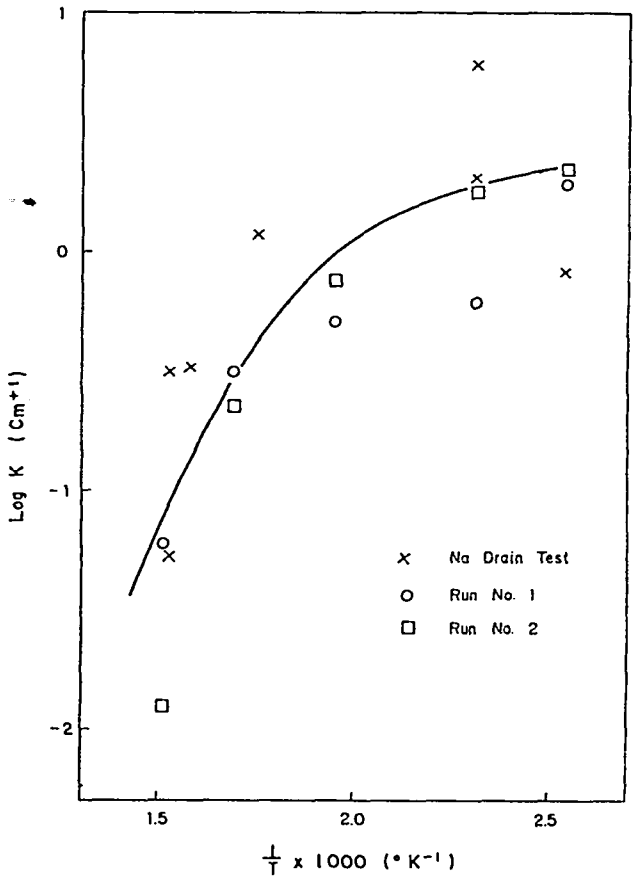

Fig. 13 Te-132 K-VALUE VS RECIPROCAL TEMPERATURE

11. Design Considerations of Fission and Corrosion Product in Primary System of MONJU

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Abstract

General influence of fission and corrosion products in primary system on MONJU plant design is reviewed. Various research and development works are now in progress to decrease the generation rate, to remove the products more effectively and to develop the methods of evaluation the behavior of radioactive products. The inventory and distribution of fission and corrosion products in the primary circuit of MONJU are given. The radiation levels on the primary components are estimated to be several roentgens per hour.

1. Introduction

The behavior and inventory of fission and corrosion product in the primary circuit of LMFBR has strong influence on the design of the plant and how it is operated. The influence of the products has two aspects; one is on the primary sodium system and the other is on the argon cover gas system. Related specific areas in each aspects of plant design are as follows:

I) in primary sodium system design
   1. shielding design for piping and components
   2. maintenance and repair considerations of piping and components
   3. design of liquid radioactive waste disposal system which process mainly the washing waste effluent from decontamination facilities for spent fuel assemblies and sodium components
   4. design of delayed neutron detecting system
   5. design measures for leaked sodium containing radioactive materials and hazard evaluation

II) in argon gas system design
   1. design of argon purification system
   2. design of failed fuel detection system

3. design of ventilation system and shielding in discharged fuel handling area
4. design measures for leakage of radioactively contaminated argon gas and hazard evaluation

The primary circuit of fast reactor differs from that of thermal reactors in the following aspects.

1. primary circuit of structure against leakage of chemically reactive sodium
2. good fission product retaining ability of sodium
3. high induced radioactivity of sodium
4. formation of radioactive corrosion products that are not familiar with thermal reactors through threshold energy reactions.

Fundamental design features of primary circuit

Since induced radioactivity of sodium is higher, the estimated radiation level in primary sodium system of MONJU is as follows:

	^{24}Na	^{22}Na
reactor operation	~10^5r/h	~10r/h
twenty days after shutdown	~10mr/h	~10r/h
sodium drainage	——	~100mr/h

It is clear that primary sodium circuit is not accessible during reactor operation and immediately after reactor shutdown. On sodium drainage, however, maintenance works can be performed for limited working time.

Though the induced activity of sodium is the major component of radioactive source term during reactor operation and for about twenty days after reactor shutdown, fission and corrosion products which are deposited on the piping and components become the main radioactive source of drained primary circuit. Even for the case of access to the primary circuit components after sodium drain, it is desirable to minimize the deposition of fission and corrosion products as little as possible in order to make the permissible time of the access longer.

