COMPARISON OF SIMULATED RADIOIODINE RATIOS WITH MEASURED VALUES FOR THREE CANDU STATIONS

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Abstract

A MATLAB model has been used to predict steady-state radioiodine concentrations for ^{133}I, ^{134}I and ^{135}I relative to ^{132}I in the coolant of three CANDU®† stations. The radioiodines, originated as fission products from tramp uranium, were directly injected into the coolant where they were subjected to radioactive decay, plate out and purification. The results of simulations were compared to long-term measured values and purification parameters in the simulations were adjusted until measured and model values agreed. The purification flow rate is the critical variable and operating purification flow rates were deduced for three stations.

1. Introduction

The heat transport purification system is used to remove contaminants and control the coolant pH. The purification system is very effective at removing nongaseous fission products, such as radioiodine, from the coolant. These fission products can be introduced into the HTS from failed fuel or tramp uranium in the core. The successful removal of radioactive impurities from the coolant and the maintenance of good coolant chemistry is one measure of the performance of the purification system [1]. The purification flow rate varies in different CANDU reactors (typically between a few kg/s and 20-30 kg/s) and affects the concentrations of radionuclides in the coolant. In this paper a dynamic model to predict the concentrations of fission products in the coolant of a CANDU-6 station [2] was adapted to estimate the purification flow rates for three CANDU plants.

2. Simulink Model

A Simulink/MATLAB [+] model was used to predict steady-state radioiodine concentrations for ^{133}I, ^{134}I and ^{135}I relative to ^{132}I in the coolant of CANDU®† stations from tramp uranium on the inner sides of the pressure tubes (from historical fuel defects) and on the surface of the fuel bundles. The model takes account of refuelling, which constantly introduces fresh natural uranium contamination via new fuel bundles while removing the irradiated uranium. The principal nuclear reactions in the tramp uranium were simulated through a large set of coupled differential equations that were solved numerically. The Simulink programming environment allows these equations, which describe the fission process,

† CANDU® (CANada Deuterium Uranium) is a registered trademark of Atomic Energy of Canada Limited, AECL.

+ Simulink® is an environment for multidomain simulation and Model-Based Design for dynamic and embedded systems. It provides an interactive graphical environment and a customizable set of block libraries that let you design, simulate, implement, and test a variety of time-varying systems (www.MathWork.com)

capture and decay processes, purification and deposition, to be modelled using combinations of the Integrator block, the Gain block, the Sum block and the Product block defined in the Simulink library.*

2.1 Half life of Purification System

It was initially assumed that the operational purification flow rate is the same value as was achieved during commissioning. The flow rate of the purification system varies for different plants but for the CANDU 6 reactor it is nominally 24 kg/s for a circulating inventory of about 120 Mg. The purification removal constant, lambda purification is defined as follows:

$$\text{Lambda Purification} = \frac{Efficiency \times Purification\ Flow\ Rate}{Circulating\ Mass} \qquad (1)$$

If it is assumed that the purification system has an efficiency of unity♦, lambda purification for a CANDU-6 is equal to:

$$\frac{1 \times 24(kg/s)}{120000(kg)} = 2 \times 10^{-4}\ s^{-1}$$

Thus it is possible to define a half-life for the purification system through the following relationship.

$$T_{\left(\frac{1}{2}\right)} = \frac{\ln(2)}{Lambda} = \frac{\ln(2)}{2 \times 10^{-4}} s = 3466s = 57.8\,\text{min}$$

So, in a CANDU-6, the purification system has a half-life on the order of one hour.

2.2 Efficiency of Purification system

The next step is to consider the efficiency of the purification system. The HTS purification system is composed of a particulate filter in series with two ion exchange columns. The ion exchange columns will remove dissolved ionic species. The purification system does not remove gaseous species.

Fission products are not typically in particulate form but will exist in gaseous form (as is the case for Xe, Kr and some iodine species), or in solution as dissolved aerosols (as is the case for Cs, Te and some iodine species). As expected, the removal rate by the purification system varies for different radionuclides driven by their chemical speciation. The ion-exchange column efficiency depends on the temperature and pH of the coolant. Generally, the performance of the ion exchange resins improves at lower temperatures.

