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Release of ^{129}I to the coolant

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SUMMARY

Various methods are used to estimate the release rate or integrated release of ^{129}I at various times during operation of a power reactor. The calculations are based on experience from General Electric and Westinghouse and on operation of Swedish power reactors. With conservative assumptions the load of ^{129}I on the ion exchangers is 1.1 MBq/year for a standard BWR and 11 MBq/year for a standard PWR, assuming continuous operation at 2500 MW(th).

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1. INTRODUCTION

For evaluation of the safety by long-time storage of ion-exchanger-waste from power reactors it is of interest to know the load of the long-lived activity ^{129}I, $t_{1/2}$ = 16 million year. Various methods are used below to estimate the integrated release and release rate of ^{129}I at various times during the operation of a power reactor operated at 2500 MW(th).

The present report is a revised edition of the report E2-79/41, dated 1979-04-26.

2. MECHANISMS FOR THE RELEASE OF FISSION PRODUCTS FROM THE FUEL

2.1 Recoil and knock-out

Even at low temperatures there will be a release of fission products which by virtue of their high kinetic energy at formation can recoil from the fuel. One would expect a quarter of those formed within a recoil range of the surface to escape from the fuel, a small fraction of these coming to rest in the can interspace and the remainder entering the canning material. The release should be proportional to yield and fission rate and the fraction released should be independent of half-life and irradiation time. In addition previously formed fission products may be knocked out by collision with hot fission fragments. It has been suggested that at temperatures up to 1000°C the release was by recoil and knock-out or evaporation of fuel and its associated fission products.

2.2 Diffusion

Early work by Booth and Rymer showed that for fuel up to about 1600°C fission gas and iodine release

could be explained on a diffusion model. The uranium dioxide behaved as though it were composed of "hypothetical spheres" of radius a, the fission product concentration being zero at the surface of the spheres. The fraction released at time t then depends on Dt/a^2 and $\lambda a^2/D$, where λ is the decay constant and D is the diffusion coefficient of the fission product which is characterized by an activation energy E.

The application of the diffusion theory has been treated in detail by Beck [1]. His short-time approximations are as follows:

A. Active fission products

$$H = 3 \cdot \sqrt{\frac{D'}{\lambda}} - 3 \frac{D'}{\lambda} \qquad \text{(eq 1)}$$

B. Stable fission products

$$H_{st} = 6\sqrt{\frac{D't}{\pi}} - 3D't \qquad \text{(eq 2)}$$

$$F_{st} = 4\sqrt{\frac{D't}{\pi}} - 1.5D't \qquad \text{(eq 3)}$$

Explanations:

$H = \frac{R}{B}$ (eq 4)

R = release rate (atoms/s)

B = birth rate (= rate of formation) (atoms/s)

$F = \frac{N}{N_F}$ (eq 5)

st = designates a stable isotope

N = accumulation of undecayed released atoms

N_F = number of undecayed atoms in fuel
D' = $\frac{D}{a^2}$ (s^{-1})
a = radius of the sphere (cm)
D = diffusion coefficient ($cm^2 \cdot s^{-1}$)
λ = decay constant (s^{-1})
t = time (s)

Further simplifications of Becks equations, valid for small release fractions (≤ 0.05), give

$$H = 3\sqrt{\frac{D'}{\lambda}} \qquad \text{(eq 6)}$$

$$H_{st} = 6\sqrt{\frac{D't}{\pi}} \qquad \text{(eq 7)}$$

$$F_{st} = 4\sqrt{\frac{D't}{\pi}} \qquad \text{(eq 8)}$$

2.3 High temperature release

At temperatures above 1600°C, gross structural changes occur in the uranium dioxide. In a temperature range 1600-1800°C there is equiaxed grain growth and from 1800°C to the melting point of 2800°C large columnar grains are formed, sometimes with central voids. These changes result in the release of fission products.

2.4 Applications of the theory

Skarpelos and Gilbert [2] have studied the release of fission products to the coolant in General Electric power reactors. Based on the operational experience they have formulated a design basis for the release. For iodines they use equation 9

$$R = 2.4 \cdot 10^7 \cdot y \cdot \lambda^{0.5}(1-e^{-\lambda \cdot t}) \qquad \text{(eq 9)}$$

where R is the release rate in μCi/s, y is the fission yield, λ is the decay constant in s^{-1} and t is the irradiation time in s. With this equation the release rates shown in Table 1 were calculated.

Equation 9 is based on the assumption that 100 out of 300 000 fuel rods are defect, i e 3.3 o/oo. Iodines seem to be released according to the theory of diffusion (R is proportional to $\sqrt{\lambda}$, the birth rate B is equal to the decay rate $N_F \cdot \lambda$ at equilibrium, i e $H = \frac{R}{B} \propto \frac{\sqrt{\lambda}}{\lambda} = \sqrt{\lambda^{-1}}$).

