Chemistry - PI 24

TRANSPORT OF RADIOACTIVITY IN NUCLEAR REACTORS

Objectives:

1. State that the three categories of radioactivity transport in the primary transport system are:

   (a) Coolant Activation Products
   (b) Activated Corrosion Products
   (c) Escaping Fission Products.

2. State that Escaping Fission Products have three sub-categories which are:

   (a) Gaseous Fission Products (Inert)
   (b) Halogen Fission Products
   (c) Depositing Fission Products

3. Give two examples each of the first two categories from objective 1 and of the three sub-categories of objective 2. In one or two sentences each, describe the fate of the substances in each category or sub-category.

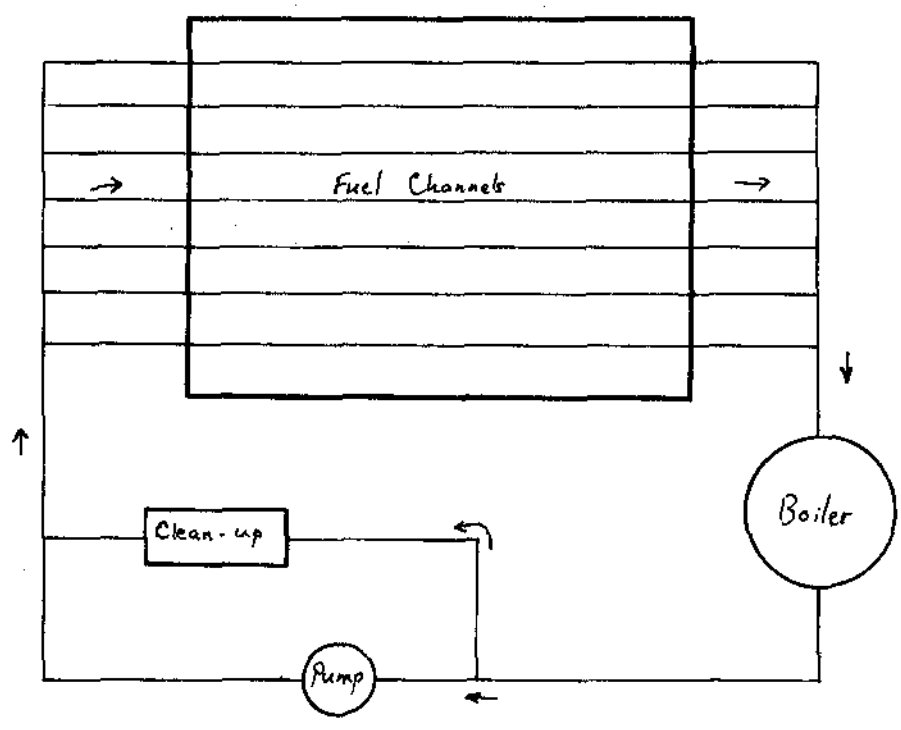


Figure 1

A CANDU Reactor

A CANDU reactor as shown in Figure 1 consists basically of a large tank filled with moderator and loaded with fuel. A coolant is pumped through the fuel channels taking the heat to the boiler where it produces steam to turn the turbine. **As a result of neutron bombardment the coolant will contain radioactivity due to (1) coolant activation products, (2) activated corrosion products and (3) escaping fission products.** Each of these activities presents its own types of problems. A clean-up loop is added to the system; part of the flow continuously passes through this loop for removal of corrosion and fission products.

COOLANT ACTIVATION PRODUCTS

Table I gives a list of the activation products from neutron bombardment of the D_2O coolant. They fit into two separate groups: the long-lived ^3H and the short-lived products.

TABLE I

Coolant Activation Products

Product	Reaction	Half Life	Decay Mode	Principal Gammas (MeV)
^3H	$^2H(n,\gamma)^3H$	12.23 yrs.	β^-	
^{16}N	$^{15}N(n,\gamma)^{16}N$	7 sec.	β^-,γ	6.13(69%), 7.11(5%)
^{17}N	$^{17}O(n,p,)^{17}N$	4.2 sec.	β^-,γ,n	0.87(8%), 2.19(0.5%)
^{19}O	$^{18}O(n,\gamma)^{19}O$	29 sec.	β^-,γ	0.197(97%), 1.37(51%)
^{17}F	$^{16}O(d,n)^{17}F$	66 sec.	β^+	0.511(200%) (β^+ annihilation)

Tritium (^3H or more commonly T) emits no gamma rays. It undergoes beta decay only releasing a beta particle with a maximum energy of 17 keV. This is not strong enough to penetrate the page of this paper. Its range in air is extremely low. Nevertheless it presents a great hazard. If absorbed by the body it can incorporate itself into biological material, where its beta is completely absorbed by the body, thereby, leading to radiolytic damage of the body cells. Tritium presents a hazard only in the D_2O moderator reactors; it is not produced with other moderator systems.

