NAA-SR-3151
COPY

RADIATION EFFECTS RESULTING FROM FUEL CLADDING FAILURE IN THE 45.5 Mw (THERMAL) ORGANIC MODERATED REACTOR

AEC Research and Development Report

ATOMICS INTERNATIONAL
A DIVISION OF NORTH AMERICAN AVIATION, INC.

NAA-SR-3151
REACTORS-POWER
26 PAGES

RADIATION EFFECTS RESULTING FROM FUEL CLADDING FAILURE IN THE 45.5 Mw (THERMAL) ORGANIC MODERATED REACTOR

BY
M. DUNENFELD

ATOMICS INTERNATIONAL
A DIVISION OF NORTH AMERICAN AVIATION, INC.
P.O. BOX 309 CANOGA PARK, CALIFORNIA

CONTRACT: AT(11-1(-GEN-8
ISSUED: JUN 1 1959

DISTRIBUTION

This report has been distributed according to the category "Reactors - Power" as given in "Standard Distribution Lists for Unclassified Scientific and Technical Reports" TID-450 (14th Ed.), October 1, 1958. A total of 620 copies was printed.

TABLE OF CONTENTS

		Page No.
	Abstract	5
I.	Assumptions and Conditions	7
	A. Reactor Operation	7
	B. Release of Fission Products to the Coolant	7
	C. Fuel Element Type	7
	D. Corrosion Rate	8
	E. Recoil of Fission Fragments	8
	F. Time of Cladding Failure	8
	G. Operation of Purification Still	9
	H. Gaseous Products and Diffusion	10
	I. Adsorption	10
	J. Additional Assumptions	10
II.	Symbols and Assigned Values	11
III.	Derivations and Calculations	13
	A. Long Lived Fission Product Activity	13
	1. Corrosion	13
	2. Recoil.	14
	3. Fission in the Coolant	15
	B. Short Lived Fission Product Activity	19
IV.	Results	21
V.	Conclusions.	25
	References	26

LIST OF TABLES

Page No.

I. Equilibrium Dose Rates from Cladding Failure 21

II. Equilibrium Values of Specific Activities in The Coolant for Isotopes Considered 23

LIST OF FIGURES

1. Equilibrium Dose Rate at Surface of 16-in. Pipe, from Fission Products Released by a 1 sq mm Fuel Cladding Failure 22

ABSTRACT

Effects of fuel cladding failure on the radioactivity in the coolant of the proposed 45.5 Mw Organic Moderated Reactor (OMR) were studied. Figure 1 shows calculated equilibrium values of the dose rate at the surface of an unshielded 16-inch, schedule 40 pipe, for continued full power operation of this reactor. Calculations were based on a cladding failure area of 1 sq mm. Since the half lives of many fission products that emit gamma rays are briefer than the coolant cycle time, the curve shows that the dose rate decreases as coolant travels farther from the core.

Table I shows dose rates, two hours after shutdown, for four different operating conditions of the purification system.

Equations for the buildup of this coolant activity are derived in the text. Also shown is the method of calculating the indicated results.

To validate certain assumptions, experiments should be conducted on: 1) corrosion rate of uranium under reactor operating conditions; 2) diffusion of fission products through uranium and cladding; 3) adsorption of fission products from the coolant by system surfaces.

I. ASSUMPTIONS AND CONDITIONS

A. REACTOR OPERATION

The operating conditions assumed for the reactor were chosen so as to give the maximum coolant activity. These are:

1) Continuous full power operation at 45.5 Mw (thermal) before and after the cladding failure.

2) Cladding failure occurring at center of core, where both flux and power peak to 2.4 times the average values.

3) A 300-day life of the element exposed to the above maximum conditions, although this is the predicted life of an element exposed to average flux conditions.

B. RELEASE OF FISSION PRODUCTS TO THE COOLANT

Following a cladding failure, fission products would be released to the coolant by three processes:

1) Direct recoil of the fission fragments to the coolant.

2) Corrosion of fission products which have built up in the fuel.

3) Fission of uranium which has been corroded from the fuel, and is circulating in the coolant stream.

C. FUEL ELEMENT TYPE

The reactor design considered in this study stipulates the use of an enriched metallic uranium fuel, with aluminum cladding metallically bonded to the uranium. The fuel element consists of a bundle of flat plates of uranium, each sandwiched between aluminum plates. [1]

It is important to note that the presence of a metallic bond between the cladding and fuel is implicit in the calculations performed in this study. With the cladding metallically bonded to the fuel, only the fission products which can be released from the exposed fuel surface area can escape into the coolant. Absence of this bond, or the use of a liquid metal bond, might permit the entire inventory of releasable fission products in a fuel plate to escape into the coolant stream through even a small cladding failure.

D. CORROSION RATE

The corrosion rate of the uranium fuel (and fission products present in the uranium) by the coolant, was taken as 9.65×10^{-11} cm/sec (0.0985 mils/mo), and as invariant with time. The average coolant temperature was assumed to be 500°F. The indicated corrosion rate is the result of Atomics International experiments conducted at 350°F with the organic open to the atmosphere. This condition permitted oxygen and water to dissolve in the organic, and thereby to contact and corrode the uranium.

