

### 3. THE BEHAVIOUR OF ACTIVATION PRODUCTS IN LMFBRs

This section discusses the behaviour of the activation products in more detail. It includes their formation, behaviour and the levels observed in operating systems.

#### 3.1 BEHAVIOUR IN THE GAS PHASE

In the absence of fuel failures  $^{23}\text{Ne}$  ( $t_{1/2} = 38$  s) and  $^{41}\text{Ar}$  ( $t_{1/2} = 1.8$  h) are the most important nuclides in the cover gas during reactor operation. After shutdown, fission gases and their decay products as well as nuclides evaporated from the sodium ( $^{24}\text{Na}$ ,  $^{22}\text{Na}$ ,  $^{65}\text{Zn}$ ,  $^{137}\text{Cs}$ ) have also to be considered.

##### 3.1.1 $^{23}\text{Ne}$

$^{23}\text{Ne}$  is formed from  $^{23}\text{Na}$  by (n,p) reactions. The resulting radioactivity in Na is very high and levels of 26 GBq/kg have been estimated at Phenix. However, the  $^{23}\text{Ne}$  activity in the cover gas is still much reduced because of the time required to coalesce atoms into bubbles and to dis-entrain the bubbles when they have grown to a critical size. De-gassing ( $t_{1/2}$ ) times of the order of one hour have been reported from reactor measurements (see 5.3.1), and typical reactor activities are of the order of 10 TBq/m<sup>3</sup> (see Table II). During reactor operation care has to be taken to ensure that the reactor roof is gas-tight and that unavoidable leakages are sufficiently delayed to minimize the escape of any  $^{23}\text{Ne}$ . Also the gas measurement system must be equipped with a delay line to reduce the background level due to  $^{23}\text{Ne}$ , although this in itself makes it difficult to measure accurately the true behaviour of other short-lived gaseous fission products.

##### 3.1.2 $^{41}\text{Ar}$

$^{41}\text{Ar}$  can be formed from the following routes:

- (n, gamma) reaction with the  $^{40}\text{Ar}$  of the cover gas by direct activation
- (n, gamma) reaction with the  $^{40}\text{Ar}$  of the cover gas entrained into the sodium
- (n, p) reaction with  $^{41}\text{K}$  present in sodium as an impurity ( $^{41}\text{K}$  accounts for 6.9% of the natural potassium).

Table II : Maximum activity levels measured in the cover gas of LMFBRs  
(in GBq/m<sup>3</sup>)

reactor	isotope half-life ref	<sup>23</sup> Ne 38 s	<sup>41</sup> Ar 1.8 h	<sup>133</sup> Xe 5.2 d	<sup>135</sup> Xe 9 h	<sup>85m</sup> Kr 4.5 h	<sup>88</sup> Kr 2.8 h	<sup>87</sup> Kr 76 min
FFTF	[169]	74000	15	1900	800	200	200	75
KNK II	[106],[170]		7	1240	134	11	13	6
PHENIX	[9],[143]	7000	1.5	2500	1500	300		200
BR-5	[171]	26000	5.5	7400	4400		3700	1800
BOR-60	[172]			25000	2000	200	70	
BN-350	[149]	18000	0.74	4000				
BN-600	[109]		5.3	4800				

The importance of each route depends on the reactor design and the specified purity of the nuclear grade sodium. At Rapsodie all the <sup>41</sup>Ar activity in the cover gas could be explained by direct activation, whereas extrapolating to Phenix led to an under-estimate of the <sup>41</sup>Ar activity by three orders of magnitude and gas entrainment was thought to be the answer. However, since the cause of the gas entrainment could not be found, calculations were performed by taking into account the activation of <sup>41</sup>K. These estimates indicated that the level of potassium impurity (about 200 µg/g) was sufficient to explain entirely the measured <sup>41</sup>Ar activity in the cover gas (9,10).

The degassing constant, was obtained by analysing the activity evolved during a fast shutdown of the reactor. The measured  $t_{1/2}$  was 48 min, which fits well with the value for <sup>23</sup>Ne activity in the cover gas and suggests that in both cases activation of dissolved atoms, rather than entrained gas bubbles, is the rate-controlling process (9).