Chemically tritium is present as water or more precisely as DTO which is formed by the exchange of the radiogenic T with the D_2O moderator

$$T + D_2O \rightleftharpoons DTO + D$$

As the reactor runs, more and more tritium is produced; thus, the tritium level of the water increases. A leak in the system becomes a serious matter as it is tritiated water that will escape into the atmosphere. There it will exchange with the H_2O vapour present to give HTO as well as DTO.

$$DTO + H_2O \rightleftharpoons HDO + HTO$$

Continuous tritium monitoring of the atmosphere is necessary for two reasons. It gives an indication of the tritium hazard for those working in the area. (This is usually backed up by bioassay measurements). It also gives a measure of the D_2O leakage rate as the tritium level is also a measure of the D_2O level provided the D_2O's tritium level is known.

Monitoring can be accomplished by passing air through a chamber of the type shown in Figure 2 or by bubbling air through ordinary water where there will be exchange transferring T from the vapour to the liquid which can then be counted.

$$DTO(v) + H_2O(l) \rightleftharpoons \begin{cases} DHO(v) + HTO(l) \\ HTO(v) + HDO(l) \end{cases}$$

The other group of coolant activities are all short-lived. They make the reactor area inaccessible during operation. Of those listed in Table I, ^{16}N presents the greatest problems. Its 6.13 and 7.11 MeV gamma rays and very penetrating and extremely difficult to shield. All of these activities decay rapidly; thus, the radiation fields are reduced in a matter of minutes after shutdown. Any impurities in the water will also contribute to the activity of the coolant. It is thus very important that the water be pure when added to the system.

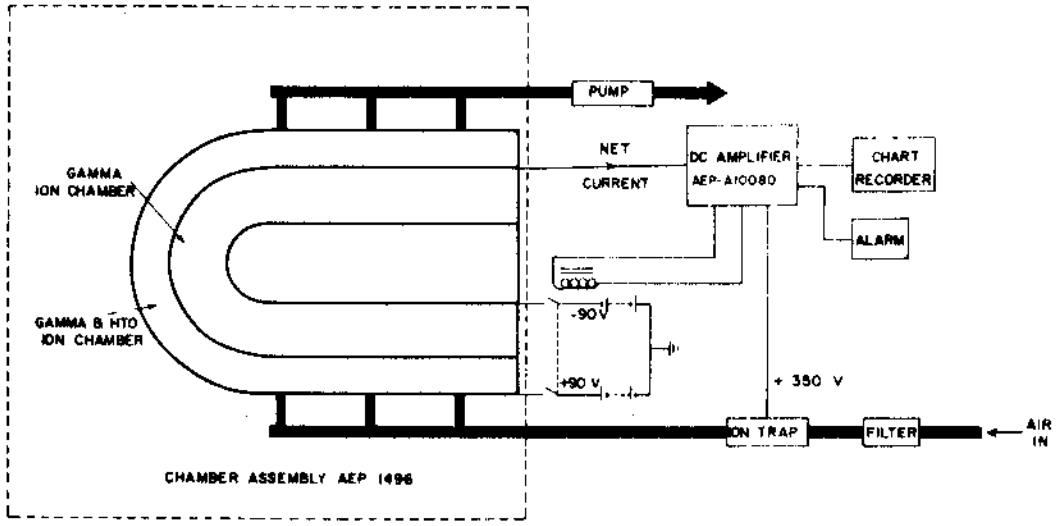


Figure 2

Block Diagram of Tritium Monitor AEP10101*

*Taken from Osborne and Cowper, AECL 2604
Due to the very low energy of the beta from the decay of tritium, the air must be counted internally. In this monitor the air is passed through the outer chamber where it is detected along with the gamma background. A second inner chamber is sensitive only to the gamma background. The difference between the two chambers gives the tritium content of the air.

