

DESIGN TO NULLIFY ACTIVITY MOVEMENT IN
HEAT TRANSPORT SYSTEMS

by

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ABSTRACT

This article describes the methods by which designers can reduce the adverse effects of system corrosion and the resultant activation of the corrosion products in Heat Transport Systems.

The presentation will cover:

- a) Choice of materials.
- b) Assessment of the need of components.
- c) Control of system chemistry.
- d) Factors considered in sizing HTS purification systems.
 - i control of activation and fission products
 - ii decontamination

1. INTRODUCTION

The time interval from the freezing of the design basis for the major systems in a nuclear power station, up until the station is commissioned and producing power, is at least five to seven years. As a result, the comparisons which can now be made between Douglas Point and Pickering are comparisons of designs produced some seven and more years ago. Certainly substantial improvements in activity transport are evident as one compares these two stations. The system designers have been watching the performance of these earlier designs and have been changing the design as a result.

This paper then is primarily about the heat transport and purification system designs which are now on the drawing boards. These systems are expected to show substantial improvements over currently operating systems, from the standpoint of the limitation of the production, movement and accumulation of radioactivity.

Let us not forget, however, that the primary heat transport system of a CANDU-PHW* reactor is a complex system and that during the evolution of a design there may be many competing and at times conflicting requirements to consider. For instance, boiler heat transfer can be improved by permitting the coolant to boil as it passes over the fuel elements, but the boiling creates a different chemistry regime, one in which there will be higher concentrations of dissolved oxygen in the lower temperature condensed coolant. The steam phase at the core outlet will also contain most of the elemental oxygen and deuterium. The greater availability of oxygen increases the probability that system materials will corrode and this in turn leads to changes in materials.

Within the context of activity movement, consideration has to be given to activated corrosion and erosion products from system materials, to fission products from fuel failures, to the effects that changes in operating conditions - pressure, temperature and chemical - might have on corrosion and erosion rates and to the steps that can be taken to reduce the resultant radiation levels around the plant to acceptable levels; levels, incidentally, which continue with passage of time to be set at lower and lower values.

The methods currently employed in the design of heat transport systems to meet this situation include:

1. Elimination of all unnecessary components.
2. Increased component reliability.
3. Attention from the earliest conceptual plant arrangements to accessibility, layout and shielding.
4. Careful choice and specification of materials including the elimination as far as is practical and/or economic of undesirable materials and of trace impurities in acceptable materials.

*Canada Deuterium Uranium-Pressurized Heavy Water

5. Control of heat transport chemistry.
6. Provision of an adequate purification system.

Before taking each of these topics and examining it in more detail, it is necessary to present a background against which to view the efforts to nullify the movement of activity and its effects.

Experience at Douglas Point, and to a lesser degree at Pickering, has shown that there is a buildup of radioactivity around the heat transport system and this creates a problem for both operating and maintenance personnel which shows up as the radiation dose they receive. At Douglas Point this has resulted in extra, non-station, personnel being employed at times, particularly during major maintenance shutdowns, because the normal station complement could not do the necessary work without exceeding the permitted radiation exposures.

At Pickering, radiation doses have not yet been such an inconvenience.

Although the whole story of activity movement has not yet been unravelled, some effects have been identified and later papers will tell of these. Suffice at the moment to say that, from our experience at Douglas Point, largely as a result of Barry Montford's work, some conclusions have been reached. At Douglas Point, it was seen that corrosion products can move around the system and that changes in chemistry, temperature, and hydraulics cause them to move. These techniques were exploited to great advantage to reduce radiation fields around the Douglas Point steam generator to 20% of their peak values.

However, of particular interest to those involved in purification system design were the observations on the redeposition of crud which showed that during normal operation, crud redeposits with a half-life of 8-12 minutes and during shutdowns this redeposition half-life extends to an hour or more. Crud movement has also been seen at Gentilly-1 and at NPD but it has not been so extensively documented at either plant.

This then is the backcloth - active material moving, depositing, moving again and redepositing. The problems - how to reduce the movement and how to catch the material which is moving? To answer these questions we have to return to the six topics mentioned previously.

