GENTILLY-2 HEAT TRANSPORT SYSTEM CHEMICAL DECONTAMINATION

by Jean Forest, Decontamination Project Engineer for Hydro-Quebec

Abstract:

Because the dose rates at Gentilly-2 around the reactor (reactor face components, feeder pipes and headers) are 5 to 7 times higher than any other CANDU-6, the refurbishment of G2, will necessitate the prior decontamination of these components. Hydro-Québec is in the middle of a decontamination project that includes artefact collection, oxide characterisation, decontamination processes qualification, application engineering, etc., that will lead to the application of a chemical decontamination process in the spring of 2011, before commencing the reactor retube activities. This paper provides details on the work and results achieved so far, the planned application strategy and the steps that lay ahead.

1.0 Past experience in Canadian Decontamination

A review was done of the decontamination applications on heat transport systems of CANDU reactors. Traditionally, application scenarios were always Full System Decontamination (FSD), except for some Pickering applications where a number of Steam Generators were isolated, keeping only one SG per quadrant, in order to reduce the mass of magnetite to dissolve. Such isolation is not possible in other CANDUs which do not have isolation valves at the boilers.

Applications used the HTS main pumps to circulate the chemicals, implying the operation of many auxiliaries, the establishment of a suitable system pressure (for pump NPSH requirement) and where the decontamination solution ends up wetting extensive parts of the HTS circuits, some of which do not actually require decontamination.

Applications were done in D2O, which implied deuterization of the resin prior to use and dedeuterization before disposing to spent resins.

Overall, despite the fact that these applications were generally successful, the Decontamination Factors (DF) achieved were rarely as high as what is deemed necessary for the Gentilly 2 retube project to be achievable. To put G2 on par with Lepreau, where the retube project is possible without HTS decontamination, Hydro-Quebec is requiring a minimum Dose Reduction Factor (DRF) of 5. The DRFs are usually smaller that the DFs, because of the radiation coming off structures not wetted by decontamination chemicals, such as calandria external tube sheet and the deeper layers of metals, rendered radioactive by activation. This means the actual required DF for G2 is closer to 6 than 5.

Historical Decontamination Results

In the table below are listed the large scale (FSD) decontamination applications done on CANDU reactors with in addition, the decontamination done at the Indian Point 2 unit (an American PWR) which also employed an AECL process.

In two instances a double back-to-back application was done (Pickering 1-1984 and Pickering 3-1989) for which a combine result is listed (grey highlighted).

Table 1: Historical Decontamination Results (Full System Decontamination applications)

#	Unit/Date	Average DF			Process
		Reactor face	SG	Piping	
1	Douglas Point - 1975	5,0	1,5	5,0	**CAN-DECON**
2	Pickering 1 - Nov. 1981	1,0	1,5	1,0	**CAN-DECON**
3	Pickering 1 - March 1983	1,3	-	1,5	**CAN-DECON**
4	Douglas Point - 1983 (*)	6,0	1,8	4,0	**CAN-DECON**
5	Pickering 2 - Jan. 1984 (*)	1,3	2,7	1,5	**CAN-DECON**
6	Pickering 2 - Apr. 1984	3,8	1,4	9	**CAN-DECON**
5&6	Combined results	5,1	4,1	13,5	
7	Pickering 1 - May 1984	5,0	3,5	3,0	**CAN-DECON**
8	Pickering 3 - Jul. 1985	5,0	10	18	**CAN-DECON**
9	Bruce 1 - Sept. 1986 (*)	< 1	2	1,6	**CAN-DECON**
10	Pickering 4 - 1986	Stopped because of mechanical problem			
11	Pickering 3 - Jul. 1989	2,2	2,3	2,8	**CAN-DECON**
12	Pickering 3 - Aug. 1989	2,4	1,5	1,6	**AP+ CAN-DECON**
11&12	Combined results	5,2	3,5	4,5	
13	Pickering 4 - Oct. 1991	3,8	3,2	10	**CAN-DECON**
14	Indian Point 2 - 1995 (PWR)	Average DF = 7,8			**5 steps (**)**

(*) Ended early because of running out of resin
(**) CAN-DEREM + AP + CAN-DEREM + AP + CAN-DEREM
Ref. COG-04-4013

Highlighted in purple are the values for "Reactor Face" and "Piping" (i.e. feeders and headers) where the results are below a DF of 5. This occurred 9 times out of 13 for "Reactor face" and also for "Piping".