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* The following differential equation was solved to simulate the number of atoms of a given species in the coolant is as follows: $\frac{dN_i(t)}{dt} = P_i - R_i$ $N_i(t)$: number of atoms of the i^{th} radioisotope in coolant ,P_i: Production Rate (atoms.s^{-1}) and R_i: removal rate due to decay, purification system, plate out or off gassing (atoms.s^{-1})

♦ The efficiency of purification system in removing different radioiodine was modeled as a separate gain. This is discussed in detail in section 2.2.

The efficiency of the purification system has been estimated based on factors for PWRs [2]. In CANDU reactors, the coolant temperature is reduced before it flows through the purification system by channelling the flow through heat exchangers. The PWR factors were selected for the modelling, because the purification system in PWRs copes with changes in temperature and boric acid concentrations and because the available information on CANDU semi-volatile fission product purification efficiencies is meagre. Table 1, shows the fraction of materials being removed by purification system.

Description	Elements		
Fraction of material removed in passing through purification system	Iodine	Tellurium	Caesium
	0.99	0.98	0.95

Table 1: Reference Purification System Efficiency Based on PWRs

The efficiency of purification system in a CANDU reactor was also estimated for radioiodine by studying the concentration measurements in loop1 and loop2 of the HTS of a CANDU-6 before and after passing through the purification system. Table 2 shows the efficiency of purification system in removing radioiodine based on measurements for a CANDU-6.

Radioiodine	Purification System Efficiency
I 132	94%
I 133	91%
I 134	95%
I 135	95%

Table 2: Purification System Efficiency Based on Measurements

Based on Tables 1 and 2, it is assumed here that the purification system has an average removal efficiency of 95% for radioiodine species. Figures 1 to 4 show the measured concentrations of ^{132}I, ^{133}I, ^{134}I and ^{135}I in the coolant before and after passing the purification system as a function of time. The measurements were recorded weekly for about a year in a CANDU-6 reactor.

