2. INTRODUCTORY REMARKS CONCERNING SOURCES, PRODUCTION AND BEHAVIOUR OF VARIOUS RADIONUCLIDES

2.1 SOURCES OF RADIOACTIVITY

The main sources of radioactivity in the coolant and cover gas come from:

i) Activation of the cover-gas and the coolant.

ii) Activation of dissolved or entrained impurities in the coolant as they pass through the core. These impurities include existing impurities, eg Li, U and entrained impurities due to corrosion.

iii) Activation of core components which subsequently enter the coolant by corrosion. The components of importance are: fuel cladding, core structural materials, control rods and wear resistant materials such as the stellites.

iv) Active fission products and fuel released from failed pins and any fuel released from contaminated fuel cladding surfaces.

v) Tritium released from ^{6}Li reactions in the coolant, ternary fission in fuel and activation of boron in control rod materials.

2.2 PRODUCTION OF VARIOUS ISOTOPES AND THEIR BEHAVIOUR IN LMFBRs

As the number of isotopes produced by neutron activation of the cladding and by fission in the fuel are too numerous to be included in this review this section deals specifically with those isotopes which, because of energy or half-life, are major contributors to circuit activity levels.

In general terms the production of all radionuclides takes place in the core of the reactor, ie within the primary sodium. Under certain circumstances the nuclides can separate from the coolant and distribute themselves between the coolant, the cover-gas, the cold-trap, and exposed steel surfaces, by processes involving precipitation, adsorption, diffusion, isotopic exchange and chemical reaction. These processes can be defined as follows:

Precipitation - The concentration of an element or compound in the bulk sodium is generally equal to its solubility at the lowest temperature of the sodium. Concentrations above this level will precipitate either in elemental form or as compounds with sodium. The cold-traps, which limit the concentration of oxygen and hydrogen in the sodium by crystallization of the sodium oxide and hydride on the mesh surfaces are also effective in removing radionuclides by precipitation processes.

Adsorption - Adsorption on component surfaces becomes important at reduced concentrations in sodium. In these situations elements can sometimes partition between the steel and the sodium and equilibrium is achieved when their respective chemical potentials are in balance. The amount of material deposited per cm^2 of surface at any particular temperature is related to a distribution coefficient K_D which is equal to the concentration of the species per unit area divided by its concentration in the sodium. The units of K_D are in cms and it is related to temperature through the expression $\ln K_D = a + H_D/RT$ where H_D is a heat term and T is the absolute temperature.

Diffusion - The diffusion of radionuclides into the stainless steel affects the level of contamination in the steel and is important for the decontamination of the component.

Isotopic exchange - This phenomenon does not appear to be important for describing the behaviour of radionuclides in LMFBRs.

Chemical reactions - The reaction with surface material and the formation of oxides is the main cause of irreversibility of the physical processes mentioned above.

2.2.1 Activation of the cover-gas

In the absence of other contaminants activity levels in the cover-gas come mainly from the reaction $^{40}Ar(n,\gamma)^{41}Ar$. The isotope is produced either by direct activation or when argon gas is entrained in the coolant and then passed through the core, or by the reaction $^{41}K(n,p)^{41}Ar$. The amount produced in the core is affected by safety requirements which limits the amount of entrained gas to < 1% of the core volume. Additional activity levels come from vapour and gaseous contaminants such as: ^{24}Na, ^{22}Na, ^{23}Ne, and possibly 3H (tritium).

2.2.2 Activation of the coolant and impurities in the coolant

The major active isotope in the coolant is ^{24}Na. The isotope, half-life 15hrs, is produced from the reaction ^{23}Na(n,γ)^{24}Na. Additional, but lower, activities come from ^{22}Na (half-life 2.6 a) produced by n,2n reactions and from ^{23}Ne (half-life 38 s) produced from ^{23}Na(n,p)^{23}Ne reactions. A comparison of the half-life of ^{24}Na and the transport time of the coolant around a primary circuit suggests that both sodium isotopes should be homogeneously distributed throughout the system. Also because of its short half-life the presence of any ^{24}Na in the sodium wetted film, covering components removed from the reactor, should only present short-term problems and over the long term the lower energy longer half-life ^{22}Na will be the dominant species.

The effect of impurities in the sodium on activity levels is not considered to be significant due to the tighter specifications now being applied to reactor grade sodium and estimated levels arising from residual impurities are less than those produced by ^{24}Na. The presence of Li and U impurities, although capable of producing extra tritium and fissile material, have negligible effects on activity levels compared to levels produced from ternary fission and failed pins respectively. It is also noteworthy that the combined values arising from pin contamination and uranium impurities in the sodium are generally used to set the background level for the Failed Fuel Pin Detection system.

Control of impurities during normal plant operation, by continuous or periodic cold trapping of the coolant, should ensure that increased levels of impurity will only occur if contaminants enter the circuit either during core changes, or from failed components or renewable items of plant. Evidence of contamination from other sources is highlighted by KNK.2 experience with ^{65}Zn, and spillage in other reactors from liquid metal seals containing Pb, Sn, Bi. For further details see Section 3.

2.2.3 Activation of core components

Most of the core components, which include fuel cladding, sub-assembly items, containments for control and neutron shield rods and support structures generally, are made from the Type 300 stainless steels. Variants which have been considered for fuel cladding are the high nickel austenitic alloys and for fuel element wrappers the high Cr ferritic steels. High cobalt alloys (stellites) have or are being used for rubbing pads and possibly pump bearings and although control rods are mainly made from B_4C, tantalum has been considered as a possible alternative.