ACTIVATED CORROSION PRODUCTS

Table II gives a list of several of the possible radioactive corrosion products in a reactor system along with their modes of production and decay. They are listed according to half life and it can be seen that they cover a very wide range of half lives.

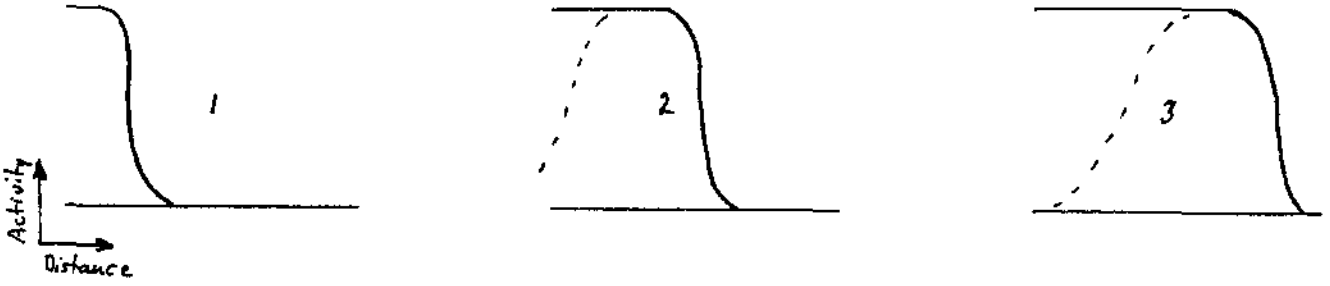
TABLE II

Activated Corrosion Products

Product	Reaction	Half Life	Decay Modes	Principal Gammas (MeV)
^{28}Al	$^{27}Al(n,\gamma)^{28}Al$	2.3 min	β^-,γ	1.78
^{56}Mn	$^{55}Mn(n,\gamma)^{56}Mn$	2.58 hrs.	β^-,γ	0.847(99%)
^{64}Cu	$^{63}Cu(n,\gamma)^{64}Cu$	12.8 hrs.	EC,β^+ β^-,γ	0.511(38%)
^{51}Cr	$^{50}Cr(n,\gamma)^{51}Cr$	28 days	Ec,γ	0.325(9%)
^{59}Fe	$^{58}Fe(n,\gamma)^{59}Fe$	45 days	β^-,γ	1.095(56%) 1.292(44%)
^{124}Sb	$^{123}Sb(n,\gamma)^{124}Sb$	60 days	β^-,γ	0.603(100%) 1.692(50%)
^{58}Co	$^{58}Ni(n,p)^{58}Co$	71 days	β^+,γ	0.511(200%)
^{54}Mn	$^{54}Fe(n,p)^{54}Mn$	300 days	EC,γ	0.835(100%)
^{60}Co	$^{59}Co(n,\gamma)^{60}Co$	5.3 yrs.	β^-,γ	1.173(100%) 1.333(100%)

The great difficulty encountered with the activated corrosion products is their tendency to "plate-out" on various metal surfaces. This means that the various valves, flanges, pipes, etc will tend to become very radioactive and hence make servicing difficult. It is ^{60}Co perhaps that gives the greatest problem due to its long half life and high energy gammas.

The exact nature of the "activity transport problem" is not fully understood. At the present time, it appears that the mechanism is by some type of chromatographic process. Some people feel that the surface of the metal behaves as though it were an ion-exchange resin. The corrosion products as they are carried through the heat transport system are repeatedly adsorbed and and desorbed from the walls. Starting with clean metal walls, a band of activity advances with the flow until the walls are fully coated as shown in Figure 3 (solid line).



solid line - continuous production of activity
broken line - short-term production of activity
1,2, and 3 are after three successive time intervals.

Figure 3

Transport of Activity on Clean Metal Walls

If left flowing sufficiently long without any new production of radioactive material, it is possible that the walls could be washed clean again as depicted in Figure 3 by the broken line. Production does not, however, stop. It continues.

The various corrosion products have different affinities for different wall materials; thus, the transport of these various activities differs with the different wall materials.

FISSION-PRODUCT ACTIVITIES

Ideally no fission-product activity should reach the coolant system as the fuel is "safely" clad within a zircaloy sheath. Fission product activities are; however, released both as a result of surface contamination of the fuel bundles with uranium and as a result of rupture of the fuel bundles. It is the latter source that is most important.