Westinghouse [3] expresses the release of iodine in their power reactors with the escape rate coefficient ν, which is defined as follows

$$\nu = \frac{R}{N_F} \qquad \text{(eq 10)}$$

At equilibrium $B = N_F \cdot \lambda$ and accordingly

$$\nu = \frac{R}{N_F} = \frac{R}{B/\lambda} = H \cdot \lambda \qquad \text{(eq 11)}$$

Westinghouse assumes an escape rate cofficient for iodines of $1.3 \cdot 10^{-8}$ s^{-1} and a defect level of the fuel of 1%.

3. CALCULATIONS

All calculations are based on a 2500 MW(th) reactor. Calculations have been carried out based on various assumptions such as the design basis of General Electric and Westinghouse. ^{129}I is produced with a yield of 0.0066 [4] and has a long half-live, $t_{1/2} = 15.9 \cdot 10^6$ year, $\lambda = 1.38 \cdot 10^{-15}$ [4].

3.1 Calculations based on D'

It is therefore more realistic to regard ^{129}I as a stable fission product and use the adequate equations 7 and 8.

The release of iodines is characterized by an empirical diffusion coefficient D'. D' consists of a

composition of values (including release from the interface to the coolant) but the same composition is assumed to be valid for both ^{129}I and ^{131}I. For an radioactive isotope i the release fraction H_i is

$$H_i = 3\sqrt{\frac{D'}{\lambda_i}} \qquad \text{(eq 6)}$$

i e $$D' = H_i^2 \cdot \frac{\lambda_i}{3^2} \qquad \text{(eq 12)}$$

For a stable isotope the release fraction F_{st} then is

$$F_{st} = 4\sqrt{\frac{D't}{\pi}} = \frac{4 \cdot H_i}{3}\sqrt{\frac{\lambda_i t}{\pi}} \qquad \text{(eq 13)}$$

3.1.1 Oskarshamn 1

In ref [5, 6] release fractions for iodines were calculated for 3 periods preceeding the refuelling stops 1975, 1976 and 1977. The value for ^{131}I were $(3.6, 7.5 \text{ and } 4.7)\cdot 10^{-4}$ with defect levels of 1.8, 2.1 and 1 o/oo, respectively. No corrections were included for release of iodines from uranium in the coolant.

Based on these values I assume the following conservative values:

Fraction of defect rods:	$2\cdot 10^{-3}$
Release fraction, H, for ^{131}I:	10^{-3}

According to equation 13:

$$F_{st} = \frac{4}{3}\cdot H(^{131}I) \cdot \sqrt{\frac{\lambda(^{131}I)\cdot t}{\pi}} =$$

$$= \frac{4}{3}\cdot 10^{-3} \cdot \sqrt{\frac{9.95\cdot 10^{-7}}{\pi}} \cdot t =$$

$$= 1.33\cdot 10^{-3}\cdot 5.63\cdot 10^{-4}\cdot \sqrt{t} =$$

$$= 7.5\cdot 10^{-7}\sqrt{t}$$

In Table 2 the results are shown after 2, 5 and 10 years of operation. The defect level 2 o/oo is used to calculate the inventory in the defect rods. The total inventory of ^{129}I in the fuel may be calculated for a 2500 MW(th) reactor as follows assuming $3.1 \cdot 10^{16}$ fissions per MWs:

$$N = 2500 \cdot 3.1 \cdot 10^{16} \cdot y \cdot t \text{ (atoms)} \qquad \text{(eq 14)}$$

and the activity, A, is

$$A(^{129}I) = \frac{N(^{129}I) \cdot \lambda(^{129}I)}{3.7 \cdot 10^{10}} \text{ Ci} \qquad \text{(eq 15)}$$

The contribution from uranium in the coolant is small. Assuming 10 g of uranium in the coolant the production rate of ^{129}I is $9 \cdot 10^{-10}$ µCi/s ($P = \sigma_f \cdot \phi_{th} \cdot y \cdot n(U_{235})$, σ_f = 580 barns, $\phi_{th} = 3.5 \cdot 10^{13}$ $n \cdot cm^{-2} \cdot s^{-1}$).

3.1.2 General Electric

Calculations have been made in a similar way based on the design basis of General Electric and Westinghouse, data from Ringhals 1 and 2 and design basis for a standard PWR and BWR, see Table 3.

For General Electric reactors $R(^{131}I)$ = 700 µCi/s according to Table 1. The birth rate, B, in defect rods are calculated as $B = 3.1 \cdot 10^{16} \cdot 2500 \cdot 0.0033 \cdot 0.0291 \cdot \frac{9.95 \cdot 10^{-7}}{3.7 \cdot 10^{4}} = 2.00 \cdot 10^{5}$ µCi/s.