More recent Atomics International experiments, performed at 700°F in a sealed autoclave, gave a corrosion figure of 6.8×10^{-11} cm/sec (= 0.069 mils/mo) for as-cast natural uranium. In this case, however, the organic was used as received, and had 2 percent or more water content, which might make the corrosion rate high.

These experiments were made with the coolant quiescent, and would therefore not take into account possible erosion of uranium by abrasive action of the flowing coolant. For small area ruptures, which are believed to be the more likely case, the exposed uranium would be protected from full flow by the walls of the cladding fissure or pit. In consequence, it is believed erosion would not be appreciable, at least for pin hole type failures.

E. RECOIL OF FISSION FRAGMENTS

The recoil range of fission fragments[2] in the fuel was taken to be 6.35×10^{-4} cm. One-fourth of the fragments originating in the volume of fuel associated with this thickness, and with the area exposed, were assumed to escape into the coolant.[2] The 1/4 th figure, actually too great for failure areas smaller in diameter than the cladding thickness, was used so that the dose rates calculated per sq mm of failure could be multiplied by a larger area and still be valid.

F. TIME OF CLADDING FAILURE

The maximum contribution to the coolant activity by the corrosion process will occur if the fuel has been irradiated as long as possible before the failure (maximizing the buildup of active products in the fuel), while allowing enough

of the element lifetime after the failure to permit equilibrium of fission and active fission products in the coolant to be approached. Escape of fission products by recoil and the number of atoms of fissionable material corroded into the coolant do not depend upon the length of reactor operation preceding the failure, if burnup is neglected.

Since the equations derived below indicate that elapsed time on the order of 100 days is required after the failure, for fission products circulating in the coolant to approach equilibrium,* the remaining 200 days of the assumed element life was taken as operating time preceding the failure. Actually most of the fission products reach equilibrium in 200 days, and ± 100 days on this time would make only a small difference in the final activity obtained.

The area of uranium exposed by the failure has been tacitly assumed to appear after 200 days. This assumption covers all probable cases except one in which the area exposed by the failure is increasing rapidly in the period between 200 and 300 days. In such an instance, use of the final area upon removal of the element would give conservative results.

G. OPERATION OF PURIFICATION STILL

It was assumed that 25 lb per hr of polymer would be removed as residue from the system, and that it would be necessary to process from 100 to 250 pounds of coolant per hour in the purification still in order to accomplish this removal. The problem of whether fission products from the total amount of coolant processed remain with the still residue, or pass through with the purified coolant and return to the system, was taken into account by studying four separate cases of still operating conditions. In each case, however, it was assumed that all the fission products associated with 25 pounds of coolant would be removed with the 25 pounds of residue per hour.

Case 1 assumed that the fission products would not be concentrated into the residue by the still. This situation can be termed a still efficiency of zero, where still efficiency is defined as the fraction of fission products removed by the still from the processed amount of coolant returned to the system. In

*The time required to reach equilibrium is 200 to 300 days when considering Pr^{144}, Rh^{106}, and fission in the coolant. In order to avoid complexity, however, the figure of 100 days to reach equilibrium was used, and the activities from Pr^{144}, Rh^{106}, and fission in the coolant were assumed saturated.

case 1, then, only the impurities associated with the 25 pounds of residue removed per hour are deleted from the system. In this case, the amount of coolant which must be processed in order to get the 25 pounds of residue is immaterial, since the fission product concentration of this amount of coolant has been assumed to be unaffected by the still.

Cases 2, 3, and 4 assume a still efficiency of 1 (= 100 percent), and coolant processing rates of 100, 175, and 250 lb per hr, respectively. The amount of coolant deleted of all fission products is the same as the processing rate in these cases.

H. GASEOUS PRODUCTS AND DIFFUSION

Gaseous products released were assumed to remain in the coolant. It was also supposed that the magnitude of the effect of diffusion of fission products through the fuel and cladding, particularly gaseous products, would be negligible. This assumption is supported by experimental evidence[3] that diffusion of gaseous products through the fuel in the case of a cladding failure, or through aluminum cladding, is not the principal mechanism of release of fission products to the coolant in the MTR. Further support of this contention is that major activity does not diffuse through the cladding in existing reactors.

I. ADSORPTION

No study of adsorption phenomena (plating out of fission products on the system walls) for organic systems could be located by the author. Analysis of adsorption, therefore, was not included in the present work.

J. ADDITIONAL ASSUMPTIONS

1) Assumed system constants not listed above are contained in Section II

2) In Sections III and IV, several assumptions are made which are kept, for clarity in their mathematical context.

