEL IMPLEMENTATION

The first draft of the model is being developed in a spreadsheet application that uses macros when necessary to perform transient calculations. The first station to apply the model is the Exelon Station of Braidwood Station in Braidwood, Illinois.

Braidwood station has two four-loop Westinghouse pressurized water reactors, each generating approximately 1300 MW. Exelon provided information about all unit operations necessary for RCS coolant modeling, such as system volume, temperature, initial tritium concentrations, and boron, lithium and tritium concentrations throughout the length of the cycle. In addition, Exelon provided an exhaustive list of potential transport operations and known volumes of water transferred between the unit operations.

At the time of publication of this paper, the modeling effort had progressed to the point of establishing the connections between the unit operations, and a reasonable estimate of the tritium generation was developed. Refer to Fig. 3 below.

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Fig. 3. Comparison of predicted and measured tritium concentrations for Braidwood Unit 1, Cycle 11

From May 2003 to December 2003, very little dilution water is used and the linear increase represents the combined generation as a function of boron concentration, lithium concentration, fission rate, and IFBA release. Not shown in this paper is that the same generation rate constants also provided very similar results for Unit 1, Cycle 10, as well as Unit 2, Cycles 10 and 11. On December 24th, make-up water was added to the RCS to dilute the boron concentration to reduce activity suppression. The agreement achieved was accomplished with a regular schedule of dilutions ranging from 1,000 gallons (3,800 L) to 30,000 gallons (113,550 L).

One possible scenario to manage tritium release is to allow it to concentrate in the RCS. One method to achieve this goal is boron removal with ion exchange resin instead of dilution at the later stages of the cycle. In effect, this removes the dilution component, but the boron concentrations are held the same as the dilution case. This scenario was modeled, and the results are shown below in Fig. 4.

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for Braidwood Unit 1, Cycle 11

The tritium concentration positive slope is less than the initial slope because of the lower boron concentration later in the cycle. The tritium concentration increases; however, it does not increase to levels higher than that initially observed before dilutions began.

CONCLUSIONS

The tritium management model is in development. The model requires the satisfaction of the water and tritium mass balance. Programming is in progress, and results for tritium generation show reasonable agreement between measured and predicted tritium concentrations. The operation scenario of using ion exchange resin to remove boron instead of dilution make-up water shows an increase of tritium concentrations that is within known plant operating experience.

REFERENCES

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- 2. Bryant, Daniel, South Texas Project, Personal Communication, 2005
- 3. Kuemmel, Bernd, 'The _Temp, Humidity, and Dew Point_ Often Needed Answers', http://mmf.ruc.dk/~bek/relhum.htm, 2005