RADIOACTIVE CORROSION PRODUCT RELEASE
POTENTIAL IN SODIUM-COOLED REACTORS

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ABSTRACT

A mathematical analysis of the rate of release and deposition of radioactive corrosion products from the core of liquid sodium cooled reactors is outlined and numerical results presented as a function of reactor design parameters for a number of reactors. It is shown that the activity release potential of advanced sodium-cooled reactors (FFTF and CRBRP) is about two orders of magnitude larger than that of older reactors such as EBR-II, and RAPSODIE. Radiation levels up to 6 R/hr are predicted for FFTF and CRBRP primary system components after 20 years of operation.

The model can be used to predict the sensitivity of activated corrosion product release to changes in operating parameters for various existing and planned reactors.

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G. R. Taylor and R. P. Colburn

INTRODUCTION

An important factor in the economics of fast breeder reactors is the assumed plant availability. To obtain the desired 80 percent or better availability, highly reliable design and construction is of first priority. In addition, ease of maintenance and repair of the system is mandatory. One factor which must be considered in planning for maintenance is the level of radioactivity which will exist at the various plant components during plant life.

The generation of radioactivity is inherent in the production of fission nuclear power. The fission products are in general radioactive, and the neutron flux transmutes the elements of the reactor structural materials and coolant to nuclides which also are generally radioactive. The obvious radiological hazards to the public are dealt with by containment in depth; i.e., the fuel is clad, the coolant is contained in high integrity systems, and the whole is kept within a containment vessel. These containments must however be opened for refueling, fuel reprocessing, and for system maintenance and repair, necessitating the development of suitable methods for handling radioactive materials. Because of the unavoidably high radiation level of the fuel proper and its immediate support structure, refueling and reactor structures maintenance must be done remotely. On the other hand, for systems external to the reactor vessel, this is not necessarily the case. The radioactivity in the primary system external to the reactor vessel will be only that which have been transported there; it will be contained in the coolant, or in a superficial but not necessarily loosely adherent layer on the system's surfaces.

This paper is concerned only with that portion of the extra core radio-activity originating from the corrosion and transport of activity from the core cladding and other structural members. Any contribution of fission products from breached fuel pins is not considered.

ANALYSIS OF ACTIVITY RELEASE AND RELEASED INVENTORY

The rate of release of a given active nuclide from the reactor to the coolant is given by:

$$(1) \quad \dot{N} = \int N \dot{V} dv + 1/4 \int \sigma_p N^p \phi_s dv$$

where:

$$(2) \quad \dot{V} = s a e^{bT}$$

$$(3) \quad N = N^o e^{-\lambda t} + \phi \sigma_p N^p (1 - e^{-\lambda t}) / \lambda$$

The first term on the right-hand side of Equation 1 gives the corrosion contribution to activity release while the second term gives the instantaneous contribution due to recoil of active products resulting from (n, p) reactions.

The reactor is assumed to be a right circular cylinder with neutron flux given by¹

$$(4) \quad \phi = \phi_m J_0(B_R r) \cos B_z z$$

The corrosion of 316 stainless steel in cm/sec is assumed to be of the form:

$$R = a e^{bT}$$

¹See Notation for definition of symbols.

and the temperature as a function of position is given by:

$$(5) \quad T = T_i + J_o(B_R r) \Delta T_m (1 + \sin B_H z / \cos B_H \delta) / 2 K_r$$

The factor K_r allows for the possibility of non-uniform fuel enrichment leading to more uniform reactor coolant outlet temperatures, while the value of N° varies with the specific refueling pattern chosen (e.g., for total fuel replacement at each refueling, $N^\circ = 0$).

The inventory of released activity is calculated by integration of

$$(6) \quad \dot{P} = \dot{N} - \lambda P$$

The results of these analyses are summarized in Table 1.

Numerical Results

The major contributors to LMFBR plant radioactivity from 316 stainless steel clad fuel and 304 stainless steel structures result from the activation reactions

The pertinent data for these nuclides are summarized in Table 2.