II. SYMBOLS AND ASSIGNED VALUES

The i subscript is used to designate a member of the fission product spectrum. Where the function is applicable only to direct fission products, i is replaced by A. Where the function is applicable only to daughters of direct products A, i is replaced by B.

a = area of fuel exposed to coolant by cladding failure - 1 mm^2

γ_i = fission yield of isotope i, atoms/fission

T = time of reactor operation, sec

t = time after failure (100 days = 8.64×10^6 sec for equilibrium)

t' = time following a period of reactor operation, sec

$C_i(T)$ = density of isotope i in the fuel at time T, atoms/cm^3

M_i = rate of introduction of isotope i into the coolant by corrosion, atoms/sec

r = corrosion rate 9.65×10^{-11} cm/sec (= 0.0985 mils/mo)

d = recoil range of fission products[2] in uranium = 6.35×10^{-4} cm

s = probability that a recoiling fission product originating within the volume associated with the exposed uranium area and the recoil range will get into the coolant[2] = 1/4

f = average fission rate at full power = $45.5 \times 10^6 \times 3.25 \times 10^{10} = 1.4 \times 10^{18}$ fissions/sec

f_c = effective fission rate in the coolant, fissions/sec

$N_i(t)$ = number of isotope i atoms in the coolant at time t, atoms

R_A = rate of introduction of isotope A into coolant by recoil process, atoms/sec

$F_A(t)$ = rate of production of isotope A in coolant by fission in coolant at time t, atoms/sec

V = volume of uranium in reactor = 63 elements x 5190 cm^3/element = 3.27×10^5 cm^3

ϵ = enrichment of fuel,[1] = 1.8 a/o

λ_i = physical decay constant of isotope i, sec^{-1}

λ_{sj} = removal constant of purification system, (4 cases, $j = 1, 2, 3, 4$)

γ_{ik} = number of gamma rays of energy E_k given off by isotope i per disintegration, gammas/disintegration

p = ratio of peak flux at center of core to average flux = 2.4

ϕ_c = average thermal neutron flux in the coolant in the core = 5×10^{13} n/sec-cm^2

W = weight of all coolant, 80,000 lbs = 3.63×10^7 grams

g = fraction of time coolant is in core = 0.020

σ_{500} = fission cross section of U^{235} at 500°F = 529 barns

ρ_i = density of isotope i, gm/cm^3

N_a = Avogadro's number, atoms/gm mol

A = atomic weight, gm/gm mol

D_j = total dose rate at surface of 16-inch, schedule 40 pipe from long lived fission products in coolant at equilibrium, mr/hr (for still condition j)

Q_k = geometric conversion factor for 16-inch, schedule 40 pipe to change specific activity at energy E_k to dose rate, $\frac{mr/hr}{gamma/gm\text{-}sec}$

$\Gamma(t)$ = gamma energy per fission at time t after reactor shutdown, Mev/fission

$\Sigma\Gamma(t)$ = sum of $\Gamma(t)$ over successive 50-second coolant cycles from 0 to 7200 seconds, Mev/fission

$D(h)$ = contribution to the dose rate from short lived fission products at time h along coolant cycle, mr/hr

h = time measured along coolant cycle, (= 0 at center of core), sec

c = coolant cycle time = 50 sec

III. DERIVATIONS AND CALCULATIONS

Summation of the effects of known individual fission product isotopes accounts for the order of 1 Mev of gamma energy per fission, and gives an accurate value of the gamma ray activity 1/2 hour or more after reactor shutdown. The specific fission products involved are termed "long lived" throughout this report. The total gamma energy is known to be in excess of 6 Mev/fission, the difference appearing as very short half life isotopes for which fission yields and other constants have not been determined. These products are referred to as "short lived" throughout this report. Since it was not known whether the gamma activity appearing in the coolant would be predominately this short lived activity or the result of buildup of long lived isotopes, the short and long lived activity were evaluated separately.

A. LONG LIVED FISSION PRODUCT ACTIVITY

The fission product isotopes considered as contributing to the long lived coolant activity were taken from Clark.[4] The same modifications to a few of the constants for these isotopes as used by Ashley[5] were incorporated.

Before studying the buildup of each particular fission product in the coolant as a function of time after the rupture takes place, it was necessary to determine the rate of release of fission products to the coolant by each of the three processes assumed to be contributory.

1. Corrosion

The rate of introduction of an isotope i to the coolant is equal to the product of the concentration of this isotope in the fuel, the corrosion rate, and the area of fuel exposed by the rupture, or symbolically

$$M_i = C_i(T)\, ra \quad atoms\ i/sec \qquad ...(1)$$

The function $C_i(T)$ is different for direct fission products (denoted by A) and daughter products (denoted by B); and is given by

$$C_A(T) = \frac{pfy_A}{V\lambda_A}\left(1 - e^{-\lambda_A T}\right) \quad atoms\ A/cm^3 \qquad ...(2)$$

and

$$C_B(T) = \frac{pfy_A}{V}\left[\frac{1}{\lambda_B - \lambda_A}\left(e^{-\lambda_B T} - e^{-\lambda_A T}\right) + \frac{1}{\lambda_B}\left(1 - e^{-\lambda_B T}\right)\right] \quad atoms\ B/cm^3 \qquad ...(3)$$

These equations neglect both the removal of an isotope A or B by neutron capture and the direct yield of B isotopes in fission (y_B). Actually, the cumulative yields of the B products were used for y_A in equation (3). Also, no third products with sufficient delay in the decay chain were present to warrant equations for third products.