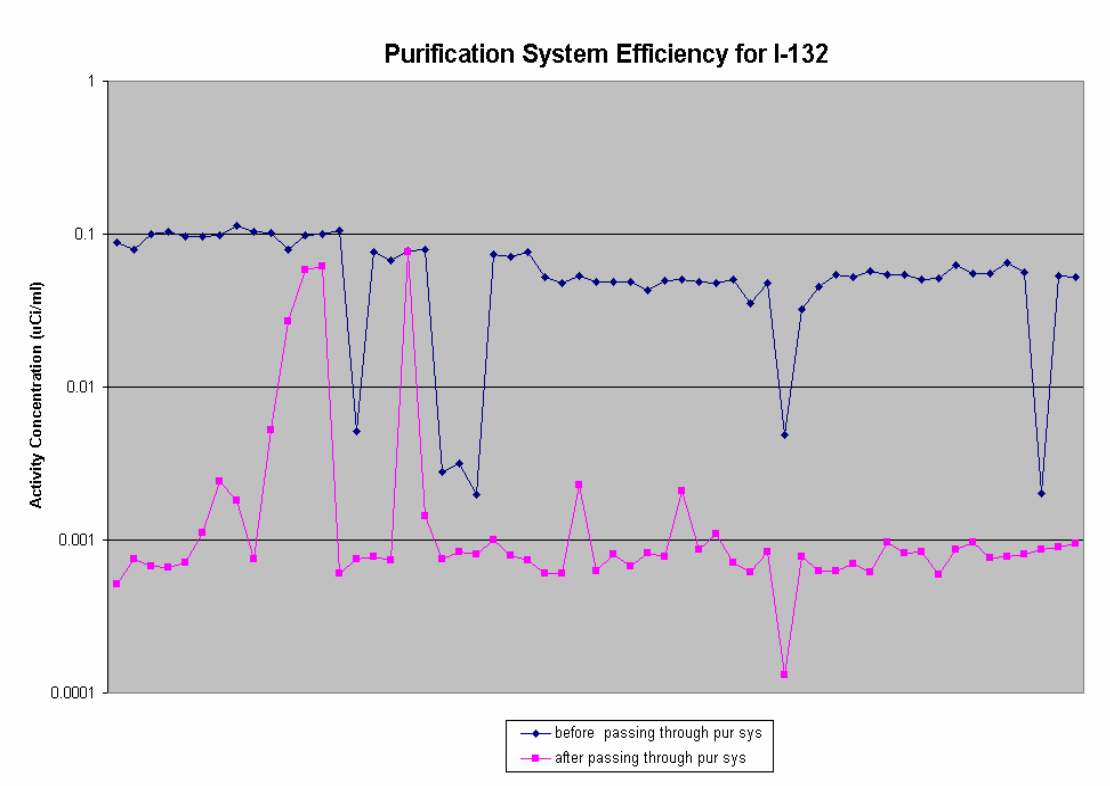

Figure 1: Measured Concentration of ^{132}I after and before the Purification System

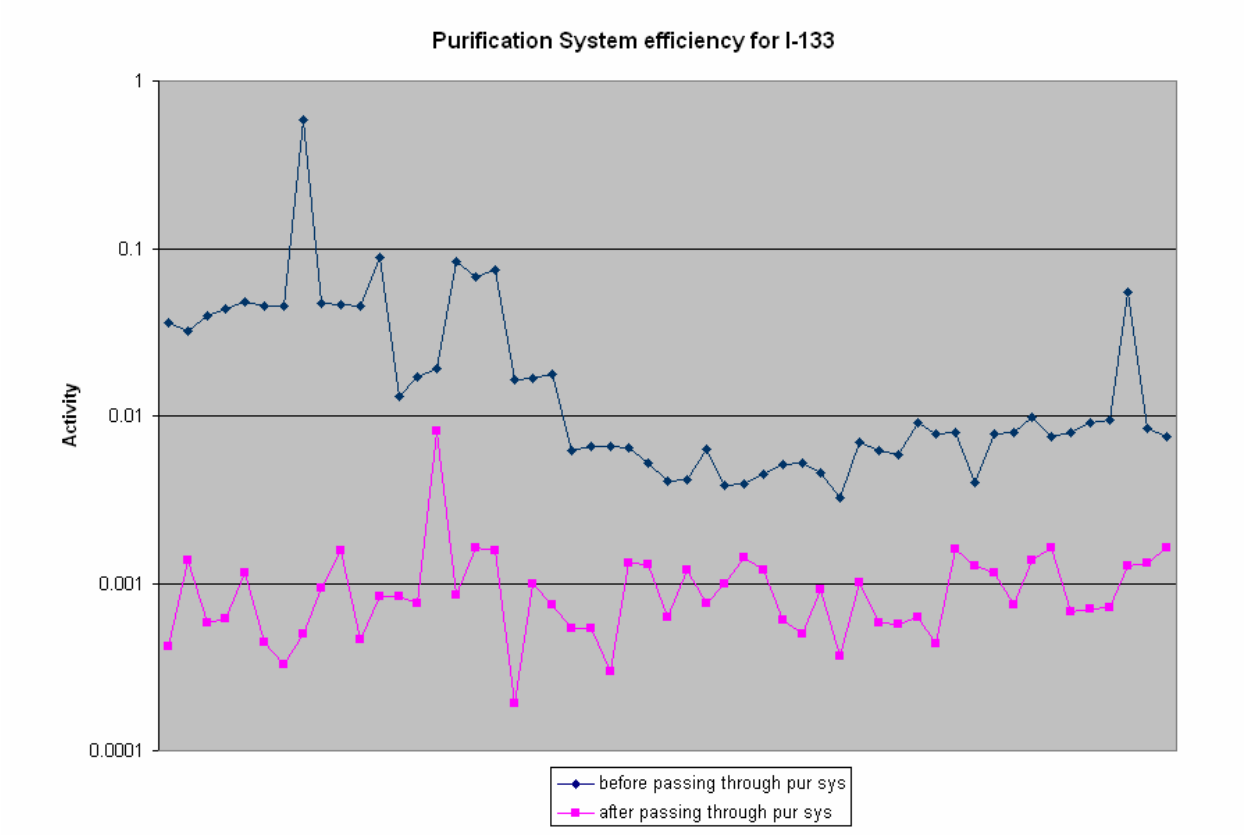

Figure 2: Measured Concentration of ^{133}I after and before the Purification System