Fission is a very complex process liberating several hundreds of radioactive species. For this discussion we will divide them into groups according to both their chemical behaviour and their half lives. The list will be restricted to those activities easily observable from UO_2 fuel failures.

(a) Gaseous Fission Products

These products (shown in Table III) include the various Kr and Xe isotopes. These gases move with the coolant. They are not trapped on the ion-exchange columns, but remain in solution in the water. They undergo beta decay to produce Rb and Cs isotopes which will stop on the cation resins as well as the walls.

$$^{87}Kr \xrightarrow{\beta^-} {}^{87}Rb \xrightarrow{\beta^-} {}^{87}Sr$$

$$^{137}Xe \xrightarrow{\beta^-} {}^{137}Cs \xrightarrow{\beta^-} {}^{137}Ba$$

A small pinhole or hairline crack might release activity only when under stress, eg, during a power change or a fuel shuffling. (Refer to Figure 5(b)). A large crack or hole could release activity continuously. The length of time required for fission-product gases to diffuse through the fuel and out into the system would be shorter with a large leak than a small leak. The large leak would likely show the presence of both long and short-lived activities while the small leak would show only the long-lived activities as the short-lived activities would probably decay before escaping into the coolant.

TABLE III

Gaseous Fission Products

Nuclide	Half Life	Principal Gamma (MeV)
^{87}Kr	76 min	0.403 (84%), 0.850 (16%) 2.57 (35%)
^{88}Kr	2.8 hr	0.191 (35%), 0.850 (23%) 2.19 (18%), 2.4 (35%)
^{85m}Kr	4.4 hr	0.150 (74%), 0.305 (13%)
^{138}Xe	14 min	0.260 (100, 1.78 (66) 2.02 (58)
^{135m}Xe	15.6 min	0.527 (80%)
^{135}Xe	9.15 hr	0.250 (91%), 0.61 (3%)
^{133}Xe	5.65 days	0.081 (37%)

(b) Halogen Fission Products

The halogens are volatile and fairly easily released from the fuel. They have a tendency to be captured by the anion ion-exchange resins; and thus, removed from solution. The halogen activities (shown in Table IV) fall into two very distinct groups: A mixture of short-lived Br and I isotopes having half lives less than a minute that undergo a rather unique form of radioactive decay - delayed neutron emission and a mixture of longer-lived I activities that undergo the more conventional β-γ decays.

TABLE IV

Halogen Fission Products

Nuclide	Half Life	Principal Gamma (MeV)
Short-lived group		
^{87}Br	55.8 sec	Delayed Neutron Precursors
^{88}Br	15.9 sec	
^{137}I	23 sec	
Long-lived group		
^{132}I	2.4 hr	0.52 (20%), 0.673 (144%) 0.773 (89%)
^{134}I	52 min	0.85 (95%), 0.89 (65%) 1.15 (10%)
^{135}I	6.7 hr	0.86 (11%), 1.14 (37%) 1.28 (34%), 1.46 (12%) 1.72 (19%), 1.80 (11%)
^{133}I	21 hr	0.530 (90%)
^{131}I	8.05 days	0.364 (82%)

(c) Depositing Fission Products

These nuclides are removed from the coolant mainly by deposition on the walls. They are not likely to be recirculated through the reactor system as they "plate-out" on the walls; thus, effectively making the reactor itself a clean system. The sequence in Figure 4 would consist only of the first step as the cooling water re-entering the core would no longer be carrying any radio-activitiy with it; the radioactivity would be on the walls of the boiler, etc.

Table V gives a list of several of the longer-lived depositing fission products used to locate failed fuel elements. As these materials deposit on the walls, their recycle time in the system is essentially infin-ite. They thus are capable of giving a good signal com-pared to background.

^{239}Np is included as a fission product in the list on Table V. It is not really a fission product, but a neutron activation product of the ^{238}U in the fuel.

$$^{238}U + n \longrightarrow {}^{239}U \xrightarrow{\beta^-} {}^{239}Np \xrightarrow{\beta^-} {}^{239}Pu$$

The end product of the beta decay chain is the 2.4 x 10^4yr ^{239}Pu which undergoes fission when bombarded by thermal neutrons ($\sigma_{(n,f)}$ = 750 barns). The ^{239}Np is in-cluded in this discussion because it behaves as though it were a fission product.