### 3.1.3 <sup>37</sup>Ar

This long lived isotope ( $t_{1/2} = 35$  d) disappears by electronic capture and causes no trouble during reactor operation, nor during shutdown, since the

activity level is low ( $0.2 \text{ GBq/m}^3$  at Phenix (10)). Its measurement is not easy and necessitates gas sampling.

Three formation routes are possible:

- (n, gamma) reaction with  $^{36}\text{Ar}$  of the cover gas entrained into the sodium ( $^{36}\text{Ar}$  accounts for 0.34% of the natural argon)
- (n, 2n) reaction with  $^{38}\text{Ar}$  of the cover gas entrained into the sodium ( $^{38}\text{Ar}$  accounts for 0.06% of the natural argon)
- (n, alpha) reaction with  $^{40}\text{Ca}$  present as an impurity in the sodium.

To minimize the risk of plugging by CaO precipitates, the calcium content of nuclear grade sodium is very low (typically  $5 \mu\text{g/g}$  in France). However, although this reaction could account for the measured activity in Phenix  $^{37}\text{Ar}$  can form by other routes, which supports the argument for limiting the amount of argon entrained in the sodium (9).

### 3.2 BEHAVIOUR IN THE SODIUM

#### 3.2.1 $^{24}\text{Na}$

The major radioactivity in the sodium of an LMFBR is due to  $^{24}\text{Na}$  (about  $0.5 \text{ TBq/m}^3$ , see Table III) and calculations for the biologic protection of the reactor are based on this nuclide. However because its half-life is only 15 h, it is of less importance during reactor shutdown and only nuclides of half-lives of at least several days are of interest during this period.

#### 3.2.2 $^{22}\text{Na}$

Because of its long half-life (2.6 a)  $^{22}\text{Na}$  radioactivity increases in sodium with reactor thermal power history. Typical reactor values are around  $20 \text{ MBq/kg}$  (see Table III). After a long reactor shutdown, its radioactivity in the aerosols deposited in the cover gas plenum can make maintenance work difficult, especially in regions such as annuli and crevices and Stamm (11) has reported a much higher specific  $^{22}\text{Na}$  activity from sodium deposits at the rotating plug of KNK than in the bulk sodium.

Table III : Maximum activity levels measured in the sodium of LMFBRs

isotope		<sup>24</sup> Na	<sup>22</sup> Na	<sup>65</sup> Zn	<sup>124</sup> Sb	<sup>131</sup> I	<sup>137</sup> Cs	<sup>134</sup> Cs
half-life		15 h	2.6 a	244 d	60.3 d	8 d	30 a	2.2 a
reactor	ref \ units	TBq/kg	MBq/kg	MBq/kg	MBq/kg	MBq/kg	MBq/kg	MBq/kg
EBR-II	[27]	0.1	5		21	11	13	1.6
FFTF	[27]	0.41	20		2.6E-2	1.8	8.1	5.5
KNK II	[13],[28]	2.5	25	6.4	1.4	38	34	
PFR	[104],[24]		14				11.7	2.2
RAPSODIE	[10]		40				55	
PHENIX	[10],[143]		22				1.7	0.33
BOR-60	[25],[109]		22	0.74		103	777	66
BN-350	[149]	0.37	28					
BN-600	[109]	0.75	26		0.15	13	150	50

### 3.2.3 <sup>65</sup>Zn

#### Behaviour of zinc in sodium

<sup>65</sup>Zn ( $t_{1/2} = 244$  d) has been observed in various reactors, although its source is not always known. In KNK, the main source was probably a zinc chromate protective coating used to cover the external surfaces of the ferritic components prior to assembly (11,12). A further possible source was an oil leakage into the primary sodium from one of the primary pumps; the oil being stabilised with zinc dithiophosphate. The possibility that other sources may have existed in the reactor is referred to in Ref (13). At DFR, the source was assumed to be due to corrosion of zinc containing alloys (14).