One application was stopped because of mechanical problems and another (highlighted in red) resulted in a DF below 1, which means the fields were higher after than before, an Antimony re-deposition problem on the end-fittings.

Highlighted in yellow are the two best results.

All of the Canadian applications were done with the AECL CAN-DECON^{TM} process, a formulation which used EDTA, Citric and Oxalic acids and corrosion inhibitors (Rodine 31). The formulation changed during the time span in Table 1, to adapt to the decontamination specific objectives and some applications were done without any corrosion inhibitor (#1, 3, 4 and 9).

In the mid-1980's AECL developed the CAN-DEREM^{TM} process, similar to CAN-DECON, but containing no Oxalic acid. This is the process used at Indian Point 2 in 1995.

AECL tested a new formulation called CAN-DEREM Plus^{TM} during Steam Generator single-tube cleaning at Gentilly-2 (1993) and Point Lepreau (1994). In these applications, there were no purification system and the process relied on higher EDTA and Citric acid concentrations to do the job.

2.0 Evolution since 2001

Early in the G2 refurbishment project, dose estimates were compiled for all the retube activities around the reactor components. Initial estimates range from 5000 to 8000 man-Rems, which would necessitate a work force estimated at a minimum of 3400 personnel. Since the timely completion of this work with such a large number of staff is not considered feasible, it became clear that the G2 refurbishment was not possible without prior decontamination. The station personnel picked up the trail in 2001 and asked AECL to prepare a Technical Document to serve as a base line and to provide a budgetary estimate.

This document, 1666-79100-TD-001 was issued in 2002 and revised in 2006. Per this document, the oxide loading projected for 2011 was 1348 kg Fe3O4 (excluding SGs). This amount is about 5 times larger than the largest primary side decontamination application so far, worldwide. AECL proposed a full system decontamination scenario.

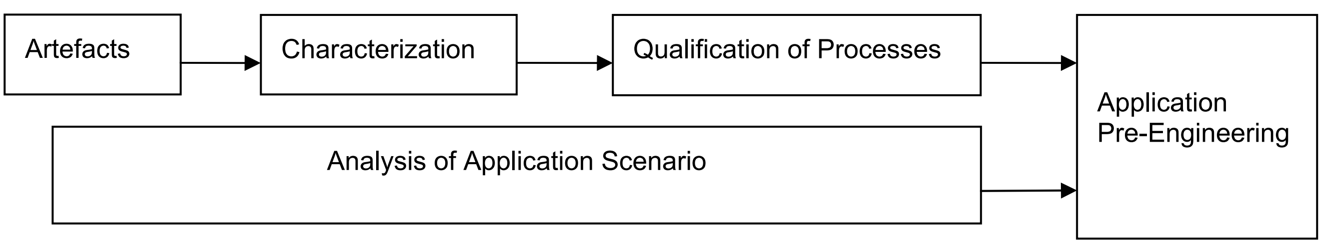
In the interim, Hydro-Quebec met with application vendors Westinghouse and AREVA (formerly Siemens Dekontamination) to get their input and further explore possibilities Their advice was: "Try to reduce the circuit size" and therefore the oxide loading to be dissolved at-once and the system volume. This is when the "small is beautiful" approach started to be nurtured by HQ personnel working on decontamination.

Realizing the unprecedented scale of the required application at Gentilly-2, HQ commissioned SNC-Lavalin Nuclear (SLN) to do a "Decontamination Technologies Review" in 2005. The report was issued at the end of 2006:

- World review of processes including AREVA, EACL, Hitachi et Westinghouse;
- Review of other decontamination methods (mechanical);
- Consider smaller HTS sub-circuits i.e. Partial System Decontamination (PSD).