TABLE V

Depositing Fission Products

Nuclide	Half Life	Principal Gamma (MeV)
Short-lived group		
^{132}Te	78 hr	0.230 (90%)
^{99}Mo	67 hr	0.181 (7%), 0.740 (12%)
^{99m}Tc		0.140 (90%)
^{239}Np	2.35 days	0.106 (23%), 0.228 (12%) 0.278(14%),+^{239}Pu X-rays
Long-lived group		
^{95}Zr	65.5 days	0.724 (49%0, 0.756 (49%)
^{95}Nb		0.765 (100%)
^{103}Ru	40 days	0.497 (88%), 0.610 (6%)
^{106}Ru	1.0 year	none
^{106}Rh		0.512 (21%), 0.622 (11%)
^{141}Ce	32.5 days	0.145 (48%)
^{144}Ce	285 days	0.34 (11%)

The greatest problem in detecting faulty fuel is the fact that in each pass through the reactor, the water picks up radioactivity from the fault and spreads it out eventually contaminating the whole water supply, thereby, increasing the background. The mechanism of this process can be seen in Figure 4. The delayed-neutron activities, due to their short half lives decay away quickly compared to the recirculation time of the reactor system (typically 14 sec. per pass); thus, they do not build up a large background. Figure 4 shows the delayed neutron (dn) monitoring system used at Douglas Point while Figure 5(b) and (c) show the type of results obtained from such systems.

Figure 4

Flow of Radioactivity Through the Reactor System Following a Fuel Failure

Upper Figure - As the coolant flows across the defect to the boilers it carries the activity with it. The activity is indicated by the dots.

Lower Figure - After one additional complete cycle the flow of liquid comes back to the boiler. The activity has made one complete loop as well, but when it recenters the core it enters all of the fuel channels, not just the one from which it came. All of the channels now have a small background indicated by the lower density of dots. Additional passes would increase this background. Only a small portion of the flow passes through the clean-up loop; thus, only a small portion is removed by it.