The behaviour of <sup>65</sup>Zn in sodium is thought to be similar to the behaviour of <sup>137</sup>Cs (see 5.3.2) in that its solubility in sodium is high (100 µg/g at 220°C (14) and no stable compounds in sodium are formed at low chemical activities of zinc in sodium. It appears to plate out on steel surfaces by

a chemi-sorption process and its diffusion into stainless steel is also similar to that of  $^{137}\text{Cs}$ , with bulk diffusion coefficients, measured by Sagawa et al, ranging from  $4.0 \times 10^{-14} \text{ cm}^2 \cdot \text{s}^{-1}$  at  $260^\circ\text{C}$  to  $8.8 \times 10^{-14} \text{ cm}^2 \cdot \text{s}^{-1}$  at  $498^\circ\text{C}$  (15-20).

The behaviour of  $^{65}\text{Zn}$  on steel surfaces has been investigated in the Harwell Active Mass Transfer Loop by Newson et al (21). The deposition was found to be governed by the temperature-dependent solubility of zinc in steel (zinc solubility increases when temperature decreases) and the solubility of zinc in sodium. Enhanced levels of zinc were found on particles deposited on pipework and this was attributed to the high solubility of zinc in iron and nickel (about 2000 times more in Ni than in Fe at the operating temperatures). 65% of the deposited zinc was also found in the cold trap which operated at  $150^\circ\text{C}$ . It therefore seems that in reactor systems zinc deposition could be dictated by both sodium temperature and hydraulics and high levels of zinc may be found in the cold trap and in the reactor vault, due to settling of particles.

Experiments carried out at the Risley Laboratories have also shown that zinc concentrations in the vapour phase above a sodium pool heated to  $460^\circ\text{C}$  were 70 times higher than in the bulk sodium. As a consequence,  $^{65}\text{Zn}$  evaporates strongly and is transported by sodium aerosols. This contrasts with the enrichment factor of 10 for the sodium wetted film formed on steel plates suspended above the sodium pool in a refluxing sodium vapour environment. In these experiments the loop system buffered the zinc concentration at 6-8  $\mu\text{g/g}$  of zinc and the excess of zinc was deposited in the cooler parts of the system, but not necessarily in the cold trap (22).

#### Reactor measurements of $^{65}\text{Zn}$

Measurements made at PFR where the coolant content had reached about 2.5  $\mu\text{g/g}$  of zinc showed surface zinc levels of  $3 \mu\text{g/cm}^2$  on steel components withdrawn from the primary sodium which is some 200 times the amount expected in sodium wetted films covering such items. Higher levels were also found on pure nickel (23). The presence of  $^{65}\text{Zn}$  in the sodium appeared to follow no particular trend and the operation of the primary cold trap loop from mid 1982 to May 1984 appears to have reduced the  $^{65}\text{Zn}$  level in the primary sodium to below the detection limit (24).

At KNK II,  $^{65}\text{Zn}$  activity (6.4 MBq/kg at 220°C) is the most important residual isotope after  $^{22}\text{Na}$  when the reactor is shutdown and the temperature is decreased: (see Table III). Surface levels measured at the outlet of the IHX gave values of 5.62 kBq/cm<sup>2</sup> and on the Colmonoy coatings of valves,  $^{65}\text{Zn}$  activity was 10 times higher compared to the uncoated ferritic steel surfaces (values up to 30 times were obtained with  $^{54}\text{Mn}$ ) (13). An enrichment factor of 280 (relative to  $^{22}\text{Na}$ ) was measured in the sodium of the rotating plug and 65% of the  $^{65}\text{Zn}$  was found in the cold trap (13). Similar trends have also been reported at BOR-60 where the reported  $^{65}\text{Zn}$  activity in sodium was 0.74 MBq/kg (25) and 85% of the  $^{65}\text{Zn}$  was in the cold trap (26).

#### 3.2.4 $^{124}\text{Sb}$ and $^{125}\text{Sb}$

Both these isotopes are fission products and activation products of impurities in the sodium.