The major radionuclides produced by neutron induced reactions in the core are: ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{58}Co, ^{60}Co and ^{182}Ta (see Table I). The relatively short half-lives of ^{51}Cr and ^{59}Fe (27.8 and 45 d respectively) and the low level of Ta in steels (< 0.1%) means that the activity levels produced by these isotopes are only of interest during normal plant operation and once the plant is shut-down the longer-lived isotopes ^{58}Co, ^{60}Co and ^{54}Mn become the radionuclides of concern.

Of these isotopes ^{58}Co comes from the nickel content of the steel. It has a modest half-life (71 d) and a high gamma energy (0.81 MeV) which makes it a major contributor to external dose rates once components have been removed from the reactor. ^{60}Co is derived solely from inactive Co and the ^{60}Ni content of alloy steels. Stellite wear pads can also be a major source of this isotope. It has a long half-life (5.27 a) and therefore the build-up of released activity to the coolant from this source is slow and never comes to equilibrium during typical fuel element lifetimes. By contrast, ^{54}Mn (half-life: 300 d) with a gamma energy of 0.84 MeV approaches equilibrium at a faster rate and thus becomes a major contributor to activity levels in reactor circuits (see Ref 4).

2.2.4 Active fission products and fuel from failed pins

The chemistry of fuel failures and factors affecting the release of certain isotopes to the sodium coolant has been well documented by Potter, and Mignanelli (5,6,7). Consequently these aspects will not be discussed in detail in this review.

The amount of activity released from failed pins has been shown to depend upon the isotopic content of the fuel, distribution of fission products in the fuel prior to failure, the quantity of elements present in the fuel-clad gas gap, the rating of the pin and reactor history (8). Irrespective of defect size the noble gases (Xe, Kr) are released fairly quickly and their limited solubility in sodium ensures rapid removal to the

Table I : Corrosion Product Radionuclides, and comments [97]

Nuclide	Formation reaction(s)	Half-life (days)	Gamma energy (MeV)	Comments
^{54}Mn	^{54}Fe (n,p)$^{+}$ ^{55}Mn (n,2n)	313	0.84	The most prevalent nuclide. Eliminating all the others still leaves a significant problem.
^{60}Co	^{59}Co (n,g)$^{+}$ ^{60}Ni (n,p)	1913	1.17 1.33	Source is Co impurity in Ni- and Co-base wear pads, bearings, and hardfacing materials.
^{58}Co	^{58}Ni (n,p)$^{+}$ ^{59}Co (n,2n)	71	0.81	Use of a ferritic steel for fuel cladding eliminates most of ^{58}
^{59}Fe	^{58}Fe (n,g)$^{+}$ ^{59}Co (n,p)	45	1.10 1.29	
^{182}Ta	^{181}Ta (n,g)	115	1.12 1.22	Source concentration increases if Nb-bearing steel is used in the neutron flux.
^{65}Zn	^{64}Zn (n,g)	243	1.11	Source is Zn in sodium or from contamination by $ZnCrO_3$ rust proofing paint.
^{110m}Ag	^{109}Ag (n,g)	253	0.65 0.76 1.47	Suspected source is Ag impurity in nickel. This nuclide has not been observed in significant quantities in the U.S.
^{51}Cr	^{50}Cr (n,g)$^{+}$ ^{52}Cr (n,2n) ^{54}Fe (n,a)	28	0.32	Although substantial, low gamma energy makes ^{51}Cr transport inconsequential.

$^{+}$ dominant reaction ; g = gamma ; a = alpha

gas space. Fission products of high volatility behave in a similar manner and those elements of high yield, namely Cs, I, Ba and Sr can also be released during the initial stages of fuel clad failures.

The levels of activity circulating in the coolant depends upon the solubility behaviour of the various isotopes and in principle levels produced by the partly soluble I, Te, Sb, Sn, Ag radionuclides can be reduced by operation of the cold trap. Also it is possible that Cs may be retained in the trap in association with impurities, such as carbon.

However, if fuel pin failures become excessive, there is the possibility that the cold trap may become a relatively ineffective sink and plate-out of the various products may occur elsewhere in the circuit.

Isotopes of major concern are ^{134}Cs and ^{137}Cs which have half-lives of 2 and 30 a respectively. Both isotopes can produce radiation fields comparable to those produced by deposited corrosion products and the complete miscibility of Cs in sodium coupled with its high volatility points to complex behaviour in sodium systems. Other fission products such as the lanthanides, Zr, Nb, Ru, Mo and fuel products have very low solubilities in sodium and these elements along with the stable oxide formers Sr and Ba could be released either as sodium-metal oxides or fuel particles.

2.2.5 Tritium

Although tritium is unlikely to present a major hazard during normal plant operation, the element is of biological importance and therefore a knowledge of its transport behaviour in operating plant is required.

Tritium is produced by ternary fission in the fuel and activation of ^{10}B in control rods where the latter involves both thermal and fast neutrons;

$^{10}\mathrm{B}\ (n,\alpha)\ ^{7}\mathrm{Li} = {}^{7}\mathrm{Li}\ (n,\alpha n)\ ^{3}\mathrm{H}$ - Thermal neutrons (^{10}B) Fast neutrons (^{7}Li)

$^{10}\mathrm{B}\ (n,2\alpha)\ ^{3}\mathrm{H}$ - Fast neutrons

Its release is not dependent upon cladding failures as the isotope readily diffuses through cladding steels.

In operating plant the levels of tritium in the cover gas and the coolant are dependent upon the mode of operation of both the primary and secondary circuits and the respective cold-trapping procedures. The transport of the isotope and concomitant activity levels are influenced by thermo-chemical gradients set up in various parts of the system and interaction with hydrogen from other sources. See Section 6 for further details.