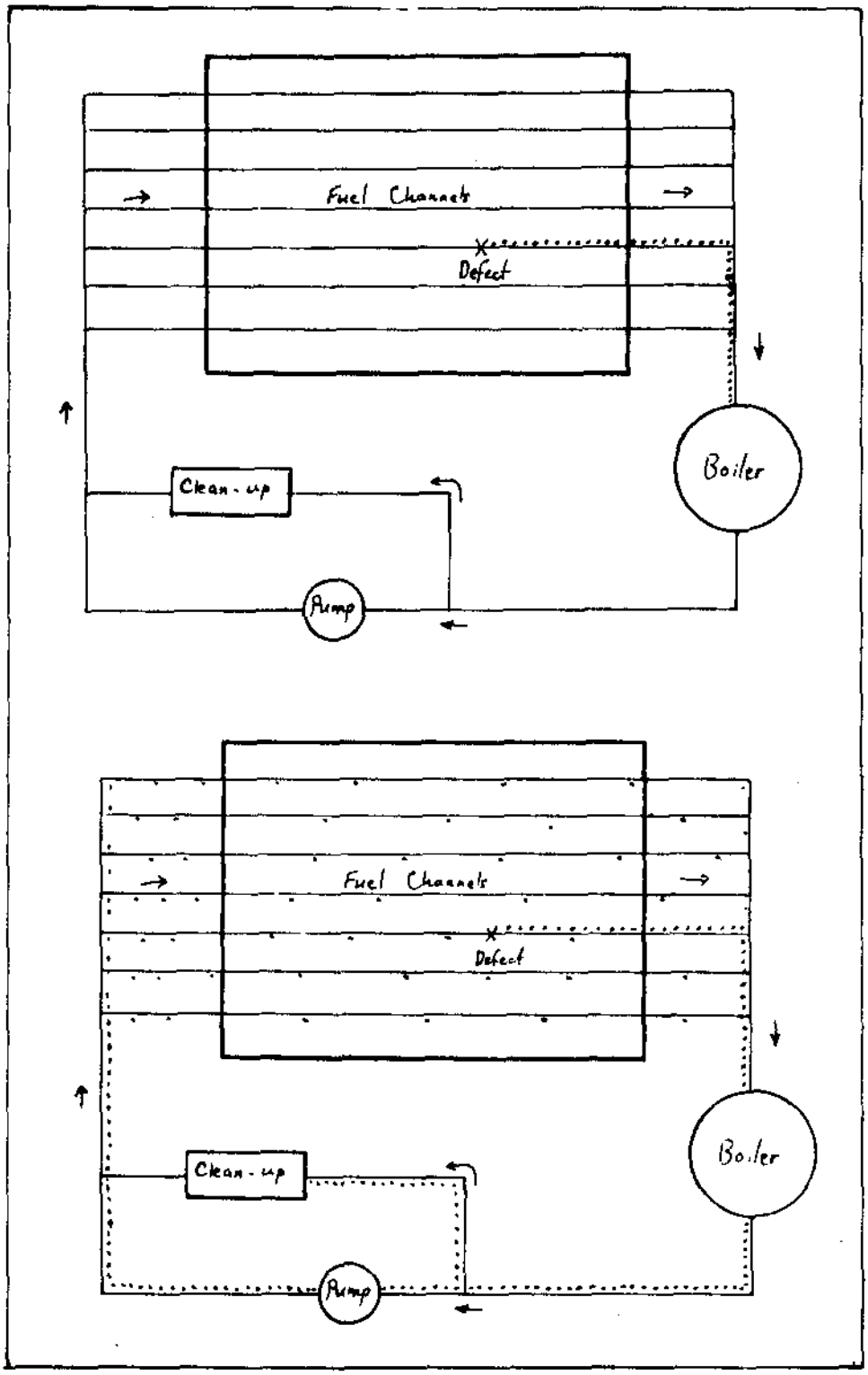

Fuel Channels
Defect
Boiler
Clean-up
Pump
Fuel Channels
Defect
Boiler
Clean-up
Pump