##### Behaviour of $^{124}\text{Sb}$ in sodium

Although the solubility of antimony in sodium is high, its deposition on nickel or steel surfaces is negligible. It is found to co-precipitate with sodium oxide in cold traps and therefore its behaviour is very dependent upon the oxide impurity level in the sodium.

$^{124}\text{Sb}$  ( $t_{1/2} = 60.3$  d) has been reported at Phenix and it was found in EBR-II sodium in 1984. In both instances its presence was due to a leaking Sb-Be neutron source used at the start-up of the reactor. At KNK, its presence was attributed to the use of 'Molykote' (a lubricant containing antimony trisulfide) for the bearing rings at the bottom of the sub-assemblies (13).

##### Reactor measurements of $^{124}\text{Sb}$

Measurements taken at EBR-II in 1984 showed a peak value of 21 MBq/kg before it decayed away, whereas transitory radiation levels measured near the primary cold trap, due to  $^{124}\text{Sb}$ , were as high as 0.3 Gy/h (27). At KNK II the reported activity for  $^{124}\text{Sb}$  was 1.4 MBq/kg and 4.7 MBq/kg for  $^{122}\text{Sb}$  ( $t_{1/2} = 2.7$  d) (28).

### 3.2.5 Other minor activation products

$^{110m}\text{Ag}$  - The source of  $^{110m}\text{Ag}$  is not always known. The highest reported activity level was measured at BOR-60 following the corrosion of structural material containing 2  $\mu\text{g/g}$  silver which produced a value of 8.5 MBq/kg (25). The isotope appears to be completely miscible in sodium and shows no segregation or deposition effects.

$^{113}\text{Sn}$  - The leakage of the tin-bismuth seal of the rotating plug into the primary sodium at EBR-II and Rapsodie has been the cause of tin activation ( $^{113}\text{Sn}$ -In  $t_{1/2} = 119$  d) in both these reactor systems (29).

$^{210}\text{Po}$  - The major activation product of bismuth is alpha-emitting  $^{210}\text{Po}$  ( $t_{1/2} = 138.4$  d). The isotope is of particular concern where hands-on maintenance of sodium-wetted components is undertaken. Fortunately, the deposition of bismuth in the cold trap keeps the circulating inventory in the sodium at a low level and restricts  $^{210}\text{Po}$  build up. At FFTF, the alpha energy measured fitted 3 nuclides:  $^{210}\text{Po}$ ,  $^{232}\text{U}$  and  $^{243}\text{Am}$ . The presence of traces of bismuth indicated that the alpha activity could be attributable to  $^{210}\text{Po}$  and not to fuel release.

$^{203}\text{Hg}$  - This isotope was reported as an activation product of a mercury impurity in the DFR coolant. Its solubility and volatility suggests that it may behave like  $^{65}\text{Zn}$ , or the soluble fission products Cs, I (30).

$^{64}\text{Cu}$  - The behaviour of copper was investigated at EBR-II using  $^{64}\text{Cu}$  as a tracer. Its short half-life (12.8 h, comparable with  $^{24}\text{Na}$ ) is probably the reason why it is not observed in operating reactors.

$^{182}\text{Ta}$  -  $^{182}\text{Ta}$  is reported only in a few reactors and is only briefly mentioned in the section devoted to the behaviour of corrosion products. It is the (n, gamma) activation product of  $^{181}\text{Ta}$  and it is part of the niobium added to stabilized steels. Because of its long half-life (115 d) and its hard gamma-radiation, it contributed much to the dose rate in the primary cell during maintenance periods at KNK I(13). Also it produced the highest percentage level of corrosion product activity when the wash-waters from the KNK fuel element cleaning facility were analysed. Values obtained were:  $^{182}\text{Ta}$  (34.4%) followed by  $^{54}\text{Mn}$  (17.2%),  $^{58}\text{Co}$  and  $^{60}\text{Co}$  (13.4% each),  $^{51}\text{Cr}$  (9.6%),  $^{65}\text{Zn}$  (7.6%) and  $^{59}\text{Fe}$  (3.8%).