The values of the integrals I_1 and I_2 as a function of $b\Delta T_m$ are shown graphically in Figures 1 and 2 for the uniform enrichment core and the two-zoned radial fuel enrichment core respectively, with the ratio of the actual to the extrapolated core radius as a parameter.

The corrosion rate of 316 stainless steel in sodium has been found to change linearly with oxygen concentration in the sodium at concentrations

above about 1 ppm but become largely independent of oxygen below this concentration. The corrosion rate has also been found to vary approximately linearly with sodium velocity at low flow rates but become independent of velocity above 3 m/sec. In sodium corrosion tests with irradiated cladding in low oxygen sodium (2.5 to 0.5 ppm) the ^{54}Mn release rate has been observed to be 3 times the rate expected for stoichiometric release and its release is limited by diffusion to the surface. Conversely ^{60}Co is preferentially retained on the corroding surface and only released at about 0.5 times the stoichiometric rate. In addition the preferential release of Ni from the cladding surface reduces the surface concentration of the ^{58}Ni precursor for ^{58}Co further reducing the release of this species.

Taking these non temperature factors into consideration the release rate of the activated species may be expressed by the coefficients $a_i = [o] V/3 a_o$ and f_i where $[o]$ = oxygen concentration in ppm and assuming $[o] = 1$ for concentration less than 1 (oxygen independent)

V = sodium velocity (m/sec)
 $= 1$ for sodium velocities > 3 m/sec (velocity independent)

f_i = isotope release factor to account for selective release or retention of the active species and their precursors from the corroding surface

$$f_{\text{Co-60}} = 0.5 \quad f_{\text{Co-58}} = 0.2$$

$$f_{\text{Mn-54}} = 3.0/[o] \text{ for } [o] < 3$$

$$= 1 \text{ for } [o] \geq 3$$

a_o = corrosion rate coefficient for base case in the oxygen and velocity independent region
 $= 4.19 \times 10^{-16}$ cm/sec

b = exponential temperature coefficient
= $0.0144\text{ }^{\circ}\text{C}^{-1}$

The assumed operating parameters for the FFTF and the CRBRP are given in Table 3. From these data the inventory of released activity from the core only was calculated assuming a two-zoned fuel enrichment with radial out-to-in refueling. Values of the radioactivity release integrals are given in Table 4. It is emphasized that these calculations yield only the activity coming from the core; contributions from the breeder blankets and reflectors must be added for the total release. Detailed calculations for FFTF show that the core will contribute 85% of the total ^{58}Co , 83% of the ^{54}Mn , and only 25% of the ^{60}Co . The very significant extra-core ^{60}Co activity results primarily from the increased ^{59}Co adsorption cross-section at the lower energy neutron flux spectrum which exists in the reflector regions. The total release with extra core contribution in FFTF is shown graphically in Figure 3. If one assumes similar fractional contributions from regions surrounding the core occur in other LMFBR's, the total releases may be expressed by dividing the calculated core contribution by Fe = .85, .83 and .25 for ^{58}Co , ^{54}Mn and ^{60}Co respectively as shown in Table 5.

The effect of various design parameters on the plant 20 year activity inventory from the core is shown in Table 5. Changing the inlet temperature by 50°C results in about a two fold change in release. A change from annual to semi-annual refueling decreases ^{58}Co by about 71%, ^{60}Co by 57%, and ^{54}Mn by 42%. Thus only relatively minor savings can be effected by reasonable changes in these design parameters.

An increase in the oxygen activity in the sodium from the base case of 1 ppm to 5 ppm results in a factor of 5 increase in the release of ^{60}Co and ^{58}Co but only a factor of .6 increase in ^{54}Mn thus increasing the relative importance of the cobalt isotopes.

The release of activity is linearly dependent on the concentration of the precursor in the cladding. There is relatively little latitude in reducing the precursors for ^{54}Mn and ^{58}Co if stainless steel cladding is to be used. However specifying low Co concentrations in the cladding, $<0.02\%$, is a feasible method to minimize ^{60}Co release.