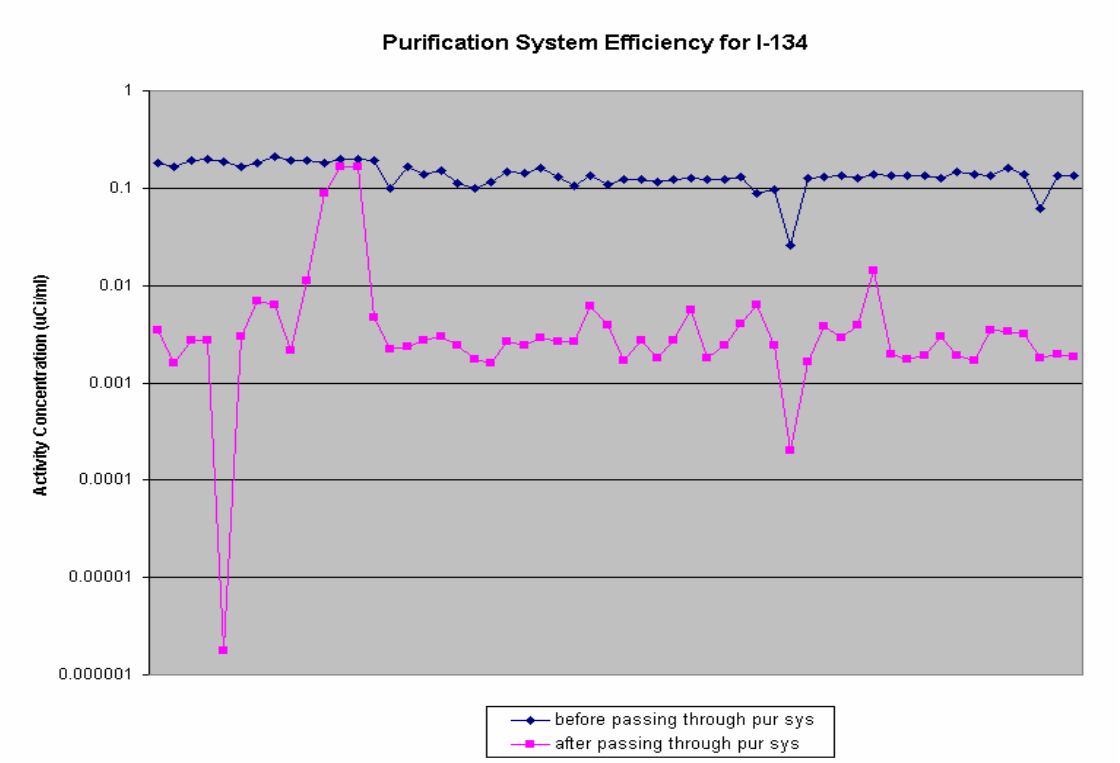

Figure 3: Measured Concentration of ^{134}I after and before the Purification System