From this report and the AECL TD-001, HQ established the following action plan.

Action Plan for successful Gentilly-2 decontamination :



Any battle, for chance of success, requires knowing the enemy. To that end, artefacts were required. AECL recommended an inlet feeder pipe sample (CS material) and a liner tube from inlet end-fitting (SS material).

Logical steps to follow are Characterization of deposits and Decontamination Process Qualification.

In parallel, an in depth analysis was to be one of the possible Application Scenarios (PSD), which permanent (station) equipment to use, how and where to connect, how to circulate (pumping) and what should the best, most effective decontamination circuit be.

Before going to the actual application engineering with a vendor, Application Pre-Engineering was also started.

3.0 Artefact Procurement (2006)

In a CANDU Heat Transport System (HTS), the radioactive contaminants not removed in real time by the purification circuit (Co-60, Antimony-122/124/125, Zr/Nb-95, Mn-54, etc.) will settle and mostly become imbedded in the Fe3O4 oxide layer that forms on the colder parts of the circuit (SG cold leg tubes, pumps with their inlet/outlet piping, inlet headers and feeder pipes and inlet end-fittings). In CANDU-6 reactors, which are deigned to operate with a significant boiling at the channel outlet (4 - 5% steam quality), this two phase fluid is more aggressive to the outlet feeder pipe base metal, which thins down more rapidly with in turn, a higher oxide build-up in the system over time.

Ideally, for a good characterization of the oxide, artefacts should be procured from the same reactor and for both most important materials: (feeder) Carbon Steel and (liner tube) Stainless Steel. Because no SFCR (Single Fuel Channel Replacement) was ever done at G2 and because of the prohibitive cost of one, the later was eventually procured from our Point Lepreau sister plant. For the CS sample, a partial inlet feeder pipe replacement was scheduled for a future annual outage.

The CS artefact is from the Gentilly-2 G11 inlet feeder pipe, removed in September 2006.

The SS artefact was procured from the Point Lepreau. It was removed from the R16 inlet end-fitting in May 1998 and had been in storage at Chalk River for some time.

4.0 Magnetite Characterization (2007)

The G-11 CS artefact sample was sent to Kinectrics inc. in Toronto in the spring of 2007 for examination and characterization of the oxide deposited over the years on the pipe inner wall surface. The information required was oxide thickness, loading (g/m2) and porosity, the deposit profile of the radioactive contaminants and inclusion (elemental) analysis.

Two important findings came out of this report, which directly affect the decontamination strategy:

A) Non-linear profile of decontamination

Cobalt-60, which is the dominant radioactive contaminant, is deposited in a profile "*consistent with a model based on the deposition of oxide from solution at constant rate and with a specific activity corresponding to the measured value for loose oxide followed by a radioactive decay of the deposited oxide over the entire service duration*".

% oxide removed	% Co-60 removed	Equiv. DF
51%	80%	5
74%	91%	11
87%	96%	25
96%	98%	50

This implies that high DFs are possible without the need to dissolve all the oxide down to the base metal.

Of interest also, is the observation that Antimony-124 "diffused" to deeper layers, as concluded from it's presence right down to near the base metal surface, despite its relative short half-life (66 days) compared to Co-60 (5,3 years). Not surprising, given the oxide porosity (30% average).

B) Higher oxide loading than anticipated

At 1000g of oxide per square meter of pipe, this pointed in the direction of a possible higher total oxide loading than what was predicted by AECL in 2006.

Oxide loading estimate revision

In the fall of 2007, AECL was informed of the Kinectrics results and asked to revisit their "Iron Transport Model" and confirm the oxide loading projection for a 2011 application at Gentilly-2.

In December 2007, HQ was informed that the revised oxide loading now stood at **2158 kg** (up 60% from the previous value of 1348 kg), values which do not include the magnetite present in Steam Generator, as HQ, at that point in time, was inclined more and more towards a Partial System Decontamination application scenario.