Substituting equations (2) and (3) into (1) gives

$$M_A = \frac{pfy_A}{V\lambda_A}\left(1 - e^{-\lambda_A T}\right)ra \qquad atoms\ A/sec \qquad ...(4)$$

$$M_B = \frac{pfy_A}{V}\left[\frac{1}{\lambda_B - \lambda_A}\left(e^{-\lambda_B T} - e^{-\lambda_A T}\right) + \frac{1}{\lambda_B}\left(1 - e^{-\lambda_B T}\right)\right]ra \quad atoms\ B/sec \qquad ...(5)$$

for the number of atoms entering the coolant per second as a result of corrosion for isotopes of the A or B variety, respectively.

2. Recoil

Only direct fission products A were considered to leave the fuel by the recoil process, since the direct fission yield of B (daughter) products was neglected (see III. A. 1 above). The number of atoms of a fission product A leaving the fuel per second from a one millimeter rupture area is

$$R_A = \frac{pfy_A ads}{V} \qquad atoms\ A/sec \qquad ...(6)$$

3. Fission in the Coolant

The number of atoms of U^{235} corroded into the coolant per second is

$$M_{25} = C_{25}(T)ra = \frac{N_a \rho_u \epsilon ra}{A} \qquad atoms\ 25/sec\ , \qquad ...(7)$$

the expression to the right assuming negligible burnup.

The U^{235} is introduced to the coolant at this rate, and is removed from the coolant by the purification system, plating on the system surfaces, natural decay and fission in the coolant. Neglecting the plating effect is conservative unless there is large preferential plating of U^{235} in the core region over the rest of the system. Natural decay of U^{235} and fission in the coolant are negligible when compared to removal by the purification system and, therefore, the rate of change of the amount of U^{235} in the coolant is given by

$$\frac{dN_{25}(t)}{dt} = M_{25} - \lambda_{sj} N_{25}(t) \qquad atoms\ 25/sec \qquad ...(8)$$

Under the condition that $N_{25}(0) = 0$, this integrates to

$$N_{25}(t) = \frac{M_{25}}{\lambda_{sj}} \left(1 - e^{-\lambda_{sj} t}\right) \qquad atoms\ 25\ (in\ all\ coolant) \qquad ...(9)$$

The U^{235} in the coolant will undergo fission in that part of the cycle during which it is in the reactor core and release direct fission products into the coolant at a rate equal to

$$F_A(t) = N_{25}(t)\sigma_{500}\phi_c g y_A = \frac{N_a \rho_u \epsilon ra \sigma_{500} \phi_c g y_A}{\lambda_{sj} A} \left(1 - e^{-\lambda_{sj} t}\right) atoms\ A/sec \qquad ...(10)$$

for which the exponential term is zero for $F_A(\infty)$, the equilibrium rate.

Having formulated equations for the rate of introduction of fission products to the coolant by the three contributing processes, it is possible to find the equations for the buildup of each isotope as a function of time after a rupture occurs. For direct fission products, the rate of introduction will be the sum of equations 4, 6, and 10; and the rate of removal will be the sum of the rates of natural decay and removal by the purification system; or symbolically,

$$\frac{dN_a(t)}{dt} = M_A + R_A + F_A(t) - \lambda_A N_A(t) - \lambda_{sj} N_A(t) \qquad atoms\ A/sec \qquad ...(11)$$

The term M_A in this equation is a function of reactor operation time T, assumed to be 200 days, which will continue to increase with t. That is, the concentration of those few isotopes which have not yet reached equilibrium in the fuel at the time of failure might be expected to increase after the failure. After the rupture occurs, however, it has been assumed that 1/4th of the fission fragments being generated in the outside 6.35×10^{-4} cm of exposed fuel would escape to the coolant.

Therefore, the concentration of those isotopes which are in equilibrium at the time of failure will tend to decrease, because the effective production rate will be less than the value before the failure; while those not in equilibrium will continue to build up, but at a lower rate than preceding the failure. The bulk of the isotopes belongs to the decreasing group; of those in the group which continues to build up, only Zr^{95} and its daughter Nb^{95} contribute significantly to the final activity. In order to gain mathematical simplicity, it is convenient to assume that the concentration of the fission products in the fuel will not change after the assumed time of rupture.

From the above discussion, it is seen that the activity would decrease, with the one exception noted; and since there are several isotopes which contribute more significantly to the final activity than does the exception, the assumption made is conservative.