Consequently $H(^{131}I) = \frac{R}{B} = \frac{700}{2 \cdot 10^{5}} = 3.5 \cdot 10^{-3}$.

The fraction of defect rods, f_v, is 0.0033. We may calculate the integrated release A_I by comparing these data with the data from Oskarshamn.

$$A_I(GE) = A_I(O1) \cdot \frac{H(GE)}{H(O1)} \cdot \frac{f_v(GE)}{f_v(O1)} =$$

$$= A_I(O1) \cdot \frac{3.5 \cdot 10^{-3}}{1 \cdot 10^{-3}} \cdot \frac{0.0033}{0.002} =$$

$$= A_I(O1) \cdot 5.8$$

3.1.3 Westinghouse

The release rate coefficient $\nu = 1.3 \cdot 10^{-8} s^{-1}$. For ^{131}I $H = \frac{\nu}{\lambda} = \frac{1.3 \cdot 10^{-8}}{9.95 \cdot 10^{-7}} = 1.3 \cdot 10^{-2}$. The fraction of defect rods f_v is assumed to be 0.01.

$$A_I(W) = A_I(O1) \cdot \frac{1.3 \cdot 10^{-2}}{1 \cdot 10^{-3}} \cdot \frac{0.01}{0.002} =$$

$$= A_I(O1) \cdot 65$$

3.1.4 KWU

KWU has given a relation between $\nu(^{131}I)$ and heat rating [7]; at low heat ratings ν is $2 \cdot 10^{-8}$, near to the value of Westinghouse. The release rate of ^{129}I should then be about the same as calculated under 3.1.3.

3.1.5 Ringhals 1

Based on information from Ringhals [8] the release rate R in July 1978 was calculated at 2.3 µCi/s and the "birth" rate B at $8.1 \cdot 10^3$ µCi/s in 6 defect pins. The release fraction $H = \frac{R}{B}$ is $2.8 \cdot 10^{-4}$. The fraction of defect rods is 0.00014.

$$A_I(R1) = A_I(O1) \cdot \frac{2.8 \cdot 10^{-4}}{1 \cdot 10^{-3}} \cdot \frac{0.00014}{0.002} =$$

$$= A_I(O1) \cdot 2 \cdot 10^{-2}$$

3.1.6 Ringhals 2

The release rate in March 1978 is 136 µCi/s [8]. The birth rate in $8(^{+8}_{-4})$ defect pins (assumed in 4 detected defect elements) is $(1.6^{+1.6}_{-0.8}) \cdot 10^4$ µCi/s and H

is $(8.8^{+8.8}_{-4.4})\cdot10^{-3}$. The fraction of defect rods is $0.32^{-0.16}_{+0.32}$ o/oo.

$$A_I(R2) = A_I(O1)\cdot\frac{8.8\cdot10^{-3}}{1\cdot10^{-3}}\cdot\frac{0.00032}{0.002} =$$

$$= A_I(O1)\cdot1.4$$

3.1.7 Standard BWR

The release fraction is assumed to be $1\cdot10^{-3}$ and the fraction of defect rods is assumed to be $1\cdot10^{-3}$.

$$A_I(BWR) = A_I(O1)\cdot\frac{1\cdot10^{-3}}{1\cdot10^{-3}}\cdot\frac{1\cdot10^{-3}}{2\cdot10^{-3}} =$$

$$= A_I(O1)\cdot0.5$$

3.1.8 Standard PWR

The release fraction is assumed to be $1\cdot10^{-2}$ and the fraction of defect rods is assumed to be $1\cdot10^{-3}$.

$$A_I(PWR) = A_I(O1)\cdot\frac{10^{-2}}{10^{-3}}\cdot\frac{10^{-3}}{2\cdot10^{-3}} =$$

$$= A_I(O1)\cdot5$$

4. CALCULATIONS BASED ON GE's DESIGN BASIS

If we assume that equation 9, expressing GE's design basis, is valid even for ^{129}I the results shown in Table 4 are obtained.

$$R = 2.4\cdot10^{7}\cdot y\cdot\lambda^{0.5}(1-e^{-\lambda\cdot t}) \qquad \text{(eq 9)}$$

However, equation 9 is used far outside the region, where it has been found to be valid and the extrapolation made is therefore doubtful. The results are three orders of magnitude lower than those calculated above.

5. CALCULATIONS BASED ON WESTINGHOUSE DESIGN BASIS

Westinghouse [3] assume an escape rate coefficient ν for iodines of $1.3 \cdot 10^{-8} s^{-1}$ and a fraction of defect rods equal to 0.01. The release rate R is equal to $\nu \cdot N$. The results are given in Table 5. Even this calculation is doubtful because the model is used far outside the interval, where it has been tested.