Effect of Activity on Plant Maintenance

During plant operations, the intense radiation due to the ^{24}Na formed by neutron activation of the ^{23}Na will dwarf the contribution from the activated corrosion products in the primary sodium system. However, this activity decays with a 15-hour half-life and there also exists the possibility of partially draining the system and/or removal of components during shutdown for maintenance and repair. Under these circumstances, the active corrosion products and fission products from failed fuel will determine plant activity. The magnitude and nature of the released corrosion product activity on plant maintenance depends of course on its ultimate disposition throughout the plant. Corrosion and mass transfer studies indicate that the corrosion products released from the core will deposit on the surfaces in the primary loop and that only a small fraction will appear in the cold trap. The conservative approach therefore is to base shielding design and maintenance plans on the assumption that all of the corrosion products deposit on the primary system surfaces.

To get a feeling for the magnitude of the problem, a uniform distribution of the corrosion products on the primary surfaces was assumed. Using $6.1 \times 10^7 \text{cm}^2$ and $7.7 \times 10^7 \text{cm}^2$ as the primary system surface areas for FFTF and CRBRP respectively, the dose rate at the surface of the intermediate heat exchangers was estimated to be as shown in Table 6. With the indicated dose rates of about 6 R/hr, contact maintenance would clearly be difficult and remote maintenance or prior decontamination might be required under these circumstances.

The deposition patterns observed for these activated species in sodium corrosion test loop showed the ^{60}Co activity tending to deposit immediately downstream from the irradiated source. This suggests a mass transport controlled model probably describes the distribution of ^{60}Co and ^{58}Co more accurately than the uniform distribution model.

^{54}Mn was observed to migrate largely to the cold leg of the system and larger concentrations of this species can be expected on the cold end of the IHX and cold leg components. However, for the sake of comparison for this model, uniform distributions over the primary system components were assumed.

Comparison of Liquid Metal Reactors' Activity Potential

Despite the fact that there have been several liquid metal cooled reactors in operation, there is very little substantial data in the open literature concerning the distribution of activated corrosion products. This can perhaps be explained by the presence of fission products (e.g., in SRE, DFR, BR-5, Fermi), by a failure to look for corrosion product activity, and perhaps due to an actual absence of significant activity. Also, in pool type reactors (e.g., DFR and EBR-II) corrosion product activity would not be noticed as a problem until a component is removed from the tank for maintenance or repair.

Nevertheless, there are some data available, which it would be desirable to examine vis-a-vis activity predictions for the various reactors.

First, it is instructive to compare predicted corrosion product activity sources for the reactors using a uniform basis of comparison. This comparison is given in Table 7 using as a basis the simplifying assumptions of total core refueling and uniform fuel enrichment, i.e., from Table 1,

$$\dot{N} = GI_2 (1 - e^{-\lambda t})$$

and GI_2 is a measure of the reactors relative corrosion product release potential.

The last column in Table 7 gives a measure of the relative corrosion product inventory to be expected in each reactor for the same operating periods and same cladding material.

It can be seen from the last column that FFTF can be expected to have 189 times as much activity as EBR-II, 18 times as much as BOR-60, and 1.22 times as much as PHENIX, under these conditions, while the Demonstration Plant will have 1.32 times the activity release of FFTF.

It is clear from the foregoing, therefore, that the FFTF and Demonstration Plants have a significantly higher relative potential for corrosion product activity than for the earlier liquid metal reactors.

The comparison of the relative activity potential of the various reactors is considered one of the most significant parts of the present effort, and one which is subject to least uncertainty. Application of the approximate model to the various operating sodium reactors requires the specification of source, operation, and fuel cycle history to predict activated corrosion product release during that operation. The factors associated the operation times and actual reactor history are approximated by an assumed regular fuel cycle; the released activity inventory is given by:

$$\lambda P_n = GI_2 f(1 - e^{-\lambda\tau} - \lambda e^{-\lambda\tau})(1 - e^{-n\lambda(\theta + \tau)})/(1 - e^{-\lambda(\theta + \tau)})$$

As shown in Table 7, combinations of low flux, and temperatures, often combined with small stainless steel surface areas relative to those conditions proposed for the FFTF, have resulted in very low corrosion product activity predictions for these small early reactors.