By considering M_A as constant after the failure time of 200 days of operation and $N_A(0) = 0$, it is possible to integrate equation (11) with the result that

$$N_A(t) = \frac{M_A + R_A + F_A(\infty)}{\lambda_A + \lambda_{sj}} \left[1 - e^{-(\lambda_{sj}+\lambda_A)t}\right] + \frac{F_A(\infty)}{\lambda_A} \left[e^{-(\lambda_{sj}+\lambda_A)t} - e^{-\lambda_{sj}t}\right] \quad atoms\ A \qquad ...(12)$$

which has an equilibrium value of (see note in Section I. F.)

$$N_A(\infty) = \frac{M_A + R_A + F_A(\infty)}{\lambda_A + \lambda_{sj}} \qquad atoms\ A \qquad ...(13)$$

This is the number of isotope A atoms in all the coolant.

Daughter products (B) will be formed in the coolant as a result of the decay of A products, and will also be released by corrosion of the fuel. They will be removed from the coolant by radioactive decay and the purification system. This can be expressed mathematically as,

$$\frac{dN_B(t)}{dt} = \lambda_A N_A(t) + M_B - (\lambda_B + \lambda_{sj}) N_B(t) \qquad atoms\ B/sec \qquad ...(14)$$

Substituting the general value for $N_A(t)$ given in equation (12) and integrating with the boundary condition that $N_B(0) = 0$, we have

$$N_B(t) = \frac{\left[1 - e^{-(\lambda_B+\lambda_{sj})t}\right]}{\lambda_B + \lambda_{sj}} \left\{\frac{\lambda_A[M_A + R_A + F_A(\infty)]}{\lambda_A + \lambda_{sj}} + M_B\right\}$$

$$+ \frac{1}{\lambda_B - \lambda_A} \left[e^{-(\lambda_A+\lambda_{sj})t} - e^{-(\lambda_B+\lambda_{sj})t}\right] \left\{F_A(\infty) - \frac{\lambda_A[M_A + R_A + F_A(\infty)]}{\lambda_A + \lambda_{sj}}\right\}$$

$$+ \frac{F_A(\infty)}{\lambda_B} \left[e^{-(\lambda_B+\lambda_{sj})t} - e^{-\lambda_{sj}t}\right] \qquad atoms\ B \qquad ...(15)$$

The behavior of this function was studied for the list of fission products in Table I to determine if there is appreciable peaking in N_B for any of the products which might result if $\lambda_A << \lambda_B$. Such is not the case for any of the products studied, and the equilibrium value of

$$N_B(\infty) = \frac{1}{\lambda_B + \lambda_{sj}} \left\{ \frac{\lambda_A [M_A + R_A + F_A(\infty)]}{\lambda_A + \lambda_{sj}} + M_B \right\} \quad atoms\ B \qquad ...(16)$$

will be attained in almost all cases with only a small error in the assumed value of t of 100 days.

Equations (13) and (16) give the number of atoms of any isotope i in the coolant at equilibrium for each of the still cases j considered. The dose rate from a 16-inch, schedule 40 pipe carrying coolant containing the equilibrium amounts of fission products is,

$$D_j = \sum_{A,K} \frac{\gamma_{AK} Q_K \lambda_A N_A(\infty)}{W} + \sum_{B,K} \frac{\gamma_{BK} Q_K \lambda_B N_B(\infty)}{W} \quad (j = 1, 2, 3, 4)\ mr/hr \qquad ...(17)$$

where $N_A(\infty)$ and $N_B(\infty)$ are, in least terms,

$$N_A(\infty) = \frac{ra\gamma_A}{\lambda_A + \lambda_{sj}} \left\{ \frac{pf}{\lambda_A V} \left[\left(1 - e^{-\lambda_A T} \right) + \frac{ds}{r} \right] + \frac{N_a \rho_{25} \epsilon \sigma_{500} \phi_c g}{A \lambda_{sj}} \right\} \quad atoms\ A \qquad ...(18)$$

$$N_B(\infty) = \frac{ra\gamma_A}{\lambda_B + \lambda_{sj}} \left\{ \frac{fp}{V} \left[\frac{\left(1 - e^{-\lambda_A T} \right)}{\lambda_A + \lambda_{sj}} + \frac{\lambda_A}{\lambda_A + \lambda_{sj}} \frac{ds}{r} + \frac{1}{\lambda_B - \lambda_A} \left(e^{-\lambda_B T} - e^{-\lambda_A T} \right) + \frac{1}{\lambda_B} \left(1 - e^{-\lambda_B T} \right) \right] + \frac{\lambda_A N_A \rho_{25} \epsilon \sigma_{500} \phi_c g}{\lambda_{sj} (\lambda_A + \lambda_{sj}) A} \right\} \quad atoms\ B \qquad ...(19)$$

The total equilibrium contribution to the dose rate from long lived fission products for each of the four still cases considered was calculated by means of equation (17) by summing the contribution from each product listed in references 4 and 5. Study of the various decay constants involved will reveal

that equations (18) and (19) can be simplified for most of the fission products. Advantage was taken of this in computing the results. The activity calculated in this section not only represents the contribution from long lived fission products to the total coolant activity during operation, but also represents the activity which will be present about two hours after reactor shutdown, when the very short lived contribution has decayed.