5.0 Process Qualification (2008)

By January 2008, HQ had a contract in place with two vendors to Evaluate and Qualify five different process cycles:

Westinghouse Richland Service Center (WA):

1. CITROX™ – AP – CITROX
2. CITROX – NP – CITROX
3. CITROX – (AP or NP) – CITROX – DFD Lite™

AECL Chalk River Laboratories (ON):

4. CAN-DERM™ – AP – CAN-DEREM
5. CAN-DERM Plus™ – AP – CAN-DEREM Plus

The tests were conducted per a Technical Specification written by SLN, in test loop that tried to reproduce as much as possible, the operating parameters expected during the application at Gentilly-2 in 2011, which are oxide loading, surface to volume ratio, flow (Re number), purification half-life, etc. The tests objectives were to determine process decontamination efficiency and demonstrate safety for the materials not being replaced during refurbishment. A series of corrosion coupons were included in the tests.

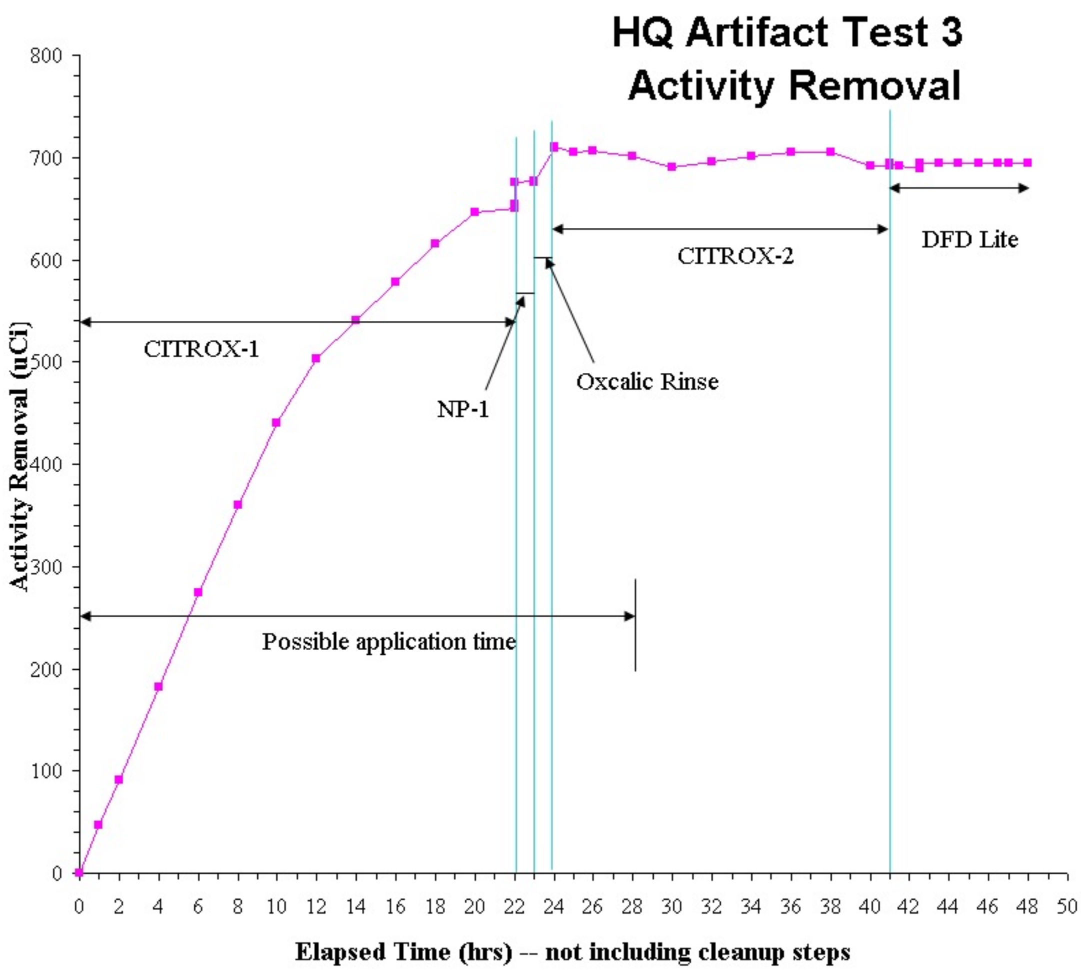
Results:

Consistent with Kinectrics findings, <u>all</u> the decontamination processes, at <u>both</u> laboratories achieved high DFs:

- DF on Carbon steel (feeder pipes) : > 700
- DF on Stainless Steel (End-fittings) : ~ 50

... and no process was rejected because of excessive corrosion.

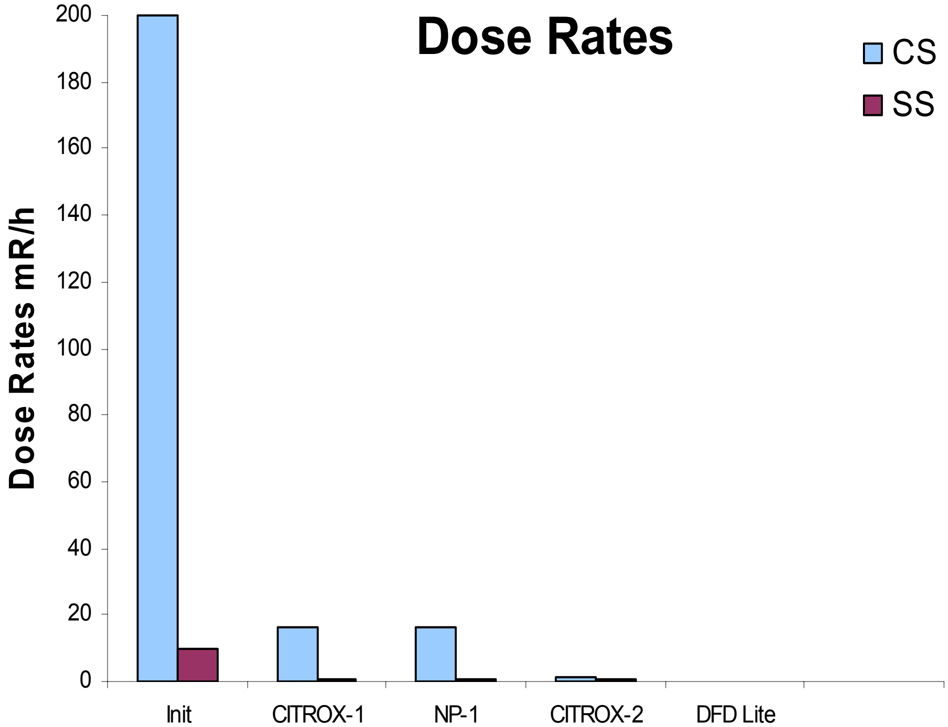
Below we reproduce a typical activity removal trend from the Westinghouse trials and the corresponding dose rates evolution bar graphs.



