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SPECIFIC ACTIVITY OF THE NPR
PRIMARY COOLANT LOOP

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SPECIFIC ACTIVITY OF THE NPR
PRIMARY COOLANT LOOP

INTRODUCTION

In a coolant system such as NPR's, the coolant activity level increases with each succeeding pass through the reactor flux until a saturation limit is reached. Therefore, the activity level of the NPR coolant system will be much higher than that of the old reactor once-through systems. The following report is the determination of the specific activities (disintegrations/cc·sec) of the various coolant impurities which determine the total activity of the coolant system.

SUMMARY

Of all the thermal and fast neutron reactions contributing to the activity level of the NPR primary loop coolant, the $^{16}\text{O}(n,p)^{16}\text{N}$ fast neutron reaction is the primary one. ^{16}N 's specific activity builds up until it reaches a saturation level of 5.0×10^5 dis/cc·sec. The primary decay gamma of ^{16}N has an energy of 6.13 Mev.

Two neutron sources in the coolant are ^{17}N and the (γ,n) reaction of deuterium. ^{17}N has a specific activity of 1.08×10^2 dis/cc·sec. (1.1 Mev neutron per disintegration), which decays quickly because ^{17}N has a 4.14 sec. half life. The (γ,n) reaction becomes as prominent as ^{17}N to the over-all neutron activity after about 5 seconds transit time from the active zone.

Tritium concentration is increased by the introduction of lithium into the primary loop; however, the bleed system (550 gpm) decreases the total activity level of tritium in the primary loop. Lithium increases the tritium level by a factor of 32.8. The specific activity for tritium because of Li (1.16×10^2 dis/cc·sec) is based on a Li concentration of 1.3 ppm.

Cr^{51} , Fe^{59} , and Co^{60} specific activities tend to build up by plating in the reactor for long periods of time. Therefore, calculated values will possibly be modified at a later date. Based on existing data, the actual specific activity values may run 100 times higher than calculated.

DISCUSSION

I. Expressions for Calculation of Specific Activity

The equations for calculation of the specific activity of effluent impurities are based on the following definitions:

- $A_n(t)$ - specific activity after n cycles (dis/cc·sec)
- t' - reactor transit time (seconds)
- T - total primary loop transit time (seconds)
- Φ - average thermal neutron flux in the coolant region (neutron/cm²·sec)
- N - concentration of given impurity in coolant (nuclei/cc)
- σ - cross section for thermal neutron absorption of given isotope (cm²)

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- λ - decay constant of activated nuclide (1/sec)
 T - half life of activated nuclide
 Q_b - coolant bleed rate (cc/sec)
 Q_1 - reactor flow rate (cc/sec)

For a recycling coolant system the specific activity after n cycles is

$$A_n(t) = \frac{\Phi N_0 (1 - e^{-\lambda t'}) (1 - e^{-(n+1)\lambda T})}{(1 - e^{-\lambda T})} \quad (1)$$

As n approaches infinity, then $A_n(t)$ becomes the following:

$$A_\infty(t) = \frac{\Phi N_0 (1 - e^{-\lambda t'})}{(1 - e^{-\lambda T})} \quad (2)$$

if; $\lambda t' \ll 1$ $\lambda T \ll 1$

then; $(1 - e^{-\lambda t'}) \approx \lambda t'$ $(1 - e^{-\lambda T}) \approx \lambda T$

When the preceding approximations are substituted into equation (2), the result is

$$A_\infty(t) = \Phi N_0 \frac{t'}{T} \quad (3)$$

For the specific activity of a second generation radioactive nuclide, one which has resulted from the activation of an isotope that is the result of a thermal neutron activation of an impurity isotope, the following is used:

$$A_2(t) = \frac{\Phi^2 N_0 \sigma_1 \sigma_2 (1 - e^{-\lambda_1 t'}) (1 - e^{-\lambda_2 t'})}{\lambda_1 (1 - e^{-\lambda_1 T}) (1 - e^{-\lambda_2 T})} \quad (4)$$

where: σ_1 is the thermal cross section value of the first generation isotope,
 σ_2 is the cross section value of the second generation thermal neutron reaction.
 λ_1 is the decay constant for first generation nuclide.