B. SHORT LIVED FISSION PRODUCT ACTIVITY

The relation[6]

$$\Gamma(t') = \int_{t'}^{t'+T} \alpha \tau^{-\beta} d\tau \qquad \textit{Mev/fission} \qquad \ldots(20)$$

where

α	β	valid range (sec)
1.10	0.000	$0.0 \leq \tau \leq 0.1$
0.53	0.318	$0.1 \leq \tau \leq 1.0$
0.53	0.824	$1.0 \leq \tau \leq 10.0$
1.26	1.200	$10.0 \leq \tau \leq 10^7$

gives the gamma energy per fission at time t' after reactor shutdown for any period of operation T. The short half life part of this activity was considered to come only from direct recoil out of the fuel and fission in the coolant stream, because the short lived activity released by the corrosion process will be negligible in comparison. Equation (20), which applies to situations in which the normal spectrum and distribution of uranium fission products are generated, is used in this case, with the implicit assumption that the recoil process yields a spectrum of fission products to the coolant with the same distribution as is generated by fission in the fuel.

The gamma energy given by equation (20) was evaluated as a function of decay time from 0 to 7200 seconds, at which time the calculation in Section III. A. above applies. An operating period of one second, which is the residence

time of the coolant in the core, was used. The physical significance of the curve plotted from this data is that it gives the decay of the activity picked up by a 1-sec fraction of the coolant during one pass through the core region. Another way to interpret the curve is in terms of continuous operation and the 50-second coolant cycle time. Considering any cycle of operation after equilibrium of fission product activity has been reached, the first 50-second portion of the curve gives the decay of the activity picked up by a 1-sec fraction of the coolant at the start of the cycle.

The next 50-second portion gives the decay of the activity picked up on the preceding cycle. The third 50-second portion gives the decay of the activity picked up on the second cycle preceding the one being considered, and so on. Therefore, if the decay curve is divided into 50-second intervals, and the activity in these intervals is summed, the total equilibrium activity will be obtained. This was done for the case at hand. The summation was made to 7200 seconds. The $\Gamma(t')$ curve was very nearly linear in the interval 400-7200 seconds, so the average value in the interval times the number of 50-second cycles could be added as one line of the proper slope, saving the summation of 136 lines in the interval. Actually, the first cycle contributes most of the activity. The buildup from following cycles is small, as would be expected from the fact that very short half life products are involved.

The gamma energy per fission released as a function of time along the coolant cycle having thus been determined, it was possible to convert this to dose rate at the surface of a 16-inch, schedule 40 pipe. The recoil rate given by equation (6) and the rate of fission in the coolant given by equation (10) give the effective fission rate in the coolant (f_c). The contribution to the dose rate from the short lived fission products as a function of time h along the coolant cycle is then

$$D(h) = \frac{f_c C Q_K \Sigma \Gamma(t)}{W} \qquad mr/hr \qquad \ldots(21)$$

An average energy of 1 Mev for the short lived fission products was assumed in the evaluation of $D(h)$.

IV. RESULTS

The contribution to the dose rate at the surface of a 16-inch, schedule 40 pipe, from the long lived fission products under the assumed conditions, per sq mm of uranium exposed to the coolant by the cladding failure, is given in Table I. This dose rate will be present two hours after reactor shutdown, when the effect of the short lived products has died away.

No further effort was expended to obtain the dose rate for other values of time after shutdown, since the dose rate noted above, when multiplied by failure areas deemed probable, would not present a serious problem for accessibility or maintenance.

TABLE I

EQUILIBRIUM DOSE RATES FROM CLADDING FAILURE

Purification System Case	Coolant Processing Rate (lb/hr)	Residue Removal Rate (lb/hr)	Coolant Fission Products Removal Rate (lb/hr)	Dose Rate at Surface of 16-in. Pipe ($mr/hr\text{-}mm^2$)
1	100	25	25	0.0094
2	100	25	100	0.0064
3	175	25	175	0.0056
4	250	25	250	0.0049

Table II lists the isotopes considered in calculating the dose rate from long lived products and their equilibrium specific activities. Also included are several fission products which do not produce significant gamma radiation, but which are of interest in evaluating the health physics hazards associated with coolant spillage.

The dose rate from the long lived fission products (see above) for still case 1 was added to the dose rate along the coolant cycle from short lived products. The result is the total dose rate at the surface of a 16-inch, schedule 40 pipe per sq mm of uranium exposed to the coolant by the failure, and is shown in Figure 1. The purification system has little effect on the short lived activity; the effect of the most favorable still case considered would be to reduce the values in Figure 1 by 0.0045 mr/hr.