2. ELIMINATION OF UNNECESSARY COMPONENTS

The most prominent example of this is the reduction in the number of heat transport system valves. The original impetus came from a desire to improve on the leak tightness of the system. However, since

many valves had high cobalt content hard facing materials, their elimination removed a potential source of cobalt-60. So whereas at Douglas Point some 400 valves are exposed to HTS water, at Bruce there are fewer than 20 valves exposed to high temperature coolant.

Closely related to this is the reduction of the number of components. Thus whereas each Pickering unit has sixteen pumps and twelve boilers, the 600 MW(e) reactors have four pumps and four boilers. The main advantage expected from this is a reduction in total maintenance time. This then leads into the second topic -- reliability.

3. COMPONENT RELIABILITY

By increasing component reliability, for example using proven, bellows-sealed valves instead of conventionally packed valves, the frequency of maintenance can be greatly reduced. Similarly attention has been given to the design of the internals of ion exchangers, the shaft seals on pumps and the choice of boiler tube material, all from the standpoints of reducing the frequency of maintenance and also of making any anticipated maintenance easier.

4. ACCESS, LAYOUT AND SHIELDING

The layout of the components can have a great effect on the total dose absorbed by the station personnel. Ease of access to and convenient working conditions in radiation areas must be given attention early in the design process. Consideration must be given to equipment position, material handling and the arrangement of stairways, passages, platforms and shield walls. As examples - in current designs, all transfers of spent radioactive, or potentially radioactive, ion exchange resins are by slurry methods through shielded pipes. Boilers and pumps are in shielded compartments with shielding between the main pumps and their drive motors.

5. CHOICE OF MATERIALS

If it were possible to obtain construction materials which could withstand the maximum operating conditions of the heat transport system (up to 310°C (590°F) and 11.2 MPa(gauge) (1630 psig)) without

corroding, and yet still be economic, corrosion product transfer would not be a concern. However, this utopian condition is not yet with us.

Factors which affect the choice of HTS materials include:

1. Corrosion rates
2. Variation of corrosion rates with
 - (i) chemistry
 - (ii) temperature
3. Impurities in the materials - particularly cobalt
4. Neutron economy (for core materials).

As a result, the CANDU-PHW heat transport system includes:

1. Zirconium alloys for pressure tubes
2. Carbon steel for headers, feeders and major piping
3. Nickel alloys as the boiler heat transfer surface. Monel-400, Inconel-600 and Incoloy-800 have all been used or proposed for use in boilers; Monel was used at Douglas Point and Pickering 'A' and is the proposed material for Pickering 'B', Inconel was used at NPD and Bruce and Incoloy is being used in all the present 600 MW(e) reactors.

In specifying boiler tube materials, there has been a continuing downward movement in the permitted cobalt level. The maximum allowable cobalt concentration is now 0.015% or 150 ppm. For the carbon steel piping, this approach is continued, and for the Bruce 'A' G.S. the carbon steel pipe has an average cobalt concentration of only 0.006%. Without these restrictions and control, about 12 g/a of Co would have come from C.S. corrosion on each Bruce reactor. As a result of them, the quantity is down by a factor of ten.

Monel has given very good service both at Douglas Point and Pickering where the coolant is expected to contain less than 5 ppb of oxygen, but if the HTS oxygen content is allowed to rise above 5 ppb, the Monel corrosion rate increases rapidly (see Figure 1). Since Bruce and the 600 MW(e) reactors will have boiling in the reactor core, oxygen will be present in the coolant leaving the reactor core and flowing through the external parts of the system, and so Monel is not a candidate material for boiler tubes. Inconel was chosen for Bruce partly as a result of good performance at NPD and partly as a result of the choice of this material for the boilers in the pressurized light water reactors. Our present choice of Incoloy stems from secondary side, chemical upset condition investigations, primarily by Siemens, and the observation that Incoloy is not inferior to Inconel, but is cheaper.