The derivation for equation (4) is based on the following: the differential equation covering the buildup of activity of a second generation isotope is

$$\frac{d[N_2(t)]}{dt} = \Phi N_1 \sigma_2 - \Phi N_2(t) \sigma_3 - N_2(t) \lambda_2$$

where: $\Phi N_1 \sigma_2$ is second generation buildup

$\Phi N_2(t) \sigma_3$ is second generation burnup

$N_2(t) \lambda_2$ is second generation decay

$$\frac{d[N_2(t)] e^{(\lambda_2 + \sigma_3)t}}{dt} = \Phi N_1 \sigma_2 e^{(\lambda_2 + \sigma_3)t} \quad *$$

$$A_n(t)_2 = \frac{\Phi N_0 \sigma_2 [1 - e^{-(\lambda_2 + \sigma_3)t'}] [1 - e^{-(n+1)(\lambda_2 + \sigma_3)T}]}{(\lambda_2 + \sigma_3) [1 - e^{-(\lambda_2 + \sigma_3)T}]}$$

* For solution technique see: The Reactor Handbook, AECD-3646, Vol. 2 Engineering, May 1955, Chapter 1.12.

But, the number of first generation nuclides (N_1) is

$$\frac{\Phi N_0 [1 - e^{-(\Phi \sigma_2 + \lambda_1)t'}] [1 - e^{-(n+1)(\Phi \sigma_2 + \lambda_1)T}]}{(\Phi \sigma_2 + \lambda_1) [1 - e^{-(\Phi \sigma_2 + \lambda_1)T}]}$$

Therefore:

$$A_n(t)_2 = \frac{\Phi^2 N_0 \sigma_2 [1 - e^{-(\Phi \sigma_2 + \lambda_1)t'}] [1 - e^{-(\Phi \sigma_2 + \lambda_1)t'}] [1 - e^{-(n+1)(\Phi \sigma_2 + \lambda_1)T}] [1 - e^{-(n+1)(\Phi \sigma_2 + \lambda_1)T}]}{(\Phi \sigma_2 + \lambda_1)(\Phi \sigma_3 + \lambda_2) [1 - e^{-(\Phi \sigma_3 + \lambda_2)T}] [1 - e^{-(\Phi \sigma_2 + \lambda_1)T}]}$$

As n approaches infinity and if;

$$\Phi \sigma_2 \ll \lambda_1, \quad \Phi \sigma_3 \ll \lambda_2$$

then

$$A_\infty(t)_2 = \frac{\Phi^2 N_0 \sigma_2 (1 - e^{-\lambda_1 t'}) (1 - e^{-\lambda_2 t'})}{\lambda_1 \lambda_2 (1 - e^{-\lambda_1 T}) (1 - e^{-\lambda_2 T})}$$

To obtain specific activity of second generation nuclide, multiply the preceding by λ_2 . Thus, equation (4) is established.

For constant feed and bleed rates, the following adjustment is made to equation (2):

$$A_\infty(t)_b = \frac{\Phi N_0 (1 - e^{-\lambda t'})}{(1 - e^{-\lambda T}) [1 + \frac{Q_b}{Q_f} (\frac{1}{\lambda T} - 1)]} \quad * (5)$$

When the bleed rate is 550 gpm and $\lambda t' \ll 1$, $\lambda T \ll 1$, then equation (5) becomes

$$A_\infty(t)_b = \Phi N_0 \left[\frac{2.81 \times 10^{-2}}{1 + 3.2 \times 10^{-3} (\frac{1}{0.72} - 1)} \right] \quad (6)$$

Numerical values in equation (6) are based on present reactor parameters. Equation (6) determines the specific activity of radioactive isotopes whose half life is greater than ten minutes. When a given radioactive isotope has a half life less than ten minutes, equation (2) or (3) is used.

* HW-55834, E. R. Astley. Based on equations (9) and (12).

II. Specific Activity Values

The specific activity values are given in Table I. Values in Table I are based on time zero (coolant just leaving active zone on rear face) and an infinite number of cycles through the active zone.

$O^{16}(n,p)N^{16}$ is the dominant reaction because of the large concentration of natural O^{16} as a component of the effluent water. N^{16} also yields the most energetic gammas (6.13⁷³, 7.12⁵ Mev) during decay as compared to the gamma energies of isotopes from other activated effluent impurities.

In the N^{17} decay scheme a 1.1 Mev neutron is produced. N^{17} is the coolant's main neutron source for approximately the first 5 seconds after leaving the active zone. (see Figure 4) Then the (γ,n) reaction of deuterium begins to contribute significantly as a neutron source (compare Figures 2 and 4).

The addition of lithium to the effluent, through the means of a pH controller, increases the tritium level in the loop coolant. The tritium level is increased by a factor of 32.8 through the introduction of lithium. (See Table 1, comparison between $H^2(n,\gamma)H^3$ and $Li^6(n,\alpha)H^3$). Fortunately, the tritium level in the primary loop is decreased by the bleed system of the loop as bleeding essentially adds an extra decay constant.