Considering these effects of fission and corrosion products on plant system design, following are the related basic requirements in plant design.

(1) decreasing the rate of fission and corrosion products release into sodium
(2) removal of fission and corrosion product from primary circuit
(3) accurate evaluation of inventory and distribution of radioactive products in primary system

Research and Development Program

Research and development works with many varieties are now in progress to realize the requirements mentioned previously. They include;

(1) decreasing fission and corrosion product release

fission product

- o strict quality assurance procedures against fuel clad failures

corrosion product

- o limited use of stellite in primary components
- o development of stainless steel containing less cobalt which is main source of long life nuclide ^{60}Co.

(2) removal of fission and corrosion product from primary circuit

- o obtaining deposition distribution profile in cold trap and removal efficiency for radioactive impurities
- o development of argon cover gas purification system.

(3) evaluation of inventory and distribution of radioactivity in primary circuit

fission product

- o measurement of fission product release rate from irradiated fuel pellet
- o measurement of fission product distribution in experimental sodium loop.

corrosion product

- o fundamental experiments to investigate corrosion mechanism and corrosion rate
- o measurement of corrosion product distribution in small sodium loop

synthetic evaluation

- o availability of calculating code of radioactivity distribution in primary circuit after long term reactor operation
- o comparison between measurements and calculations on the experiments performed in fast experimental reactor JOYO which is scheduled to be critical next year.

2. General description of MONJU primary system

MONJU is a loop type prototype LMFBR of 300MWe. Its main characteristics and the flow diagram of the primary cooling system are given in Table 1 and in Fig. 1 respectively.

Table 1. Principal characteristics of MONJU primary system

Reactor Thermal Power	714 MWt
Electrical Power	300 MWe
Reactor Type	Loop type
Average Fuel Burnup	80,000 MWD/t
Primary Heat Transport System	
Number of Loops	3
Flow Rate	5.12×10^6 kg/h - Loop
Reactor Outlet Temperature	529 °C
Reactor Inlet Temperature	397 °C
Primary Auxiliary Sodium System	
Overflow and Makeup System	
Flow Rate	2.0×10^4 kg/h
Purification System	
Number of System	1
Flow Rate	1.0×10^4 kg/h
Cold Trap Inlet Temperature	~200 °C
Cold Trap Outlet Temperature	~150 °C
Cold Trap Cooling Medium	N_2 gas
Argon Cover Gas System	
Flow Rate	8 Nm^3/h

Reactor coolant flows upward through the core and is heated up from 397 °C to 529 °C in the reactor core. Primary pump is placed in its cold leg and is of centrifugal type. Intermediate heat exchanger is a straight tube cluster type with primary sodium flowing shell side.

The purification system of the primary sodium is composed of overflow tank and cold traps with the flow rate of 10 ton/h. Maximum capability of the system of pumping up from overflow tank to reactor vessel is 20 ton/h. The capacity of the cold traps is so chosen that they can serve for entire plant life.

The argon cover gas circulates through the closed system during reactor normal operation and the radioactively contaminated argon gas is not released to the environment. In the purification system of argon gas, rare gas and volatile elements are removed by activated charcoal beds and cryogenic distillation column. This process is illustrated in Fig. 2. The effectiveness of this process has been demonstrated on a pilot mockup plant,[1] which showed the rare gas removal efficiency of 10^{-7}, while the required efficiency of MONJU design is about 10^{-4}.

3. Release of radioactive nuclides into sodium

3.1 Fission product and tritium

The plant design is based on the assumption that fission product is released from failed fuel of 1%, where the effect of fission product retention in fuel pellets and gas plenum is now under examination. Assumptions taken in calculating the release of fission product and tritium into sodium are:

(i) immediate release of produced rare gas

(ii) release rate of 10% for halogens which react easily with other elements

(iii) release rate of 1% for the other elements which are less mobile, and

(iv) 100% release of tritium through stainless steel clad.

The kind of nuclides investigated and calculational model of their production are similar to Radio Isotope Buildup and Decay (RIBD) code.[2][3] The estimated inventory of principal nuclides is shown in Table 2.

It is assumed that tritium is produced by the following reactions.