CORROSION RATE $\text{mg}/\text{dm}^2 \text{ day}$

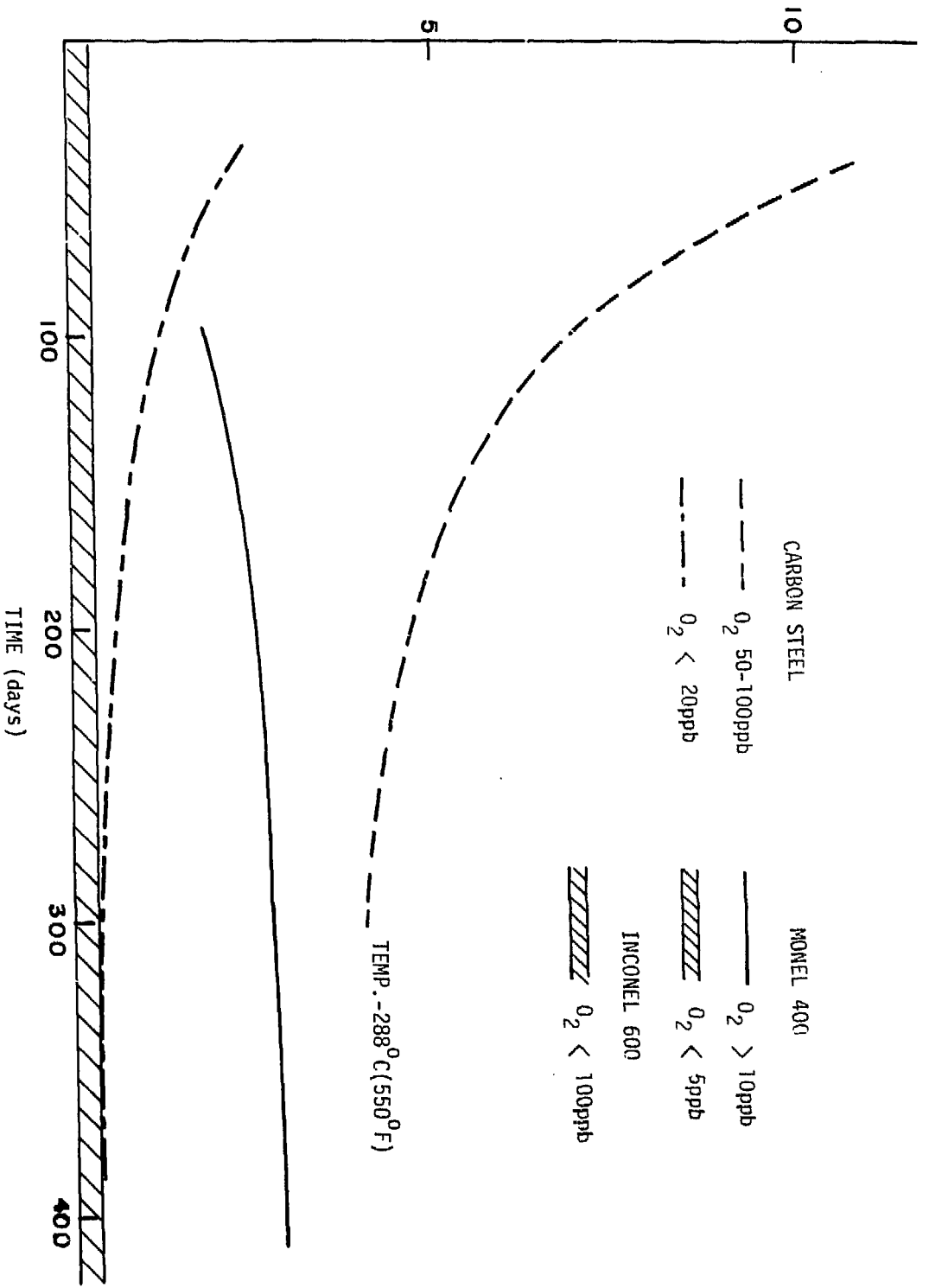


FIGURE 1: EFFECT OF $[O_2]$ ON CORROSION RATE

Typical areas of the major materials in contact with the coolant are: boiler tubing, 10,000 to 20,000 m²; feeders and headers, 2700 m²; and zirconium alloys, 4500 m². The range on boiler tubing area reflects the effect of boiling and no boiling in the PHT system coolant.

6. CONTROL OF HTS CHEMISTRY

The use of carbon steel as a major construction material dictates the use of a high pH, low oxygen coolant (<5 ppb O₂). These requirements are achieved by dosing the HTS coolant with lithium hydroxide to an "apparent pH" of 10.0 to 10.5 and maintaining a dissolved deuterium content in the coolant of 5 to 10 ml/l. ("Apparent pH" is the reading obtained when a D₂O solution is monitored with a pH instrument which has been buffered with H₂O chemicals.)