DISCUSSION

There is of course a possible pessimism in the calculations in that no

credit was taken for migration of the activity to the cold trap, and the hot and cold in-vessel surfaces are not given any special weight as activity sinks. However, if irreversible mass transfer controlled deposition is assumed it can be shown that even 100 percent effective cold trapping cannot compete with deposition at any reasonable cold trap flow rate. There may of course be a gradual re-release of deposited material and a continual transfer to the cold trap. The magnitude of this effect cannot be estimated at present.

To investigate general accuracy of the mathematical assumptions of the functional form of the flux and temperature, the FFTF calculations were compared with the results of a detailed point-by-point computer calculation* using "detailed" flux, flux spectrum, temperature, and adsorption cross section data. The analytical solution was high by a factor of 1.30 for ^{58}Co , 1.24 for ^{54}Mn and 2.1 for ^{60}Co . These differences could perhaps be due to our choice of adsorption cross sections. Changing the cross sections by the indicated ratios would of course make the exact and approximate solutions identical.

In conclusion, it would appear from these calculations that corrosion product activity is going to be a problem in LMFBFR's which must be attacked from several angles, i.e., surveillance, development of decontamination techniques, system design, and development of radioactive corrosion product trapping techniques.

*W. L. Kuhn, "Activated Corrosion Product Radiation Levels in the FFTF Heat Transport System Cells and Closed Loop Modules", HEDL-TME-76-10, September 1977.

NOTATION (in order of appearance in the development)

- ϕ = neutron flux, neutrons/cm²-sec.
 ϕ_m = maximum neutron flux in reactor.
 J_n = Bessel function of order n.
 r = radial cylindrical coordinate with origin at core center axis.
 z = axial cylindrical coordinate with origin at core center plane.
 B_z = axial buckling
 = π/H
 H = core height including extrapolation distance, cm.
 $H-2\delta$ = actual core height.
 B_R = radial buckling.
 = $2.405/R_e$
 R_e = core radius including extrapolation distance, cm.
 R = actual core radius
 $\dot{N}$ = rate of release of nuclide N from reactor to coolant, atom/sec.
 N = concentration of nuclide N in cladding or other reactor structure, atoms/cm³, superscript ° refers to zero time.
 $\dot{V}$ = corrosion rate of reactor structure, (cm³/sec) per cm³ space in the reactor.
 dv = differential space element in the reactor, cm³
 = $r \, dr \, d\phi \, dz$
 s = corroding surface area per unit volume reactor space, cm⁻¹
 D_r = recoil range of activated nuclide, cm.
 T = temperature, °C.
 ae_i^{bT} = linear corrosion rate as a function of temperature, cm/sec.
 $a_i = a_o [0] V/3$; $a_o = 4.19 \times 10^{-16}$; $b = .0144^\circ\text{C}^{-1}$
 N^P = concentration of the precursor of nuclide N in the cladding, atom/cm³.
 σ_p = microscopic neutron reaction cross section of precursor nuclide, cm².
 fi = isotope release factor.
 Fe = factor for contribution from region surrounding the core.

T_i = reactor inlet temperature, °C.
 ΔT_m = maximum temperature rise across the core, °C.
 λ = decay constant for nuclide N, sec^{-1} .
 K_r = a factor to take into account radial zones of fuel enrichment, dimensionless.
 S = total corroding surface area in the actual reactor core, cm^2 .
 α = ratio extrapolated core volume to actual core volume
 $= (R_e/R)^2 (1 - 2\delta/H)^{-1}$
 R_i = radius of inner core zone, cm.
 $= R/\sqrt{2}$
 t = general time during an operating cycle, sec.
 τ = reactor operating time per cycle, sec.
 θ = downtime between reactor operations, sec.
 P = plant inventory of released active nuclide N, atoms
 $\dot{P}$ = dP/dt
 P_n = plant inventory of released active nuclide N after nth operating period, atoms.
 v = sodium linear flow velocity, m/sec. = 3 for velocity > 3 m/sec.
 $[O]$ = oxygen concentration in sodium, ppm by weight.
 $= 1$ for concentration < 1.