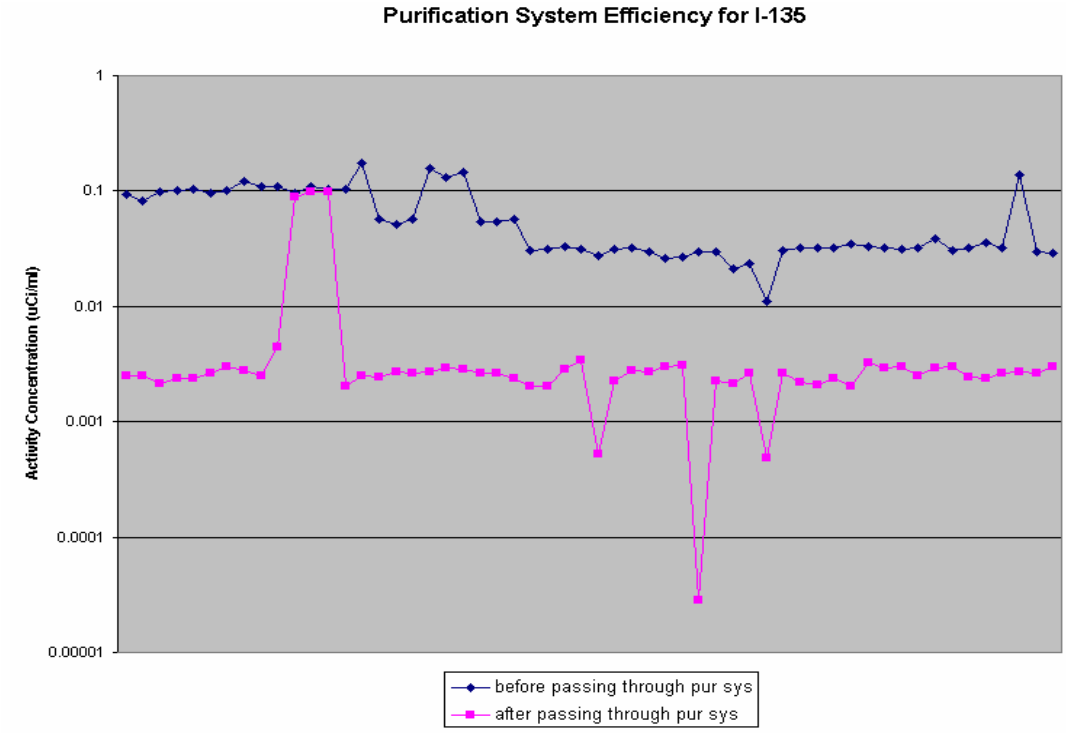

Figure 4: Measured Concentration of ^{135}I after and before the Purification System

2.3 Fission Product Plate-out

The deposition of fission products on heat transport system surfaces was modelled as a separate subsystem in the model.

Some of the soluble fission products, such as Iodine, Cesium, Tellurium and Rubidium, in the coolant are deposited on the surfaces of heat transport equipment and piping. There is too little information available on the exact effect of plate-out on the concentrations of radioiodine in the coolant to model this effect with confidence; however from comparing the simulated values with the measurements, the removal constant for the plate-out (lambda) is estimated to be about 100 times lower than for the purification system. [2] Thus, the effect of plate-out is negligible compared with removal by the purification system.

3 Results and Discussion

The Simulink model was used to predict the steady state radioiodine concentrations in the coolant of 3 CANDU stations. The parameters such as half life, capture and fission cross sections of the isotopes are known and unchanged; however the variables such as total coolant circulating mass vary in different CANDU designs and have been changed for each model.

The results of simulations were compared to long-term measured values and purification flow rates were adjusted until measured and model values of radioiodine concentrations agreed. The effect of deposition on radioiodine compared to the purification system is negligible so the purification flow rate is considered as the critical variable. Different purification flow rates were evaluated for the three stations named CANDU station A, B and C and the results are shown in Tables 3, 4 and 5.

The agreement of the Simulink model ratios compared with the measurements ratios is expressed as (Std/Ave) in tables 3,4 and 5. Where: Std is the standard deviation of the ratio of the simulation (e.g. column 3 in table 4) to the measured ratio (e.g. column 2 in table 4) and Ave is the average of the ratio of the simulated ratio (e.g. column 3 in table 4) to the measured ratio (e.g. column 2 in table 4).

Isotopes	Measurements	24 kg/s purification flow rate	12 kg/s purification flow rate
	Reactor A ratios with respect to ^{132}I	Simulink Model Ratios with Respect to ^{132}I	Simulink Model Ratios with Respect to ^{132}I
I-133	0.20	0.19	0.23
I-134	2.12	1.69	1.65
I-135	0.50	0.52	0.59
(Std / Ave) of Simulink over the measurement values		0.13	0.22

Table 4: Deduced Operating Flow Rate (CANDU Plant A)

Isotopes	Measurements	24 kg/s purification flow rate	12 kg/s purification flow rate	6 kg/s purification flow rate	2 kg/s purification flow rate
	Reactor B ratios with respect to ^{132}I	Simulink Model Ratios with Respect to 132I	Simulink Model Ratios with Respect to ^{132}I	Simulink Model Ratios with Respect to ^{132}I	Simulink Model Ratios with Respect to ^{132}I
I-133	0.21	0.19	0.23	0.31	0.49
I-134	2.50	1.69	1.65	1.55	1.34
I-135	0.65	0.52	0.59	0.68	0.83
(Std / Ave) of Simulink over the measurement values		0.14	0.25	0.41	0.65