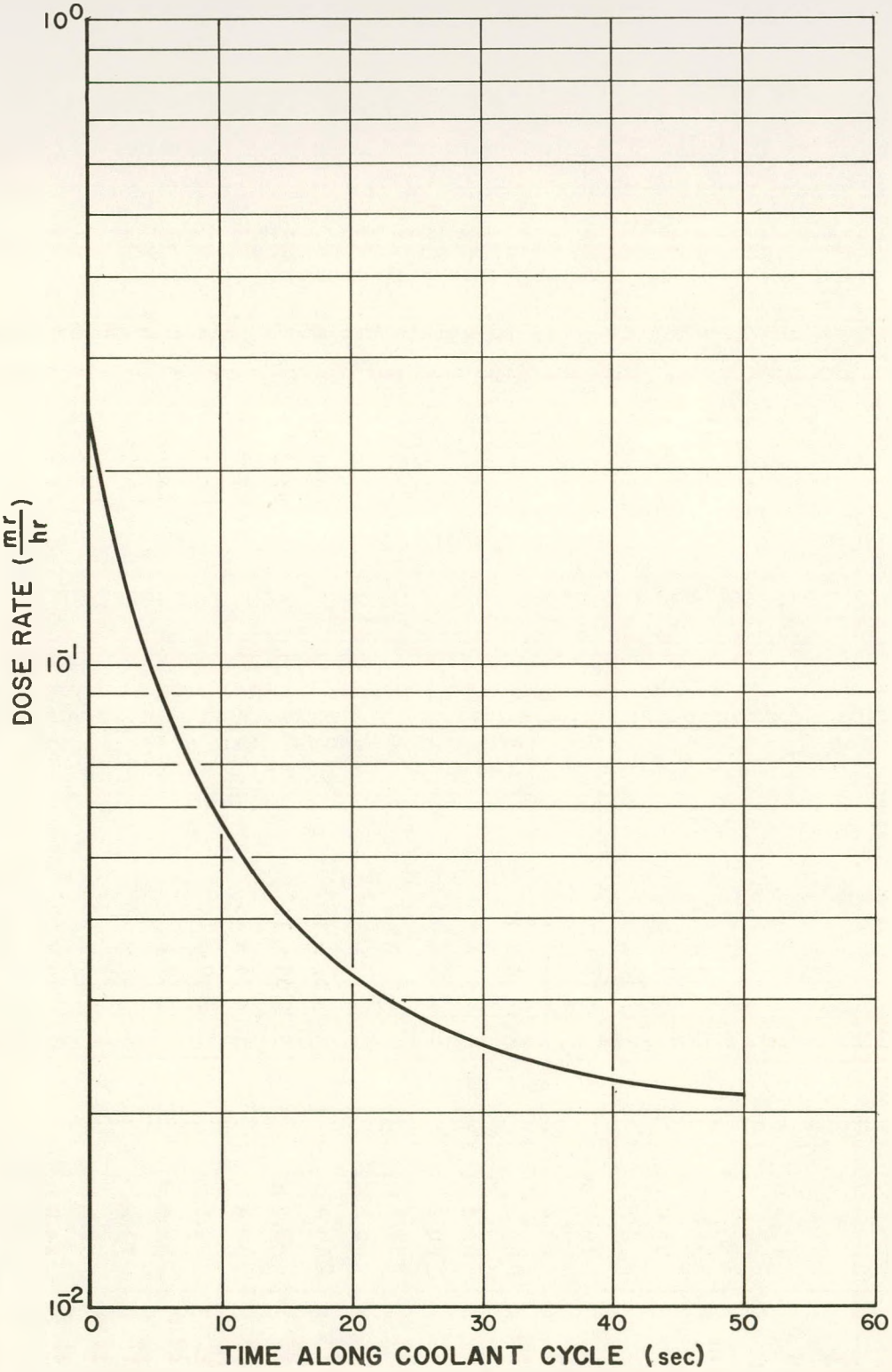

Figure 1. Equilibrium Dose Rate at Surface of 16-in. Pipe, from Fission Products Released by a 1 sq mm Fuel Cladding Failure