TABLE 1: SUMMARY OF ACTIVATED CORROSION PRODUCT RELEASE ANALYSIS

$$\dot{N} = FGI_1 f_i (1 - e^{-\lambda\tau}) e^{-\lambda(\theta + \tau)} + GI_2 f_i (1 - e^{-\lambda\tau}) + HI_3$$

$$\lambda P_n = [FGI_1 f_i (1 - e^{-\lambda\tau}) \lambda \tau e^{-\lambda(\theta + \tau)} + GI_2 f_i (1 - e^{-\lambda\tau} - \lambda \tau e^{-\lambda\tau}) + HI_3 (1 - e^{-\lambda\tau})]$$

$$\frac{(1 - e^{-n \lambda(\theta + \tau)})}{(1 - e^{-\lambda(\theta + \tau)})}$$

$$G = 0.22 a e^{bT_i} (\phi_m \sigma_p N^p / \lambda) S \cos(\pi \delta / H) (1 - 2\delta / H)^{-1}$$

$$H = 0.22 (\phi_m \sigma_p N^p D_r / 4) S \cos(\pi \delta / H) (1 - 2\delta / H)^{-1}$$

$$I_1 = (1 / b \Delta T_m) (R_e / R)^2 \int_0^B (K_r J_0(u') / J_0(u)) (e^{b \Delta T_m J_0(u) / K_r} - 1) u du$$

$$I_2 = (1 / b \Delta T_m) (R_e / R)^2 \int_0^{2.4 R / R_e} K_r (e^{b \Delta T_m J_0(u) / K_r} - 1) u du$$

$$I_3 = (2.4)(R_e / R) J_1(2.4 R / R_e)$$

Refueling Scheme:	Total Fuel Change	Replace 1/mth Core Homogeneously	Two-Zone Refueling
F	= 0 (1/m)	$\sum_{i=1}^{m-1} \frac{(1 - e^{-i\lambda(\theta + \tau)})}{(1 - e^{-\lambda(\theta + \tau)})}$	1
Fuel Enrichment Pattern:	Uniform	Two-Zoned Radial	
K_r	= 1	1 for $0 \leq u \leq 1.7 R / R_e$ $J_0(1.7 R / R_e)$ for $u > 1.7 R / R_e$	
u'	= u	$[(1.7 R / R_e)^2 + u^2]^{1/2}$ for $u > 1.7 R / R_e$	
B_R	= 2.4 R / R_e	1.7 R / R_e	

a Refuel by removing center zone, placing outer zone into center zone, and placing fresh fuel into outer zone.

TABLE 2: NUCLEAR DATA FOR SIGNIFICANT RADIONUCLIDES

Nuclide	Element Precursor Concentration in 316 SS, wt %	Precursor Natural Abundance, %	(1) σ_p cm ² (X10 ²⁷)	λ sec ⁻¹	$\frac{\sigma_p N^p}{\lambda}$ atom/cm sec.	Isotope Release Factor For Base Case fi
Cobalt-58	14	67.8	6.13	1.13×10^{-7}	422	.20
Cobalt-60	.02	100.	220.	4.17×10^{-9}	862	.50
Manganese-54	68	5.84	7.7	2.76×10^{-8}	954	3.0

(1) The activation cross sections used (given in Table 2) were those averaged for the flux spectrum at the FFTF core center:

$$\sigma_p = \frac{\sum_J \sigma_{Jp} \phi_J}{\phi_{total}}$$

where the $\sum_J$ is over the 26 group energy structure.

TABLE 3: ASSUMED FFTF AND WESTINGHOUSE DEMONSTRATION PLANT DESIGN PARAMETERS

	T_i	$ae_i^{bT_i}$ cm/sec.	ϕ_m n/cm ² sec.	S cm ²	$\frac{\cos(\pi\delta/H)}{(1 - \frac{2\delta}{H})}$	ΔT_m °C	R_e/R	τ yr.	θ yr.	$b\Delta T_m$
FFTF	427	1.96×10^{-13}	7×10^{15}	3.8×10^6	1.262	278	1.337	0.8	0.2	4.00
CRBRP	399	1.32×10^{-13}	8×10^{15}	6.33×10^6	1.262	267	1.337	0.8	0.2	3.84