Specific activity values in Table I are of a saturated-steady state nature and are based on a 550 gpm bleed rate. However, experimental data of activation levels of Cr^{51} , Fe^{59} , and Co^{60} have given activity plots which do not reach a steady state condition. There is a tendency for activity of these particular coolant impurities to continue to increase with time. At the present time, the explanation for this is a plating action in the reactor; thus, instead of a constant, even mixture of impurities in the effluent water, large amounts of particular isotopes are plating in the reactor for long periods of time. After being activated in the reactor to higher levels, these isotopes will return to the coolant and circulate through the primary loop. Consequently, the specific activity of Cr^{51} , Fe^{59} , and Co^{60} builds up. As the values in Table I are of a steady state nature, the specific activities of Cr^{51} , Fe^{59} , and Co^{60} will possibly be modified at a later date. Within the next few months a more definite picture of this plating action should be obtained.* At present, data indicate that the specific activity of Cr^{51} , Fe^{59} , and Co^{60} may run 100 greater than values in Table I.

* Personal communication from L. D. Perrigo.

TABLE I
(Based on 550 gpc Bleed Rate)

Isotope	Fast Neutron Reactions	Saturated Specific Activity (dis/cc-sec)	Half Life	Decay Products $Q(\text{Mev})$	$\lambda(\text{Mev})$
N ¹⁶	O ¹⁵ (n,p)N ¹⁶	5.00 x 10 ⁵	7.4s	4.3 ⁴⁸ , 10.4 ²⁶ , -	6.13 ⁷³ , 7.12 ⁵ , -
N ¹⁷	O ¹⁷ (n,p)N ¹⁷	1.08 x 10 ²	4.14s	3.7(neutron-1.1 Mev)	(None)
F ²⁰	Ne ²³ (n, α)F ²⁰	36.9	11s	5.2 ¹⁰⁰	1.63 ¹⁰⁰
Ne ²³	Ne ²³ (n,p)Ne ²³	3.00	38s	4.3 ⁶⁷ , 3.95 ³² , -	0.44 ³³ , -
Mg ²⁷	Al ²⁷ (n,p)Mg ²⁷	5.63	9.5m	1.75 ⁷⁰ , 1.57 ³⁰	0.83 ⁷⁰ , 1.01 ³⁰
Mn ⁵⁴	Fe ⁵⁴ (n,p)Mn ⁵⁴	9.86 x 10 ⁻⁴	280d	EC ¹⁵⁰	0.84 ¹⁰⁰
Co ^{58m}	Ni ⁵⁸ (n,p)Co ^{58m}	2.36	9.0h	IT ¹⁵⁰	0.025
Co ⁵⁸	Ni ⁵⁸ (n,p)Co ⁵⁸	2.96 x 10 ⁻²	71d	EC ⁶⁵ , 8 ⁺ 4.8 ¹⁵	0.81 ¹⁰⁰ , 1.62 ^{0.5}
O ¹⁹	O ¹⁸ +n $\rightarrow$ O ¹⁹	6.31 x 10 ³	29s	3.25 ⁵⁸ , 4.60 ⁴²	0.200 ⁹⁶ , 1.36 ⁵⁴
*Na ²⁴	Na ²³ +n $\rightarrow$ Na ²⁴	4.12 x 10 ²	15.0h	1.39, -	2.75 ¹⁰⁰ , 1.37 ¹⁰⁰ , -
*Al ²⁸	Al ²⁷ +n $\rightarrow$ Al ²⁸	6.85 x 10 ²	2.3m	2.87 ¹⁰⁰	1.78 ¹⁰⁰
*Si ³¹	Si ³⁰ +n $\rightarrow$ Si ³¹	6.74	2.6s	1.48, -	1.26 ⁰⁷
*P ³²	P ³¹ +n $\rightarrow$ P ³²	13.8	14.3d	1.71	(none)
*S ³⁵	S ³⁴ +n $\rightarrow$ S ³⁵	8.41 x 10 ⁻³	87d	0.157	(none)
*S ³⁷	S ³⁶ +n $\rightarrow$ S ³⁷	9.23 x 10 ⁻²	5.0m	1.65 ³⁰ , 4.7 ¹⁰	3.1 ¹⁰
A ³⁷	A ³⁶ +n $\rightarrow$ A ³⁷	1.00 x 10 ⁻²	35d	EC ¹⁰⁰	(none)

* Includes fission-spectrum-neutron activation contribution.

TABLE I (cont.)