The required chemical parameters are maintained through a program of regular sampling and associated laboratory analysis. This program also includes checks on conductivity, crud levels, dissolved materials (such as lithium, copper, iron, nitrate, chloride, carbonate) and radioactivity as gross beta/gamma and iodine-131. Table 1 shows typical chemistry control parameters for the heat transport coolant.

TABLE 1
REACTOR COOLANT CHEMISTRY

<u>PARAMETER</u>	<u>PERMISSIBLE RANGE</u>	<u>DESIRED VALUE</u>	<u>SAMPLE FREQUENCY</u>
pH at 25°C*	10.2 - 10.8	10.5	2/week
LITHIUM (I) as Li (mg/kg D ₂ O)	0.7 - 2.0	1.0	2/week
(II) as LiOH (mg/kg D ₂ O)	2.4 - 6.9	3.4	
CHLORIDE (mg Cl ₂ /kg D ₂ O)	<0.2	<0.05	2/week
SUSPENDED SOLIDS (mg/kg D ₂ O)	<1.0	<0.1	1/week
CONDUCTIVITY (μS/cm at 25°C)	20	15	1/week
DISSOLVED DEUTERIUM- (mC D ₂ /kg D ₂ O)	5 - 15	10	Once every 2 h
DISSOLVED OXYGEN** (μg O ₂ /kg D ₂ O)	<10	<4	1/week

* Measured on pH meter. The permissible range is determined by carbon steel corrosion rates. Zircaloy and Incoloy have much lower rates of corrosion and the rates are much less sensitive to change in pH.

† Automatic by Chromatograph

** If level rises above 10 μg O₂/kg D₂O resample to confirm analysis. Check dissolved D₂ level.

An important part of corrosion control is the pre-operational hot conditioning of the system to build up a protective magnetite film on the carbon steel surfaces. Using pump heat, the heat transport system temperature is raised between 200 and 250°C (390-480°F). The water is dosed initially with 100-200 mg/kg (100-200 ppm) of hydrazine. The pH is maintained between 9.8 and 10.5 to minimize magnetite solubility. The duration of the hot conditioning is 8 to 10 days. As a preliminary to the hot run, the system is circulated for up to a day through the purification system to remove crud and dissolved impurities.

During the hot run, a regular program of chemical sampling is maintained. Two of the more important parameters are crud colour and oxygen concentration. Coupons are also used to follow the progress of the magnetite film formation.

7. THE PURIFICATION SYSTEM

The main reason for heat transport system purification is to keep the heat transport fluid and surfaces it contacts as free as is reasonably possible of impurities which are, or may be, hazardous to the operation and maintenance of the station.

Within the context of activity movement, consideration must be given to the control of both activated corrosion products and fission products and to whole system decontamination. Also included in the overall purification design is the control of non-radioactive ionic impurities and particulate matter.

As already mentioned, the experience at Douglas Point has provided useful practical information on crud redeposition rates. If the purification system is to reduce the rate of buildup of radioactivity, it has to compete with the redeposition processes; hence the purification half-life must be similar to the "on-power" redeposition half-life. This gives a system which has a shorter half-life than the redeposition half-lives during shutdowns.

The relationship between redeposition and purification in its simplest form can be written

$$X_p = \frac{\frac{1}{t}}{\frac{1}{t_{1/2_p}} + \frac{1}{t_{1/2_r}}}$$

where X_p is the fraction of material removed by purification, $t_{1/2}$ are half-lives and the subscripts 'p' and 'r' refer to purification and redeposition respectively.

This relationship shows (see Figure 2) that the smaller the purification half-life, the higher the fraction of corrosion products removed. For this reason, continuous high flow rate purification is advocated. Because the purification systems use conventional ion exchangers which can be used only at relatively low temperatures 65°C (150°F), an interchanger-cooler combination is used to reduce heat losses.

The present 600 MW(e) reactors are provided with an HTS purification system giving a minimum purification half-life of 20 minutes which corresponds to an X_p value of about 0.33 during power operation and 0.75 during shutdowns.

The removal of fission products, released from fuel defects, also has to be considered. With what is called a "normal" defect rate of 0.5%, the ensuing radio-iodine release can be controlled by a purification system with a half-life of about two hours. Because both soluble and particulate material must be removed, the purification system includes both filters and ion exchangers.