(i) Ternary fission

(ii) Boron in control rods

$^{10}B\ (n, 2\alpha)^{3}H$

$^{11}B\ (n, Be)^{3}H$

$^{10}B\ (n, \alpha)^{7}Li\ (n, n\alpha)^{3}H$

(iii) Impurities in sodium

$^{6}Li\ (n, \alpha)^{3}H$

Table 2. Inventory of principal fission product in sodium

nuclide	half life	activity in sodium ($\mu Ci/cm^3$) operation	2 days after reactor shutdown	10 days after reactor shutdown
^{91}Sr	9.7 hours	2.7	8.8×10^{-2}	9.3×10^{-8}
^{95}Zr	65.5 days	5.0	4.9	4.5
^{95}Nb	35.0 days	4.5	4.3	3.7
^{99}Mo	66.7 hours	5.1	3.1	0.42
^{103}Ru	39.5 days	7.4	7.1	6.2
^{106}Ru	368 days	2.9	2.9	2.8
^{131}I	8.1 days	30	25	13
^{135}I	6.7 hours	44	0.3	6.8×10^{-10}
^{133}Xe	5.3 days	8.2	6.3	2.2
^{137}Cs	30.0 years	2.3	2.3	2.3
^{140}Ba	12.8 days	3.1	2.8	1.8
^{140}La	40.2 hours	12	5.4	0.2
^{143}Ce	33.0 hours	10	3.7	6.5×10^{-2}
^{147}Nd	11.1 days	27	24	14
^{149}Pm	53.1 hours	26	14	1.1
^{153}Sm	46.8 hours	28	14	0.8
^{155}Eu	1.8 years	28	28	28

3.2 Corrosion product

Major sources of the corrosion products of activated stainless steel (AISI 316) are fuel clads and assembly wrapper tubes. The composition of assumed stainless steel is shown in Table 3.

Table 3. Composition of stainless steel

element	percentage (%)
Fe	67.4
Ni	10.5
Cr	18.0
Co	0.05
Mo	2.0

The activation of zinc, antimony and tantalum is neglected on the basis that their activated products could be scarcely detected in the experimental sodium loop.[4] The following formations of principal corrosion products are considered as shown in Table 4.

Table 4. Formation of principal corrosion product

corrosion product	half life	activation reaction
^{51}Cr	27.8 days	$^{50}Cr(n,\gamma)$
		$^{52}Cr(n,2n)$
		$^{54}Fe(n,\alpha)$
^{54}Mn	303 days	$^{54}Fe(n,p)$
		$^{55}Mn(n,2n)$
^{59}Fe	45.6 days	$^{58}Fe(n,\gamma)$
^{58}Co	71.3 days	$^{58}Ni(n,p)$
		$^{59}Co(n,2n)$
^{60}Co	5.3 years	$^{59}Co(n,\gamma)$
		$^{60}Ni(n,p)$

It is clear that corrosion product content in primary sodium depends greatly on the corrosion rate. This is estimated as about 1.0 μ/year by using the empirical formula[5] presented by Zebroski, et al. The oxygen concentration in primary sodium is below 10 ppm during plant life.

Furthermore it is assumed that every corrosion product has the identical corrosion rate for the lack of satisfactory information on preferential leaching[4] of particular elements. The content of leached corrosion products is calculated by the following equation[6] considering only radioactive decay for individual nuclides.

$$Q_j = \sum_{i \neq j} \frac{\sigma_i \phi F_i \rho N_0 C A}{3.7\times10^{10} \cdot M \cdot \lambda_j} \left(1 + \frac{e^{-\lambda_j T_c}}{\lambda_i T_c} - \frac{1}{\lambda_j T_c}\right)\left(1 - e^{-\lambda_j t}\right) \qquad (1)$$

Q_j : total activity of j nuclide (Ci)

σ_i : activation cross section of target i nuclide (barn)

ϕ : core neutron flux ($n/cm^2 \cdot sec$)

F_i : fraction of i nuclide in the target material

N_0 : Avogadro's number

C : corrosion rate (cm/sec)

A : area of corrosion product nuclide source (cm^2)

ρ : alloy density (g/cm^3)

M : alloy molecular weight

λ_j : decay constant of j nuclide (sec^{-1})

T_c : length of fuel cycle (sec)

t : reactor operating time (sec)

The activation is calculated by two neutron group model and the cross section is collapsed from BNL-325[7] according to MONJU core spectrum. The leached inventory in sodium is presented in Table 5 on the reactor operation of both two and a half years and thirty years.