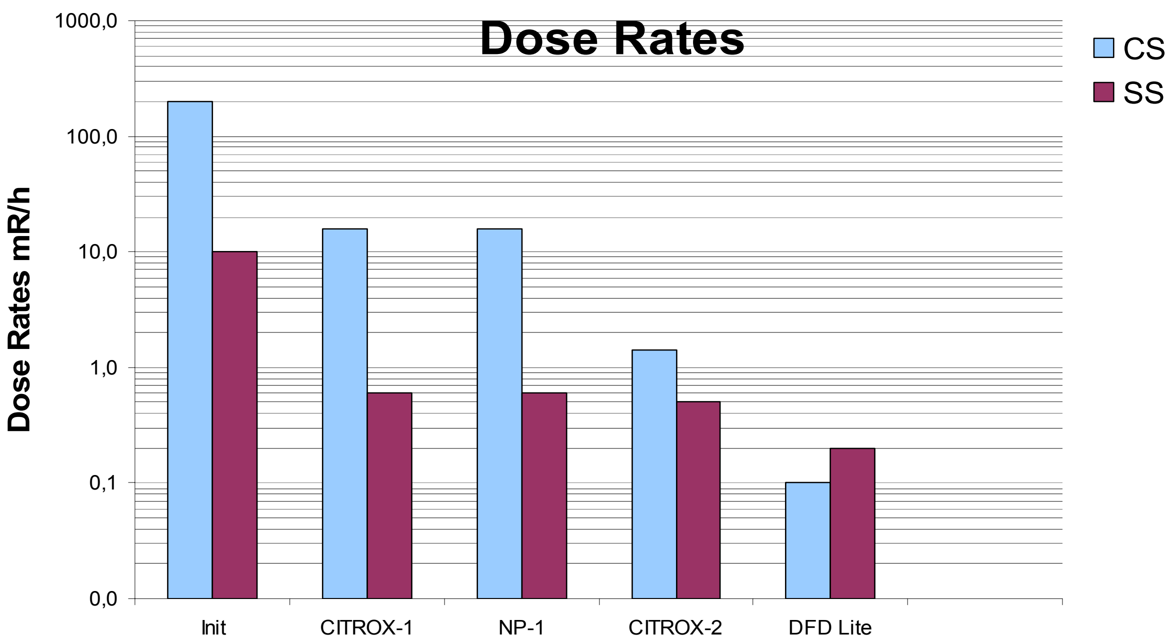
This trend seems to indicate that little is gained in the second CITROX or the DfD Lite steps. This is because the Activity Removal calculation is done by integrating the activity deposited on the resin bed, derived from the subtraction of the before/after activity measured in the solution. When these measurements get very low (near detection limit) the values can switch around randomly, giving the impression of re-contamination (CITROX-2 step) or no effective decontamination at all (DfD Lite step).

However, this trend is useful in showing that from a cost/benefit point of view the test could have stopped sometime before 30 hours.

A better sense of the decontamination progression in the later steps is given by the following bar graphs, derived from dose rate meter measurements:



With this linear scale, the effect of the DfD Lite step is not visible. See the same graph below, with logarithmic scaling:



These are laboratory results of course and HQ realises that the dynamic will be different during the application at site. Adjustment factors will no doubt apply.

The results bring two questions to mind:

1- Why are these DFs so high?

In addition to these being lab results, we believe the answer is: Because the G2 specific activity is very high in comparison to other nuclear generating stations, and because of the particular activity profile, eliminating most of the oxide layer, but not necessarily all of it, could yield an unprecedented DF in the industry.

2- Why a lower DF on SS artefact material?

If fact, this question is probably better formulated differently: Why are the Point Lepreau artefact samples DFs lower? And the answer becomes the same as above. In fact the "before" contamination on the Lepreau samples (Co60 + Sb124) was 10 times less than on the G2 artefacts. And the residual activity levels at end of decontamination cycles were similar to that of the CS samples, in either case, close to detection limit.

6.0 Application Preparation

6.1 Done so far:

- Discussions with AECL, in the context of the Integrated Work Group, on baseline scenario (FSD vs. PSD). A table of advantages and disadvantages was prepared. Initially AECL would not condone a PSD approach because it was felt that the large flow (possible only with main HTS pump operation) was crucial to the success of an application. Discussions and the results of Characterization and Qualification work have allowed to come to a consensus.

- Discussions with Westinghouse on available equipment. This vendor has a lot of very interesting equipment remaining from the Indian Point 2 application in 1995. Of particular interest are nine large (4,5 m3) Ion Exchange tanks with certification close to what we need in Canada, as well as other equipment ready on skids.

- OPEX review: The applications listed in Table 1 above were examined more in detail to classify problems in families and help determinate best practices and the ones to avoid. See Table 2 below.

- Getting the Project to advance the Resin Storage Tanks (7914) drain and refurbishment, in order to use station system piping for on-line sluicing during the application, a solution deemed much preferable to some 35 batch removal via 3 m3 containers and massive shielding + truck transport on site.

