

9. SUMMARY OF THE BEHAVIOUR OF THE MORE IMPORTANT RADIONUCLIDES IN LMFBRs

Because of the extensive nature of the review it is difficult to summarize all the topics covered in the paper, consequently this summary deals very briefly with those radionuclides, such as the noble gases and tritium, which may cause leakage problems in operating plant and the long half-life nuclides which may affect maintenance procedures during shut-down. The topics are discussed in the order presented in the paper.

Except for the short half-life ^{23}Ne (38s) the more important gaseous radionuclides in the cover-gas are the fission gases ^{133}Xe and ^{135}Xe . So far neither isotope has given problems in operating systems and Xenon can be trapped effectively on charcoal, with and without nitrogen cooling, while other gaseous products, such as the short half-life radio-kryptons, can be accommodated in suitable delay tanks before they and the longer half-life (10.7 a) ^{85}Kr , are vented to atmosphere.

The amount of activity produced by the fission gases and other species from failed pins is minimised by putting limits on the number of acceptable failures. Gas-leakage from the plant is also prevented by ensuring that the reactor roof and the cover-gas purification system are leak-tight and the use of an efficient aerosol filter prevents ^{137}Cs entering gas-lines. All these procedures help to maintain low activity levels in the gas space and elsewhere in LMFBRs, and in terms of the complete fuel cycle, the levels of gaseous activity arising are still small when compared with levels handled in reprocessing plant.

In the absence of failed pins the main sources of activity in the sodium coolant comes from ^{24}Na , existing impurities in the sodium, tramp fuel and corrosion products. Although the presence of ^{24}Na has a marked influence on shielding requirements for operating systems its short half-life (15 h) makes it a negligible contributor to activity levels 10-14 days after shut-down. Also impurities in the sodium, which are at the ppm level, are not major contributors, unless, like ^{65}Zn , they originate from other sources, such as protective coatings and pump oil.

During plant operation cold-trapping procedures help to minimize the effect of residual and released soluble species, while the requirements of

the failed fuel pin detection systems puts tight limits on acceptable levels of tramp fuel. Cold trapping, in principle, can also remove soluble radionuclides such as ^{133}I from sodium and although estimates indicate that the amount of iodine in sodium is insufficient to cause precipitation of sodium iodide at typical primary circuit cold-trap temperatures, the review indicates that iodine has been identified in most reactor cold-traps, which suggests other mechanisms may be responsible for its presence in this part of the system.

In relation to shielding requirements for component handling, cleaning, decontamination and maintenance procedures the radionuclides of concern are those having long half-lives, high gamma-energy, high chain yields (fission products) or reaction rates (corrosion products) and high fractional release or corrosion rates. Radionuclides which exhibit some of these characteristics in LMFBRs are the isotopes ^{131}I , ^{132}Te + ^{132}I , $^{95}\text{Zr/Nb}$, $^{140}\text{Ba/La}$, ^{134}Cs , ^{136}Cs , ^{137}Cs , ^{58}Co , ^{60}Co and ^{54}Mn . Additional activities also comes from ^{22}Na and contaminants, such as ^{65}Zn and the alloying constituents of liquid metal seals (Pb, Sn, Bi, Hg). Estimates indicate however that activity levels caused by the presence of ^{22}Na in a drained sodium-wetted film, 75 μm thick, are much less than those produced by the other isotopes and although the behaviour of metal contaminants, such as ^{65}Zn , is discussed in the review, good design and materials management should eliminate them from future systems.

As regards the deposition behaviour of the other isotopes it is considered that the behaviour of the more soluble I and Te is controlled by their thermodynamic partitioning between the sodium coolant and the steel and the deposition behaviour of the Zr/Nb and Ba/La pairs (assumed to be present as insoluble oxides) is mass transfer controlled. However, because these radionuclides have short half-lives the activity levels arising will not increase after one years operation, if further fuel pin failures occur, and increased activity levels will only come from the longer half-life constituents ^{58}Co , ^{60}Co , ^{54}Mn and the isotopes of cesium.