6. THE LOAD OF ^{129}I ON THE ION EXCHANGERS

From the values of integrated release given in Table 3 it is possible with some assumptions to calculate the load of ^{129}I on the ion exchangers.

We conservatively assume that the fuel pin is defected after four years of operation after a refuelling stop and that ^{129}I is released until the next refuelling stop after a total of five years operation. During this time iodine diffuses to the fuel-cladding interface. The results reported in Table 5 under five years of operation may then be regarded as the yearly load on the ion exchangers. The fraction of defect rods is accordingly regarded as an indication of the number of new defects appearing between two refuelling stops and all defect rods are assumed to be removed during the refuelling stop.

7. CONCLUSIONS

The integrated release of ^{129}I based on GE's relationship (eq 9, p 5) and on Westinghouse results assuming $\nu = 1.3 \cdot 10^{-8}$ has been calculated but the results are probably unreliable. More reliable results are obtained assuming that ^{129}I is a stable isotope and using a method based on an empirical

diffusion coefficient. A summary of results based on this method is shown in Table 3. With conservative assumptions the values for the integrated release after 5 years of operations may be taken as the yearly load on the ion exchangers.

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/Eli

Table 1

GE's design basis for a BWR/5 standard plant

Iodine isotope	λ, s^{-1}	Fission yield	Leakage rate µCi/s	MBq/s
131	$9.95 \cdot 10^{-7}$	$2.91 \cdot 10^{-2}$	$7 \cdot 10^{2}$	26
132	$8.43 \cdot 10^{-5}$	$4.26 \cdot 10^{-2}$	$9.4 \cdot 10^{3}$	350
133	$9.26 \cdot 10^{-6}$	$6.69 \cdot 10^{-2}$	$4.9 \cdot 10^{3}$	180
134	$2.21 \cdot 10^{-4}$	$7.8 \cdot 10^{-2}$	$2.8 \cdot 10^{4}$	1000
135	$2.87 \cdot 10^{-5}$	$6.17 \cdot 10^{-2}$	$7.9 \cdot 10^{3}$	290

Table 2

Integrated release and release rates of ^{129}I to the coolant of Oskarshamn 1*

	Years of reactor operation		
	2	5	10
Time, 10^7 s	6.3	15.8	31.5
$F_{st} \cdot 10^3$	6.0	9.5	13.3
Total inventory, Ci*	1.2	3	6
Inventory in defect pins, mCi	2.4	6	12
Integrated release, µCi	14	57	160
Release rate, pCi/s**	0.22	0.46	0.65
Integrated release, MBq	0.52	2.1	5.9

* Extrapolated to 2500 MW(th)

** Mean value in the preceeding period

Table 3

Integrated release of ^{129}I to the coolant of a 2500 MW(th) reactor at continuous operation. Summary of results.

	Multi-plication factor	Integrated release after various periods of reactor operation					
		μCi			MBq		
		Years of operation			Years of operation		
		2	5	10	2	5	10
Oskarshamn 1	1	14	57	160	0.52	2.1	5.9
GE	5.8	80	340	960	3.0	12.6	36
Westinghouse	65	900	3700	10400	33	137	380
Ringhals 1	$2 \cdot 10^{-2}$	0.3	1.1	3.0	0.01	0.04	0.12
Ringhals 2	1.4	20	81	220	0.74	3.0	8.1
Standard BWR	0.5	7	29	80	0.26	1.1	3.0
Standard PWR	5	70	290	800	2.6	11	30

Table 4

Release rate and integrated release of ^{129}I based on GE-data using the equation $R = 2.4 \cdot 10^{7} \cdot \lambda^{0.5} \cdot y\,(1-e^{-\lambda \cdot t})$.

	Years of operation		
	2	5	10
$R \cdot 10^{9}$, µCi/s	0.51	1.3	2.6
R, µBq	19	50	100
Integrated release, µCi*	0.03	0.15	0.56
Integrated release, kBq	1.1	6	20

* Based on the highest release rate during the preceeding period(s).

Table 5

Inventory and release rate of ^{129}I based on Westinghouse data using the equation $\nu = R/N$

	Years of operation		
	2	5	10
Total inventory, Ci	1.2	3	6
Inventory in defect pins, Ci*	0.012	0.03	0.06
$R \cdot 10^{4}$, µCi/s**	1.6	4	8
$R \cdot 10^{9}$, µCi/s***	5.8	15	29
Integrated release, µCi***	0.37	1.8	6.4
Integrated release, kBq***	14	67	240

* Assuming conservatively a 1% defect level

** Assuming $\nu = 1.3 \cdot 10^{-8}$

*** Assuming $\nu = 1.3 \cdot 10^{-8} \cdot \sqrt{\frac{\lambda(^{129}I)}{\lambda(^{131}I)}} = 4.8 \cdot 10^{-13}$

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