- Application Technical Specification (SLN: 020191-00000-45EG-0001). This document is necessary for the next step, the Application Request for Proposal.

- Decontamination Application Assessment Report (SLN: 020191-00000-45RA-0001). This recent report recommends the Partial System Decontamination scenario. See below for additional details.

- Decontamination Process Assessment Report (SLN: 020191-00000-45RA-0002). This recent report examines the Qualification Program Reports received and identifies, for each vendor, the best process.

Process Selection: The following are the preferred processes, for each vendor:

AECL: CAN-DEREM Plus™
Westinghouse: CITROX™-NP-CITROX + DfD Lite™

These processes were selected with the following criteria in mind:

- Process efficiency : Since all processes behave very well on this aspect, this criterion has lost some of its importance over the next two;
- Minimal corrosion to materials not replaced: This criterion remains high on our list because of the concern over the header nozzle condition and the piping portion (ECC/SDC) in the path of the decontamination solution that will not be replaced.
- Application safety and ease of control: Experience has shown that application is where the war can be won or lost. HQ favours a process that is simple to apply and has the least number of parameters to control or that can go astray.

6.2 Table 2: OPEX review

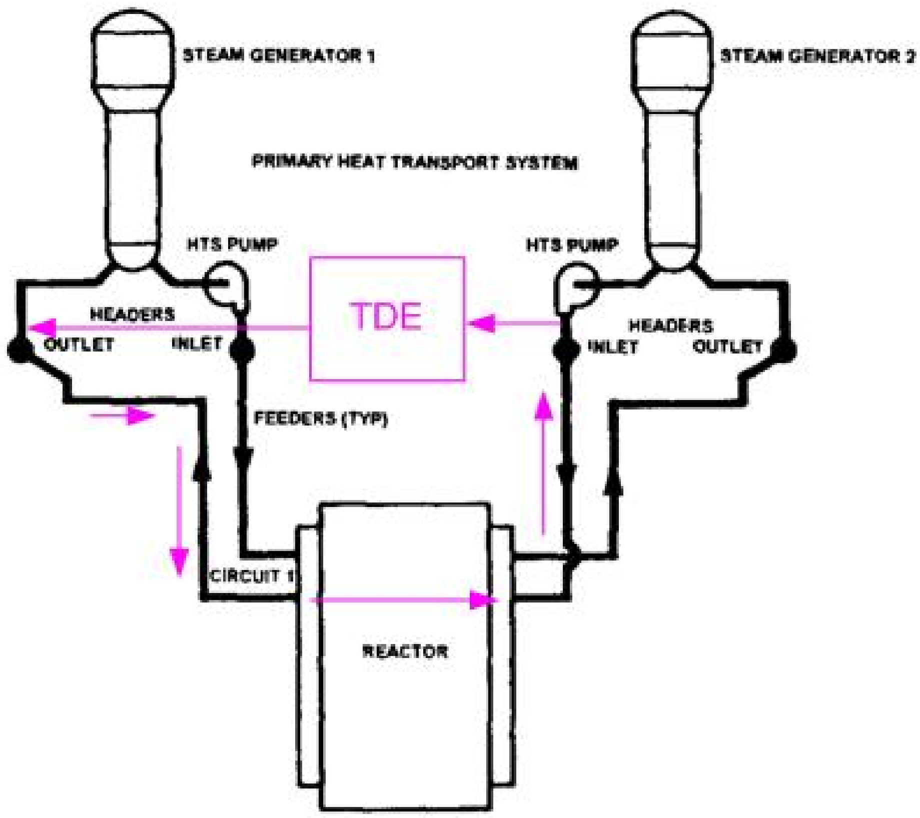
Problem (1)	Consequence	Mitigation at G2 (PSD)	FSD (2)
Lack of resin (# 4, 5, 9)	• Application stops • Low DF	• 9 IX tanks (4,5 m3 ea.) • H2O application (no slowdown due to deut-dedeut) • Ample resin reserve	Yes Yes Yes
Lower purification flow (# 2, 5, 6, 7, 9)	• Long application time • Higher corrosion	• Smaller sub-circuits • High purification flow	**No** **No**
Magnetite loading in SG (#1, 2, 4, 5, 6, 9, 11, 12, 13)	• Magnetite dissolution in SG slows down dissolution in other parts of circuit • Larger circuit volume	• SG not in circuit, or • SG mechanical clean before	Yes **Must**
Oxalate precipitation (#1, 2, 3, 4, 5, 6, 7, 9, 14)	• Final DF lower • Hot spots (local DF < 1)	• No Oxalic acid used	Yes
Antimony precipitation (#2, 3, 9, 11, 12)	• Negates decon effect • Hot spots (local DF < 1)	• In-core Sb cleaned before • Oxidation step	Yes Yes
Crud deposits (#2, 3, 5, 6, 7, 11, 12)	• Negates decon effect • Hot spots (local DF < 1)	• Reversed core flow, from clean side to dirty side • 100% flow filtration • Reduced fluid velocity to reduce end/fitting crudding	**No** **No** **No**