TABLE 4
FFTF AND CRBRP RADIOACTIVITY RELEASE INTEGRALS

	G atoms/sec	H atoms/sec.	I ₁	I ₂	I ₃
FFTF	$1.45 \times 10^9 \sigma_p N^p / \lambda$	$1.846 \times 10^{21} \sigma_p N^p D$	5.98	16.16	1.857
CRBRP	$1.85 \times 10^9 \sigma_p N^p / \lambda$	$3.52 \times 10^{21} \sigma_p N^p D$	5.39	14.51	1.857

TABLE 5: DEPENDENCE OF 20 YEAR INVENTORY IN HTS (CURIES) FROM CORE AND SURROUNDING REGIONS ON DESIGN PARAMETERS

	Base Plant ⁽¹⁾		T _i Decrease 50°C	Semi-Annual Refueling	Dissolved Oxygen ⁽²⁾ 5.0 ppm	Recoil ⁽³⁾ Contribution
	Core	Total				
FFTF						
⁵⁸ Co	46.1	54.1	26.4	38.6	270	(43)*
⁶⁰ Co	17.7	70.9	41.1	36.4	354	---
⁵⁴ Mn	658	792	386	328	1267	21
CRBRP						
⁵⁸ Co	54.1	63.7	25.5	45.7	318	(82)*
⁶⁰ Co	20.2	80.7	39.4	41.5	404	---
⁵⁴ Mn	765	922	450	388	1475	40

(1) Total inventory adjusted for contributions from region surrounding the core.

(2) Base case oxygen activity equivalent to 1 ppm [o] in equilibrium with cold trap at 130°C.

Oxygen activity equivalent to 5 ppm [o] in equilibrium with cold trap at 175°C.

(3) Recoil model not valid for ⁵⁸Co due to surface depletion of ⁵⁸Ni precursor.

TABLE 6: RADIATION DOSE RATE AT THE BASE PLANT IHX AFTER 20 YEARS' OPERATION

	$\mu\text{Ci}/\text{cm}^2$	R/hr
FFTF		
^{58}Co	.89	.38
^{60}Co	1.16	1.18
^{54}Mn	12.98	<u>4.67</u> 6.23
CRBRP		
^{58}Co	.83	.36
^{60}Co	1.04	1.07
^{54}Mn	12.0	<u>4.31</u> 5.74

TABLE 7. COMPARISON OF ASSUMED LMFBR OPERATING CONDITIONS AND RESULTING ACTIVITY RELEASE POTENTIAL

REACTOR	Power MWt	ϕ_m (n/cm ² sec) $\times 10^{-15}$	Ti (°C)	ΔT_m (°C)	$a e_i^{b T_i}$ (cm/sec) $\times 10^{13}$	S (cm ²) $\times 10^{-6}$	I ₂	GI ₂ (σ_{Np}/λ) $\times 10^8$
FFTF	400	7.	427	278	1.96	3.8	13	188.
CRBRP	975	8.	399	267	1.31	6.33	11.7	216.
EBR-II	50	3.5	371	188	.83	.215	5.7	1.0
BR-5	5	1.	430	112	1.84	.068	3.8	.13
BR-10	10	1.9	350	240	.50	.10	9.5	.25
BOR-60 (1)	60	3.7	360	224	.58	.45	9.0	3.11
(2)	60	3.7	480	192	3.26	.45	6.8	10.3
BN-350	1000	10.	300	320	.24	7.5	20.0	132.
BN-600	1470	10.	380	272	1.00	11.0	13.5	413.
RAPSODIE	20	2.	450	144	2.67	.15	4.9	1.12
FORTISSIMO	40	3.2	400	182	1.34	.30	6.5	2.31
PHENIX	625	7.2	400	256	1.34	5.0	11.5	153.

Figure 1 Activated Corrected Product Release Integrals, Uniform Fuel Enrichment Case

Figure 2 Activated Corrosion Product Release Interactions: Two-Zone Fuel Burn Case

FIGURE 3: CORROSION PRODUCT ACTIVITY IN FET-5 PLANNED HTS
(CURIES)