TABLE II

EQUILIBRIUM VALUES OF SPECIFIC ACTIVITIES IN THE COOLANT FOR ISOTOPES CONSIDERED

Isotope	Half Life	Specific Activity (disintegrations/gm-sec)	Isotope	Half Life	Specific Activity (disintegrations/gm-sec)
Br^{84}	32 m	3.70×10^{-3}	I^{134}	52.5 m	3.18×10^{-2}
Rb^{88}	17.7 m	1.73×10^{-2}	I^{135}	6.68 h	3.32×10^{-2}
Sr^{91}	97 h	2.83×10^{-2}	Xe^{135}	9.11 h	5.34×10^{-2}
Y^{91m}	51 m	1.33×10^{-3}	Cs^{136}	13.7 d	5.50×10^{-5}
Y^{91}	61 d	8.73×10^{-2}	Cs^{138}	32 m	3.33×10^{-2}
Y^{93}	10 h	3.40×10^{-2}	Ba^{139}	85 m	3.62×10^{-4}
Y^{94}	16.5 m	2.78×10^{-2}	Ba^{140}	12.8 d	5.38×10^{-2}
Zr^{95}	65 d	8.88×10^{-2}	La^{140}	10.2 h	5.75×10^{-2}
Nb^{95m}	90 h	9.09×10^{-4}	Ce^{141}	32.5 d	7.10×10^{-2}
Nb^{95}	35 d	1.43×10^{-1}	Ce^{143}	34 h	3.23×10^{-2}
Nb^{97m}	60 s	3.56×10^{-2}	Pr^{144}	17.5 m	5.01×10^{-2}
Nb^{97}	72.1 m	3.57×10^{-2}	Nd^{147}	11.6 d	2.20×10^{-2}
Mo^{99}	68.3 h	3.93×10^{-2}	Sm^{153}	47 h	9.14×10^{-4}
Ru^{103}	39.8 d	5.01×10^{-2}	Eu^{156}	15.4 d	1.22×10^{-4}
Ru^{105}	4.5 h	5.05×10^{-3}	Sr^{89}	53 d	7.91×10^{-2}
Rh^{106}	30 s	4.74×10^{-3}	Sr^{90}	28 y	2.63×10^{-3}
Sn^{125}	9.5 d	1.59×10^{-4}	Ce^{144}	282 d	6.36×10^{-2}
Te^{129m}	33.5 d	2.37×10^{-3}	Pr^{143}	13.7 d	6.77×10^{-2}
Te^{129}	72 m	1.07×10^{-2}	Xe^{133}	5.27 d	7.11×10^{-2}
Te^{131m}	30 h	2.60×10^{-3}	Kr^{85}	10 y	4.55×10^{-4}
Te^{131}	24.8 m	1.55×10^{-2}	Cs^{137}	33 y	2.55×10^{-3}
I^{131}	8.14 d	2.15×10^{-2}	Rh^{105}	36.5 h	8.90×10^{-3}
Te^{132}	77.7 h	2.19×10^{-2}	Pm^{147}	2.6 y	1.28×10^{-2}
I^{132}	2.4 h	3.17×10^{-2}	Te^{127}	90 d	5.81×10^{-4}
I^{133}	20.8 h	2.67×10^{-2}	Sm^{151}	122 y	5.43×10^{-5}

For the subject reactor, delayed neutron emitting isotopes (Br^{87}, I^{137}, Br^{89}, I^{139}, As^{85}, and Li^{9}) in the coolant have been studied previously at AI. This study shows that the dose rate in mrem/hr from delayed neutrons, for the conditions of this report, was insignificant.

V. CONCLUSIONS

From the results shown in Figure 1 it is seen that the maximum dose rate at the surface of a 16-in. schedule-40 pipe for a 1 mm^2 cladding failure will be 0.25 mr/hr. (The dose rate will attain this value 200 days after the cladding failure, under the conditions of a 100-day period of operation preceding the cladding failure, and with the reactor operating at full power during the entire 300-day period.) The dose rate is proportional to exposed area, so that the maximum dose rate for a cladding failure as large as 1000 mm^2 (= 10 cm^2) would be 250 mr/hr. Considering a fuel cladding failure of this size, the resultant dose rate is about comparable to the dose rates expected from normal coolant impurities, corrosion, and recoil products. It is evident, then, that full power reactor operation could continue with many pinhole cladding failures or several larger exposures without raising the radiation levels in the process area of the plant appreciably over normal values.

Several of the assumptions, upon which the results of this study are dependent, are subject to question. In particular:

a) The corrosion rate (including erosion) of uranium under reactor operating conditions of temperature, coolant flow, etc, might differ from the value determined under static conditions in the laboratory.

b) It may not be valid to ignore diffusion as a source of fission product leakage from the fuel.

c) Adsorption effects on system surfaces could raise dose rates above those calculated.

In view of the favorable results of this study, and the experience obtained in operation of the Organic Moderated Reactor Experiment (OMRE), it is not deemed advisable to enter into detailed investigations of the validity of the questionable assumptions at this time. Rather, exposure of an intentionally defected fuel element in the OMRE, or in an inpile loop in some other reactor, would be worthwhile to provide, in an integral experiment, an evaluation of the results obtained in this report.

REFERENCES

1. C. W. Wheelock. "11,400 Kilowatt Nuclear Power Plant," NAA-SR-2304, December 20, 1957.

2. J. Chernick and I. Kaplan, "The Escape of Fission Products from Uranium Rod," BNL-20, November 3, 1948.

3. J. H. Rainwater, "Maintenance of Primary Coolant Water Quality in the Materials Testing Reactor," IDO-16322, February 21, 1957. See 3-33.

4. F. H. Clark. "Decay of Fission Product Gammas," NDA-27-39, December 30, 1954. See Table I.

5. R. L. Ashley, "Graphical Aids in the Calculation of the Shielding Requirements for Spent U^{235} Fuel," NAA-SR-1992, November 15, 1957.

6. "Reactor Engineering Division Quarterly Report (for) September 1, 1952 through November 30, 1952," ANL-4951, p 10. The equation in the reference was modified to the form of the Way-Wigner equation in the region $10 \leq \tau \leq 10^7$ *sec*.