The flowsheet for the purification system for the proposed 1250 MW(e) reactors is shown in Figure 3. This plant has three heat transport circuits. All three feed into the purification system.

The purification system includes an interchanger, cooler, three filters in parallel with a by-pass, two ion exchangers in parallel with a by-pass, flow control and temperature monitoring. The system is designed for a purification flow of 142 kg/s (1.1×10^6 lb/h) of D_2O . The ion exchangers have a nominal diameter of 1.5 m (5 ft) and each contains approximately 1400 ℓ (50 ft^3) of a 1:1 cation/anion mixed bed. The system operates at full system pressure taking flow from the main HTS circulating pump discharge headers and returning flow to the appropriate pump suction lines. The filters have a canned arrangement of pleated paper cartridges. Suitable shielded flasks are provided for the transportation of the used canned filter cartridge assembly to disposal.

The purification flow of 142 kg/s is derived from the purification cleanup equation:

$$C = C_0 e^{-\frac{Ft}{M}}$$

where C and C_0 are impurity concentrations at times: $t = t$ and $t = t_0$.

F = purification flow rate, t = time the flow is on and M = mass of fluid in the system being purified. The half-life is the time required to reduce an

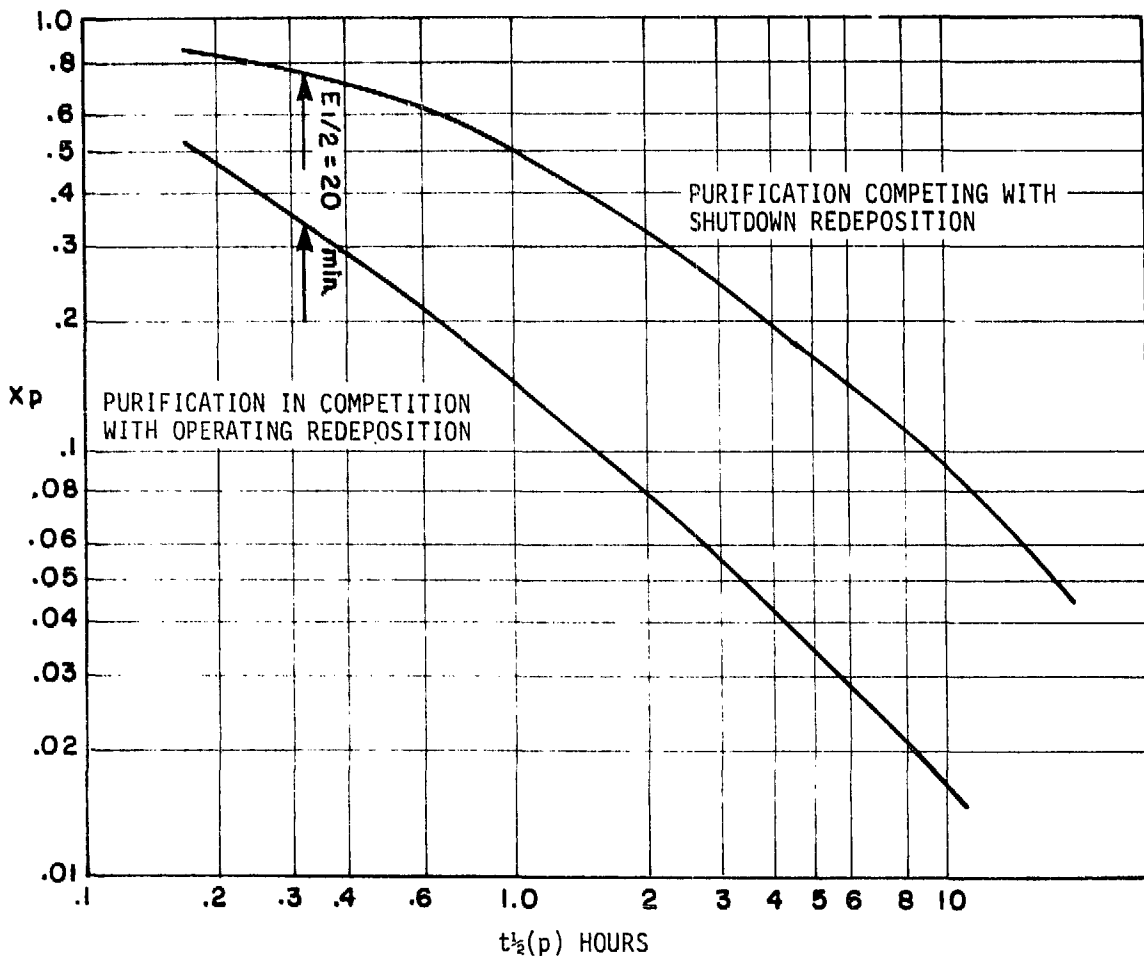


FIGURE 2: FRACTION OF RELEASED CRUD COLLECTED BY PURIFICATION SYSTEM OF VARIOUS HALF-LIVES

ASSUMING $t_{1/2}(r)$. RE-DEPOSITION HALF-LIFE = 0.167 h OPERATION
 = 1.0 h SHUTDOWN

AND:

$$X_p = \frac{1}{\frac{1}{t_{1/2}(p)} + \frac{1}{t_{1/2}(r)}}$$

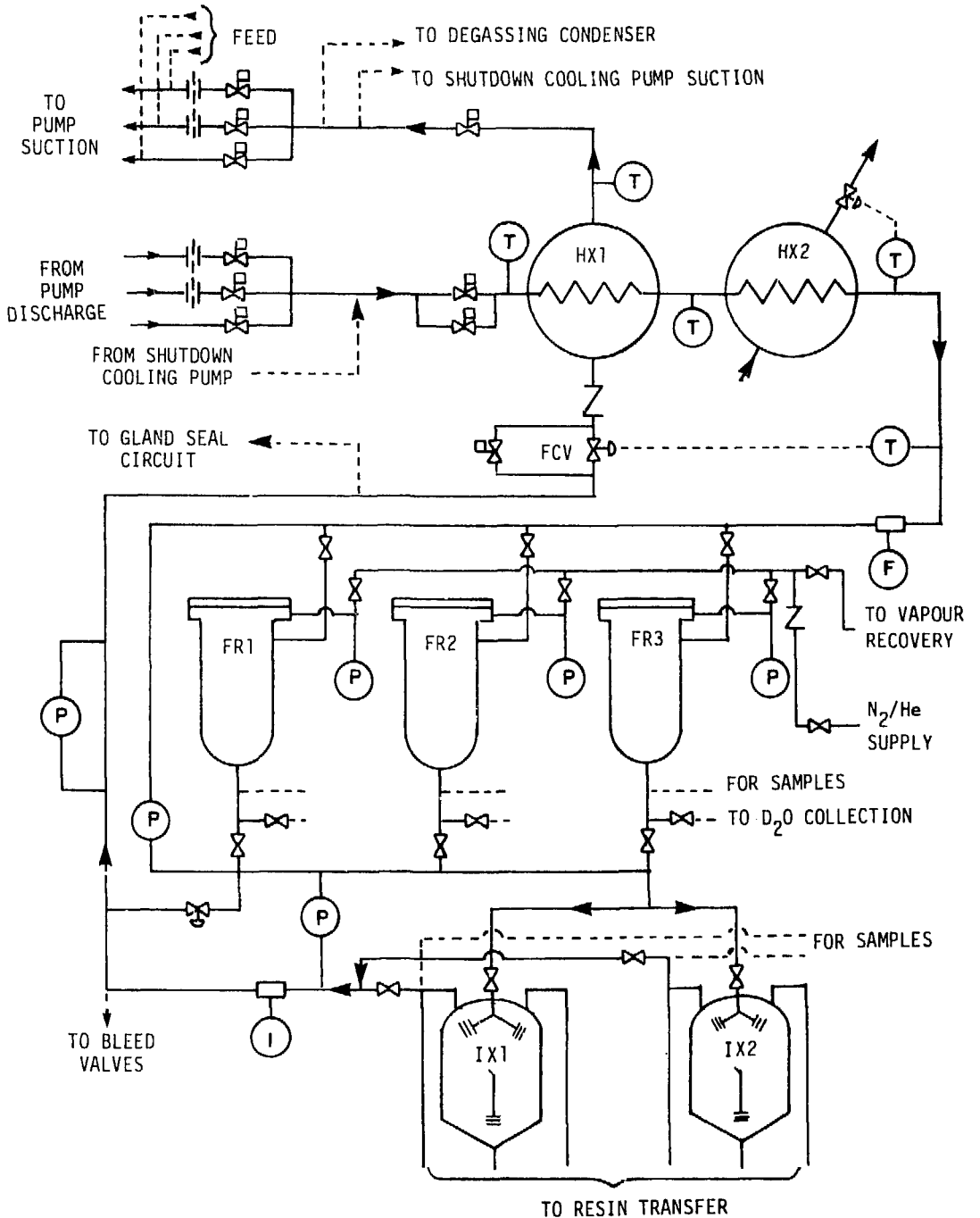


FIGURE 3: HIGH FLOW PURIFICATION SYSTEM

impurity concentration by a factor of 2 when no new impurity is being added. The relationship between flow and half-life can be obtained by manipulating the above equation to give:

$$F = \frac{\ln 2 \times M}{60 \times t_{1/2}}$$

where $t_{1/2}$ = half-life for purification (in minutes), M = HTS D₂O inventory in kg (245,100 for the 1250 MW(e) reactor at full power) and F = the required purification flow (kg/s).

Mention was made earlier of provision for whole system chemical decontamination. The need for this became evident when it was observed that although the radiation levels around the Douglas Point boilers could be reduced (and were by a factor of about 5) by a judicious combination of chemical, thermal and hydraulic cycling, there was little effect on the radiation levels around the carbon steel components. Also, during subsequent operation, there was little evidence of active material migrating from the carbon steel components to the boilers. Added to this, attempts to reduce radiation levels at NPD, using the Douglas Point techniques, were unsuccessful showing that Inconel boilers can not be decontaminated using chemical, thermal and hydraulic cycling. So other methods had to be reviewed and assessed. This work has and is continuing to be done at the AECL* laboratories at Chalk River and Whiteshell.

As far as the designers of purification systems are concerned, facilities must be provided to permit whole system decontamination. The brief details and background to the method follow but fuller details will come in later papers, especially that to be given by Paul Pettit.

An important factor in selecting a decontamination method for whole system decontamination of a PHW reactor heat transport system is the use of heavy water (D₂O) as the coolant. This largely rules out the use of conventional methods, which use relatively concentrated solutions in inhibited inorganic acids in ordinary water (H₂O) followed by copious flushing with ordinary water. Instead, milder reagents were assessed, especially those that could be regenerated and removed by ion exchange.