Table 5. Inventory of principal corrosion product in sodium

nuclide	half life	activity in sodium 2.5 yrs-operation Ci	activity in sodium 30 yrs-operation Ci	percentage (%)
^{51}Cr	27.8 days	87	87	8.8
^{54}Mn	303 days	290	320	32.4
^{59}Fe	45.6 days	12	12	1.2
^{58}Co	71.3 days	290	290	29.3
^{60}Co	5.3 years	80	280	28.3

4. Radioactivity distribution in primary circuit

4.1 Fission product

The released fission product takes one of the following behavior;

(i) stay within sodium

(ii) deposit on the cold trap and removed from primary sodium

(iii) deposit on the piping and components and stays there, and

(iv) transfer further to argon cover gas.

After evaluating research and development results,[8][9] the fractional distribution of the individual element is given as presented in Table 6.

Table 6. Fractional distribution of fission product in primary system (%)

element	deposition on cold trap	deposition on piping and components	stay within sodium	argon cover gas
rare gas (Kr, Xe)	0	0	1	99
halogen (I, Br)	33	33	33	1
Alkali metal (Rb, Cs)	90	0	10	0
balance	20	80	0	0

Concerning removal of gaseous and volatile fission product from argon gas, it is assumed on the basis of mockup experiment that rare gas is separated on the cryogenic processing system by 99.99% and halogen is thoroughly adsorbed on the activated charcoal bed. Then the distribution of the principal fission products is estimated as in Table 7.

Table 7. Distribution of principal fission products in primary system - 2.5 yrs operation

nuclide	deposition* on cold trap μCi	deposition* on piping and component $\mu Ci/cm^2$	argon cover gas $\mu Ci/cm^3$	rare gas storage cylinder $\mu Ci/cm^3$
^{85}Kr	—	—	3.8×10^{-4}	4.3×10^{5}
^{88}Kr	—	—	1.3×10^{3}	—
^{95}Nb	5.3×10^{7}	0.88	—	—
^{95}Zr	6.4×10^{7}	1.1	—	—
^{103}Ru	7.7×10^{7}	1.3	—	—
^{131}I	3.3×10^{9}	14	8.0×10^{-2}	—
^{133}Xe	—	—	3.3×10^{2}	1.6×10^{5}
^{137}Cs	2.1×10^{8}	—	—	—
^{140}Ba	3.4×10^{7}	0.57	—	—
^{140}La	2.1×10^{6}	0.035	—	—
Total	3.7×10^{9}	18	1.6×10^{3}	5.9×10^{5}

* on 10 days after reactor shutdown

It is expected that the cold trap can remove most of the tritium from primary sodium as the compound of Na^3H.

Concerning the deposition of fission product, iodine - 131 is shown to hold the prominent radioactivity during reactor operation and just after reactor shutdown, and after several tens of days, its intensity decreases to the same level as the other nuclides.

4.2 Corrosion product

The corrosion product leached into sodium is removed from the primary sodium as being deposited on piping, components and cold trap. The deposition rate is obtained as system depletion factor from the small sodium loop.[10] The remaining content in sodium of corrosion product can be predicted by the equation (2) with depletion term of radioactive decay and deposition which can be expressed by using the equation (1).

$$P_j = \frac{Q_j}{1-e^{-\lambda_j \cdot t}} \left[1-e^{-(\lambda_j+\beta_j)t} \right] \qquad (2)$$

where

P_j : remaining activity in sodium of j nuclide (Ci)

β_j : depositing removal rate of j nuclide (Sec^{-1})

Then the deposited activity ΔQ_j on piping and components is given by:

$$\Delta Q_j = Q_j - P_j \quad (Ci) \qquad (3)$$

Moreover it is assumed that the deposition term is divided into cold trap and elsewhere by 20% and 80% respectively. The estimated distribution of corrosion product is presented in Table 8.

Table 8. Distribution of principal corrosion products in primary system - 2.5 yrs operation

nuclide	half life	deposition * on cold trap μCi	deposition* on piping and component $\mu Ci/cm^2$	stay* within sodium $\mu Ci/cm^3$
^{51}Cr	27.8 days	8.2×10^6	0.14	2.4×10^{-2}
^{54}Mn	303 days	5.2×10^7	0.86	1.6×10^{-2}
^{59}Fe	45.6 days	1.5×10^6	0.025	2.8×10^{-3}
^{58}Co	71.3 days	4.2×10^7	0.7	4.9×10^{-2}
^{60}Co	5.3 years	1.5×10^7	0.26	2.5×10^{-3}
Total		1.2×10^8	2.0	9.4×10^{-2}

* on 10 days after reactor shutdown

It is shown that these corrosion products constitute the main source term during maintenance and repair work on primary piping and components due to their considerably long half lives.