Table 5: Deduced Operating Flow Rate (CANDU Plant B)

Isotopes	Measurements	24 kg/s purification flow rate	12 kg/s purification flow rate	6 kg/s purification flow rate	3 kg/s purification flow rate
	Reactor C ratios with respect to ^{132}I	Simulink Model Ratios with Respect to ^{132}I	Simulink Model Ratios with Respect to ^{132}I	Simulink Model Ratios with Respect to ^{132}I	Simulink Model Ratios with Respect to ^{132}I
I-133	0.23	0.19	0.98	0.31	0.42
I-134	2.81	1.69	1.70	1.55	1.42
I-135	0.54	0.52	0.91	0.68	0.78
(Std / Ave) of Simulink over the measurement values		0.23	0.86	0.40	0.54

Table 6: Deduced Operating Flow Rate (CANDU Plant C)

It is observed that in all three reactors the closest agreement with the measured concentration ratios is achieved with a purification flow rate of about 24 kg/s as shown by the (Std/Ave) values. This confirms that the stations are operating close to, or at the design purification flow rate value of 24 kg/s. Comparing the measurements and simulated values (Tables 7, 8 and 9) it can be observed that the shorter the radiological half-life is the more the simulation differs. Also, the activity of radioiodine in the coolant is systematically higher than the simulated values for reactor B and C. This indicates that the amount of historical tramp uranium should be higher than the assumed nominal value of one gram or that there may be defected fuel in the core. Further analysis is required to reason this discrepancy.

Purification Flow Rate	24 kg/s		12 kg/s	
R-A	Simulink Values/ Measurements	Ratio of ratios with Respect to ^{132}I	Simulink Values/ Measurements	Ratio of ratios with Respect to ^{132}I
I-132	2.60E+00	1.00E+00	4.05E+00	1.00E+00
I-133	2.50E+00	9.62E-01	4.75E+00	1.17E+00
I-134	2.08E+00	8.00E-01	3.17E+00	7.83E-01
I-135	2.70E+00	1.04E+00	4.80E+00	1.19E+00

Table 7: Comparison of Simulation Results with the Measurements (Reactor-A)

Purification Flow Rate	24 kg/s		12 kg/s	
R-B	Simulink Values/ Measurements	Ratio of ratios with Respect to ^{132}I	Simulink Values/ Measurements	Ratio of ratios with Respect to ^{132}I
I-132	2.03E-02	1.00E+00	3.17E-02	1.00E+00
I-133	1.87E-02	9.21E-01	3.56E-02	1.12E+00
I-134	1.38E-02	6.80E-01	2.10E-02	6.62E-01
I-135	1.62E-02	7.98E-01	2.87E-02	9.05E-01

Table 8: Comparison of Simulation Results with the Measurements (Reactor-B)

Purification Flow Rate	24 kg/s		12 kg/s	
R-C	Simulink Values/ Measurements	Ratio of ratios with Respect to ^{132}I	Simulink Values/ Measurements	Ratio of ratios with Respect to ^{132}I
I-132	2.70E-03	1.00E+00	4.21E-03	1.00E+00
I-133	2.25E-03	8.33E-01	4.28E-03	1.02E+00
I-134	1.63E-03	6.04E-01	2.48E-03	5.89E-01
I-135	2.61E-03	9.67E-01	4.63E-03	1.10E+00

Table 9: Comparison of Simulation Results with the Measurements (Reactor-C)

4 Conclusion

The Simulink model was run for three reactors, A, B and C and the results of the simulations were compared to long-term measured values and purification parameters were adjusted until measured and model values agreed. It was concluded that these reactors are operating with flow rates at, or close to the design value. However, in 2 out of 3 reactors the concentrations of radioiodine were higher than the results obtained from the model. It is believed that the amount of tramp uranium from defected fuel is much higher than the one gram that was assumed in this model and the radioiodine concentrations measured in the coolant are higher than the simulated values.

5 Acknowledgments

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6 References

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[3] American Nuclear Society; ANSI/ANS 18.1- 1999 Radioactive Source term for Normal Operation for Light Water Reactors