The present reagent is a mixture of complexing organic acids which dissolve the oxide film, which is formed during operation on the heat transport system surfaces, together with the embedded fission products. As these resultant chelates pass through the deuterium form cation resins of the purification ion exchangers, the metal ions are retained on the resin and the free acids (in their deuterium form) are returned to the heat transport system. Any particulate crud is removed by filtration.

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At the end of the decontamination the organic acids are removed by anion exchange resins.

The advantages of this method, called CAN-DECON, are

- There is very little downgrading of the heavy water coolant,
- the concentration of the reagent is low (about 1g/kg D₂O),
- because of the reagent regeneration, the process can continue until removal of active species has stopped,
- the only wastes are exhausted ion exchange resins and the small volume of ordinary water used to slurry these resins to the spent resin storage tank. Our experience has shown that there is only minimal leaching of the active species from these resins.

The feasibility of the CAN-DECON process has been demonstrated at

- Gentilly-1 with a whole system decontamination,
- NPD, again with a whole system decontamination,
- Douglas Point with a bleed cooler decontamination.

Also a detailed procedure is in a very advanced stage of preparation for a whole system decontamination of Douglas Point in 1975. It is expected that with the facilities provided as a normal part of the 600 MW(e) design, that a CAN-DECON process will be carried out in a period of 36 to 40 hours. For future multi-unit stations it may be preferable to provide a skid mounted unit which can be used on all units.

8. RECAPITULATION

In the design of heat transport systems, the main methods used to nullify activity transport are:

1. Choice of materials, including choice of materials with low rates of corrosion, limiting noxious impurities.
2. Elimination of unnecessary components.
3. Improvement of component reliability.
4. Laying out of equipment to permit easy access for maintenance. Provision of necessary shielding.

5. Controlling HTS chemistry to minimize corrosion.
6. Provision of an HTS purification system and ancillaries which:
 - a) competes with redeposition of crud,
 - b) removes fission products, and
 - c) provides facilities for whole system decontamination within the poison-out period.