Figure 4

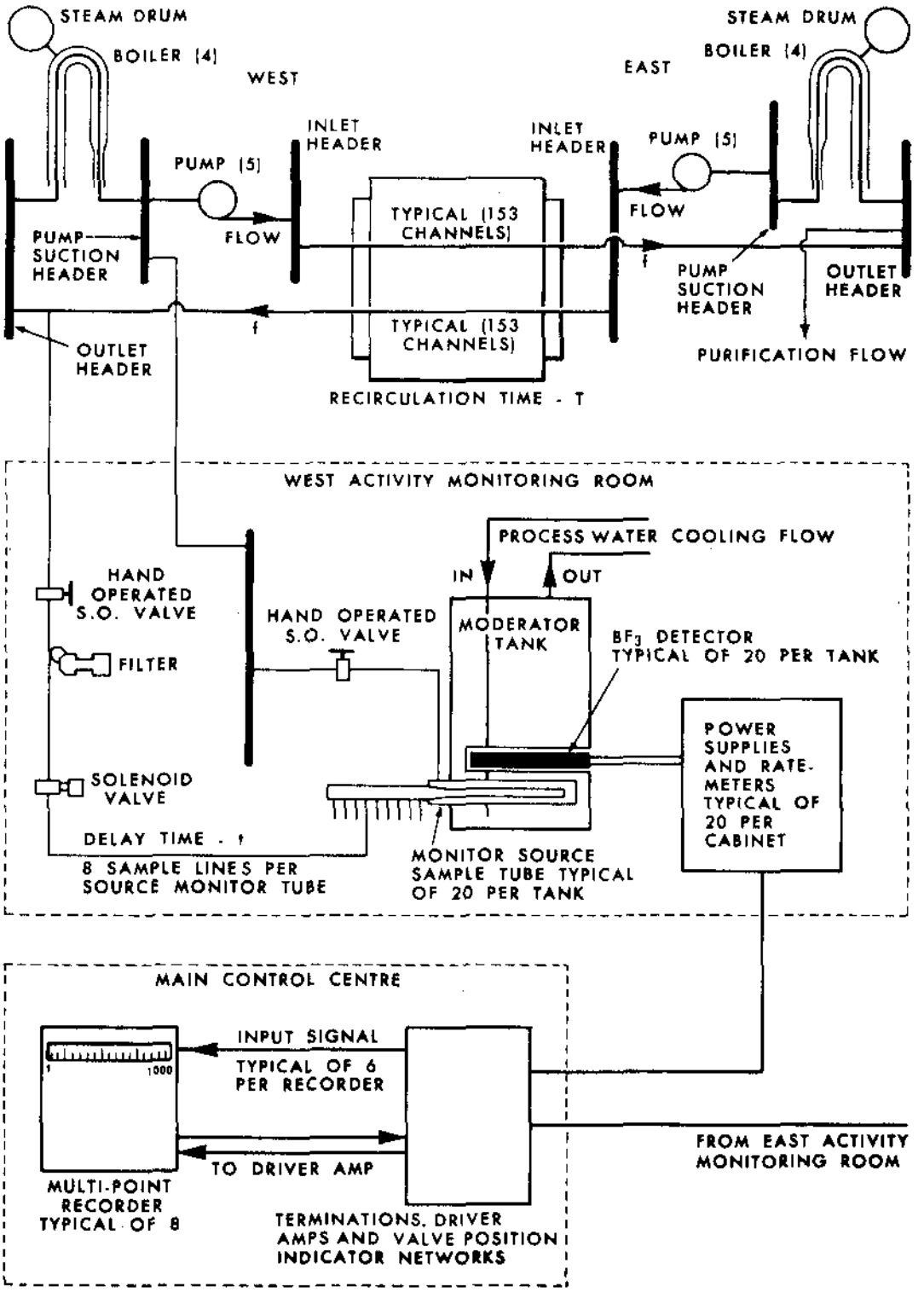


Figure 5(a)

Schematic of Delayed Neutron Activity Monitoring System

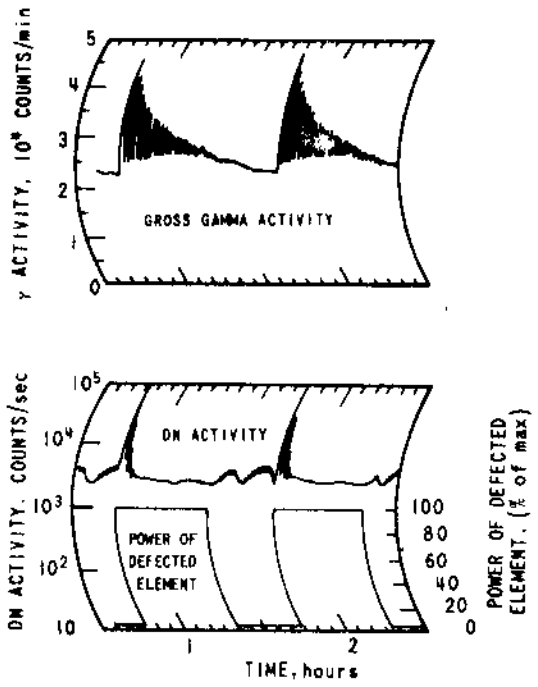


Figure 5(b)

Release of Delayed-Neutron and Gamma Activity During Power Cycles (AECL)

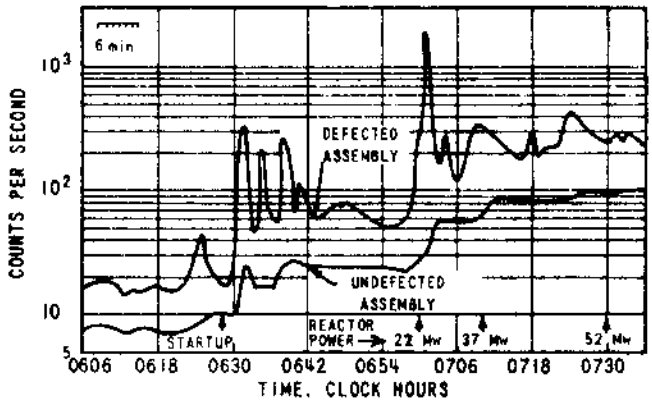


Figure 5(c)

Delayed-Neutron Monitoring of Failed Fuel Element (Shippingport PWR)

GAMMA RADIATION FROM THE HEAT TRANSPORT SYSTEM

A measure of the gross activity levels that collect on the metal surfaces shows that by far the biggest contribution comes from activated corrosion products. The contribution from fission products will remain small only as long as the fuel failures remain small. A broken fuel bundle could release very large levels of fission product activity plus ^{235}U which would contaminate the system and continue producing more fission product activity as it is bombarded. The early detection of failed fuel is necessary to prevent this from occurring.

Practice Exercises

1. List the three categories and sub-classes if applicable of activity transport in PHTS.

2. Give two examples of each category or sub-class and briefly describe the manner in which the species are removed or where they accumulate under normal circumstances.

   For the first exercise check the data in the notes. For the second, review your answer with a colleague or the course manager.

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