5. Estimation of radiation level on primary circuit

It is useful in planning maintenance work to estimate the radiation level around the primary components, which can be easily obtained from the radioactivity distribution. The radiation levels over the sodium drained primary circuit are shown in Table 9.

Table 9. Radiation level on primary piping and IHX

location	radiation level* (mr/h)	
	2.5 yrs-operation	30 yrs-operation
hot leg piping		
surface	1,100	1,500
1 meter from surface	310	440
IHX		
surface	2,200	3,300
1 meter from surface	830	1,200
tube bundles	4,300	5,400

* on drained sodium system and 10 days after reactor shutdown.

6. Further research and development program

6.1 Transfer of fission products from sodium to argon gas

The results obtained in loop experiments are reflected to the plant design in the case of fission gas transfer from sodium to argon gas.

(i) application of transfer coefficient

The isotopic concentration of a volatile element in sodium is given by

$$\frac{dx}{dt} = \frac{A}{N_S} - \lambda x - k \cdot S \frac{N_0}{N_S} \cdot x \qquad (4)$$

where

x : molecular fraction of volatile element in sodium

A : release rate of volatile element to sodium (atoms/sec)

N_S : number of sodium atoms

k : transfer coefficient (mole/cm^2.sec.mole-fract.)

S : sodium surface (cm^2)

N_0 : Avogadro's number

λ : decay constant (sec^{-1})

On the other hand the number of the volatile element in argon gas is expressed by;

$$\frac{dQ}{dt} = kSN_0 \cdot x - \lambda Q \qquad (5)$$

where

Q : number of volatile element in argon gas

If the transfer rate of volatile element from sodium to argon gas is defined as the ratio of its number in argon gas to that in sodium, this rate is expressed in equilibrium condition by;

$$\frac{Q}{xN_S} = \frac{kSN_0}{\lambda N_S} \qquad (6)$$

(ii) application of Henry's law

The Henry's law holds in equilibrium between dilute solution with volatile element and its cover gas, as expressed below.

$$K_H = \frac{x}{P^*} \qquad (7)$$

where

P^* : partial pressure of volatile element in argon gas

K_H : Henry's constant

In this case, the above defined transfer rate is given by;

$$\frac{Q}{xN_S} = \frac{N_0 \cdot V}{N_S \cdot R \cdot T K_H} \qquad (8)$$

where

V : volume of argon gas

R : gas constant

T : temperature of argon gas

It is expected that the reasonable means can be established by using equations (6) and/or (8) to predict the transfer of rare gas, halogen and alkali metal.

6.2 Improvement and extensive use of the code of computing the distribution of fission and corrosion product in LMFBR-PLANET[11]

PLANET code assumes the equivalent model to that of STP-I[12] for production and transfer of radioactive nuclides in primary circuit. Concerning the release of fission product from defective fuel to sodium, its retention in fuel pellets is expressed as the migration of diffusion, recoil, knockout and recrystallization. The principal objects of this code are;

(i) synthetic assessment of radioactivity distribution over the reactor plant

(ii) effective aid for maintenance work programming through the estimation of radioactivity distribution according to reactor operational mode, and

(iii) explicit determination of physico - chemical constant involved in corrosion and deposition model through the analysis of different loop experiments.

It is expected that to predict the behavior of individual nuclide more accurately, calculational model and many constants involved will be established in the near future through the analysis of the operational data obtained from large sodium loops and JOYO.

6.3 Experiments on radioactive products in sodium loop

Possibility of doing some experiments are being planned to make the quantitative investigations on fission and corrosion product behavior in sodium loop and to utilize their outcome for the design calculation. They include;

(i) Sodium Inpile Loop (SIL) with UO_2 fuel which has been installed in JAERI

(ii) Fission Product Test Loop (out-of-pile type) in PNC-Oharai

(iii) Hot Steel Test Loop - II (out-of-pile type) in PNC-Oharai

6.4 Study on tritium behavior

To obtain the elemental behavior of tritium, research and development will be directed toward its removal from sodium and understanding of its transfer features in sodium.