The major activity levels which occur in the sodium and on components during shut-down originate, in the absence of failed pins, from the long lived corrosion products ^{54}Mn , ^{58}Co and ^{60}Co . In discussing the behaviour of these products in LMFBRs the review has shown that the mechanisms of release and deposition of radioactive corrosion products have been extensively studied in loop experiments and models and calculation codes are

now available which give reasonably good results, especially for the deposition behaviour of ^{54}Mn in heat-exchanger units. Supporting work to establish deposition levels on components also indicates that in PHENIX ^{54}Mn contributes to 98% of the contamination on the IHX and 97% on the pumps; while estimates for 1000 MWe pool-type reactors suggest that contamination by ^{54}Mn and ^{60}Co could reach levels of 1100 Ci and 340 Ci respectively. Uncertainty exists however regarding the behaviour of the cobalt isotopes and differences exist between the results obtained on EBR II, FFTF and PHENIX, where the cobalt isotopes are, either hardly detected or are of minor importance, and the results of BOR 60, where the surface activity of ^{60}Co is greater than values recorded for ^{54}Mn . It is thought that further work is required to resolve these differences and that the composition of the fuel cladding, especially with respect to nickel content, and the oxygen level of the sodium could be factors contributing to these different effects.

Turning to the release of soluble and insoluble fission products from failed pins it is concluded that only ^{134}Cs and the long half-life (30 a) ^{137}Cs are important for the long-term contamination of the sodium and components. Although its relatively high volatility means that it can combine with sodium aerosols in the gas space, and possibly collect in gas-gaps and penetrations, the review indicates that cesium can be readily removed from sodium by carbonaceous materials and the section devoted to specialized traps indicates that carbon traps for cesium removal have been used with some success in reactor systems. This suggests that, although no suitable model is yet available to describe cesium behaviour in LMFBRs, there are ways of removing it from the sodium.

Although the acceptable number of failed pins in commercial reactors has not been resolved, Soviet experience in BOR-60, with the reactor operating with 0.1% failed pins, has shown that activity levels arising from fission products were no greater than the accumulated activity from corrosion products when measurements were taken at the next shut-down. It also seems that no particular problems have arisen during the decontamination of components removed from LMFBRs, unless oil leakage from mechanical pumps has caused carbonaceous material to collect on sodium surfaces adjacent to components. In these situations high levels of radiation may occur due to the trapping of cesium and subsequent decontamination may have to be done by mechanical means.

The measurement of tritium in operating reactors has been undertaken at a number of reactor sites using methods outlined in the review; and recent assessments of existing models, along with measurements made on PHENIX, leads to the conclusion that if the earlier KUMAR model is used to predict the distribution of tritium in LMFBRs, then adjustments are required to bring predicted values in line with recent reactor measurements. According to PHENIX data tritium levels are not as high as expected even when the permeability constants for tritium and hydrogen transfer through stainless steel are the same and the cold-trap is out of operation, and the model can only accommodate these lower values either;

- by reducing the tritium source term, which corresponds to a fission yield of 1.5×10^{-4} , by a factor of 2 or 3, or
- if the tritium source term is correctly evaluated, by increasing the hydrogen source term to increase the efficiency of cold-trapping and thus tritium removal, and by making the permeability coefficients for tritium transport through the IHX's, greater than the hydrogen value, so that in both cases tritium is more easily removed from the primary system.

Finally, although concerns have been expressed about plutonium dispersion both in the coolant and on components and its effects on the burst fuel-pin detection system, it seems that its slow release, along with the solid fission products from defected fuel pins, keeps it below detection levels both in the sodium and on components removed from reactors. Because of this, and the lack of major problems arising from activation, corrosion, and fission products; dose rates received by personnel are on the low side and releases to the environment are well below those occurring in water reactors.