(1): # refers to application number listed in Table 1 above, where these problems occurred

(2): "No" means that the Mitigation in column at left is not present under Full System Decontamination scenario

6.3 Application Scenario Description

The favoured application scenario, the one that incorporates all of the mitigation factors listed above in Table 2, is to do four sequential partial decontaminations, header-to-header only (i.e. 1/4-core at a time):



TDE = Temporary Decontamination Equipment

Application characteristics:

- In H2O (large Tritium dilution);
- Small circuits inventory, approximately 30 m3 each including TDE;
- External pumps : 200 - 300 L/s = 2 - 3 L/s per channel;
- High purification flow (~100L/s) corresponding to purification half-life < 10 minutes;
- 100% filtration flow;
- Reversed core circulation: from outlet (clean side) to inlet (where most Fe3O4 resides).

These characteristics are part of our Application Technical Specification requirements.

7.0 Steps to come

7.1 Process selection

(See last bullet of section 6.1 above.)

7.2 Pre-engineering (ongoing contract with SLN)

- Finalization of Application Technical Specification
- Pre-engineering of inter-connections between HTS and TDE
- Engineering Plan

Hydro-Quebec is preparing the Request for Proposals, which will be issued this fall, with the objective to have a contract in place by January 15, 2009.

7.3 Application preparation (2009 to 2010)

2009 will be the year where the Engineering of the application is formally done. This will be a joint effort between HQ, SLN and the application vendor. Some complementary testing is also on our radar screen. For this we have in mind to procure the Westinghouse test loop equipment, assemble it at Kinectrics and finalize our process application parameter tuning. The engineering tasks include:

- Equipment : sizing, layout, floor loadings, shielding, inter-connections;
- Application Procedures;
- HAZOP study (session) to examine all stakeholders concerns (Operations, Tech. Unit, Safety, Chemistry, Environment, Radioprotection);
- Presentation to CNSC.

In 2010, the equipment will be purchased and assembled at the vendor site for full pre-commissioning (100 hour hot functional test). At this time we will also be doing:

- Procurement of resins, filters and other staples;
- Personnel training;
- Final procedure approval by G2 Operations.

The Application at site is scheduled approximately 75 days after reactor shutdown during the Retube outage (~June 2011). Its duration is targeted for 21 days on critical path.

8.0 Acknowledgement:

This work is the result of a team effort that started in 2001. Special thanks to:

- G2 station personnel: Pierre Gauthier, Steve Plante and Wilfried Goddyn;
- Kinectrics: Vivian Chew, Guylaine Goszcziynski and Aamir Husain,;
- AECL personnel: Jaleh Semmler, David Guzonas;
- Westinghouse personnel: Darik Tippet, Jamie Herman and Powell McLean;
- SNC-Lavalin Nuclear personnel: Ralph Heart, Peter Wendl, Andrzej Krukowski;
- Niagara Technical Consultants: Jerry Smee;
- ... and to the people who direct the fine work of these specialists.