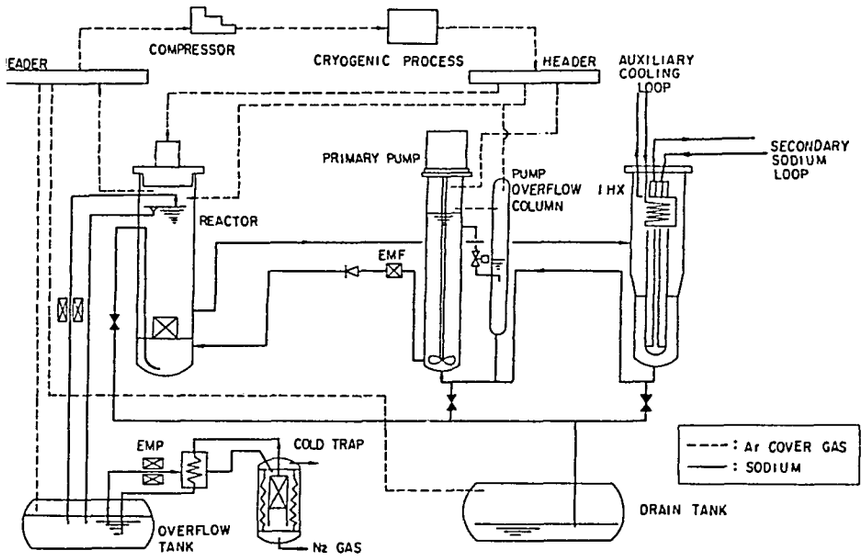

Fig. 1 Schematic Diagram of Primary Circuits of MONJU

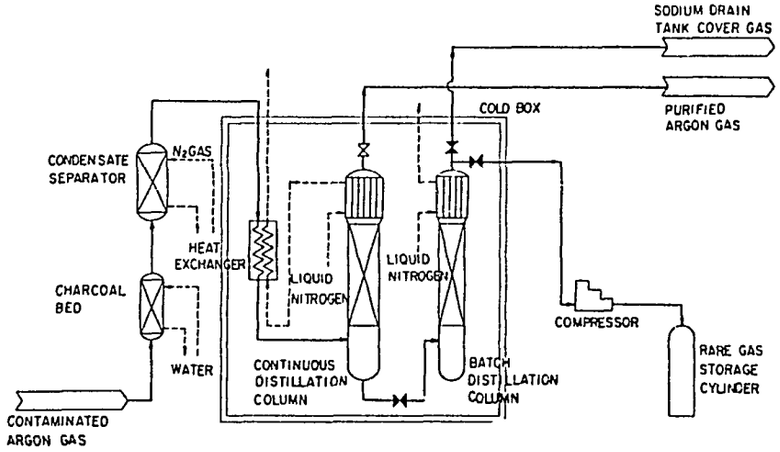

Fig. 2 Cryogenic Argon Processing System

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12. Fission and Corrosion Product Behaviour in Primary Circuits of LMFBR's. A Status Review of Work Being Undertaken in the UK

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1. INTRODUCTION

Safe and economic operation of Fast-Reactor systems require an understanding of how long lived isotopes (eg ^{54}Mn, ^{60}Co and ^{137}Cs)* and bulk corrosion products are released, transported and deposited in sodium circuits. For example build-up of radioactive species in those areas of the reactor which require periodic inspection or maintenance, such as pumps and heat exchangers, could make these operations both costly and difficult. Likewise build-up or the 'smearing' of corrosion products over pipework or heat-exchanger surfaces could effect the thermal-hydraulic performance of the system by the formation of deposits and roughening of pipework surfaces.

The UK programme to study fission and corrosion product behaviour in fast reactor systems is aimed at establishing:

a) the magnitude of material and radioactivity release to the primary coolant and the respective contribution from fuel cladding and structural components.

b) the pattern of corrosion product and activity deposition in various parts of the circuit.

c) the accuracy of estimates of activity levels deposited on the surfaces of circuit components liable to require maintenance or repair, and associated dose rates.

d) the effect of corrosion product deposition on circuit hydraulics and heat transfer.

2. CORROSION & MASS TRANSPORT BEHAVIOUR OF NON-ACTIVE AND ACTIVE CORROSION PRODUCTS

Non-Active Studies The aim of the REML experimental programme is to study mass transport of materials in sodium using both small and large loop facilities. The corrosion studies are primarily concerned with trying to understand the mechanism of stainless steel corrosion in sodium and special attention is being paid to the role individual alloying elements play in the corrosion process.

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* ^{54}Fe (np), ^{55}Mn (n 2n) } ^{54}Mn — 300 day half-life

^{59}Co (n γ), ^{60}Ni (np) } ^{60}Co — 5.27 yr half-life

Fission product ^{137}Cs — 30 yr half-life