Isotope	Thermal Neutron Reactions	Saturated Specific Activity (dis/cc-sec)	Half Life	E_{γ} (Mev)	Decay Products λ (Mev)
Cl^{38}	$Cl^{37}+n \rightarrow Cl^{38}$	1.26×10^2	37.3m	$4.8^{31}_{1.1^{31}}$, 2.8^{16}	$2.1^{47}_{1.6^{31}}$
A^{41}	$A^{40}+n \rightarrow A^{41}$	40.7	1.83h	$1.20^{99}_{2.49^1}$	1.2999
Ca^{49}	$Ca^{48}+n \rightarrow Ca^{49}$	3.51×10^{-1}	8.7m	$2.0^{89}_{0.89}$, -	$3.10^{89}_{4.05^8}$, 4.68^3
$*Cr^{51}$	$Cr^{50}+n \rightarrow Cr^{51}$	2.85×10^{-1}	27.8d	EC	0.32^{10} , -
Cr^{55}	$Cr^{54}+n \rightarrow Cr^{55}$	5.56	3.6m	2.8^{100}	(none)
$*Fe^{55}$	$Fe^{54}+n \rightarrow Fe^{55}$	7.11×10^{-3}	2.9y	EC ¹⁰⁰	(none)
$*Mn^{56}$	$Mn^{55}+n \rightarrow Mn^{56}$	1.046×10^3	2.58h	$2.86^{60}_{1.05^{24}}$, 0.75^{15}	$0.84^{99}_{1.81^{23}}$, 2.11^{14}
$*Fe^{59}$	$Fe^{58}+n \rightarrow Fe^{59}$	1.01×10^{-2}	45d	$0.46^{53}_{0.27^{46}}$, $1.56^{0.3}$	0.19^3 , -
Co^{60m}	$Co^{59}+n \rightarrow Co^{60m}$	1.54×10^3	10.5m	$IT^{99.7}_{1.54^{0.3}}$	$0.659^{99.7}_{1.33^{0.3}}$
$*Co^{60}$	$Co^{60m} \rightarrow Co^{60} + \gamma$ $Co^{59}+n \rightarrow Co^{60}$	2.38×10^{-1}	5.28y	0.31^{100} , $1.48^{0.01}$	$1.33^{100}_{1.17^{100}}$, -
Co^{61}	$Co^{60}+n \rightarrow Co^{61}$	2.78×10^{-4}	1.65h	1.22^{100}	0.76^{100}
Ni^{63}	$Ni^{62}+n \rightarrow Ni^{63}$	2.14×10^{-3}	80y	0.067^{100}	(none)
Cu^{64}	$Cu^{63}+n \rightarrow Cu^{64}$	47.7	12.8h	$EC^{43}_{0.57^{38}}$, 0.66^{19}	1.34^1
Ni^{65}	$Ni^{64}+n \rightarrow Ni^{65}$	7.14	2.56h	$2.10^{69}_{0.62^3}$, 1.01^8	$1.49^{20}_{1.11^{10}}$, 0.37^3
Cu^{66}	$Cu^{65}+n \rightarrow Cu^{66}$	51.5	5.1m	$2.63^{91}_{1.59^9}$, -	1.049
Zr^{95}	$Zr^{94}+n \rightarrow Zr^{95}$	1.14×10^{-3}	65d	$0.40^{55}_{0.36^{43}}$, 0.89^2	$0.72^{55}_{0.76^{43}}$
Zr^{97}	$Zr^{96}+n \rightarrow Zr^{97}$	2.05×10^{-2}	17h	1.91^{90} , -	$0.5-2.6$

* Includes fission-spectrum-neutron activation contribution.

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TABLE I (cont.)

Isotope	Thermal Neutron Reactions	Saturated Specific Activity (dis/cc-sec)	Half Life	β (Mev)	Decay Products γ (Mev)
H^3	$Li^6(n,\alpha)H^3$	1.16×10^2	12.26y	0.0186^{100}	(none)
H^3	$H^2(n,\gamma)H^3$	3.54			
Li^8	$Li^7+n \rightarrow Li^8$	4.08×10^4	0.84s	13,2(3 Mev)	

s - seconds

m - minutes

h - hours

d - days

y - years

EC - electron conversion

IT - Isomeric transition

A "dash" after decay energies indicates traces of lesser intensity.

Superscripts on decay energies are the number of occurrences in 100 disintegrations.

Several decay energies are not superscribed. It is assumed that these occur 100 times out of 100 disintegrations.

TABLE II

ION CONCENTRATIONS IN NPR COOLANT WATER*Dissolved Metals

Iron	0.05 ppm	4.48×10^{15} nucl/cc
Nickel	Less than 0.01 ppm	9.13×10^{14} nucl/cc
Chromium	Less than 0.01 ppm	8.31×10^{14} nucl/cc
Copper	Less than 0.001 ppm	8.49×10^{13} nucl/cc
Aluminum	0.05 ppm	3.01×10^{15} nucl/cc
Manganese	0.002 ppm	1.63×10^{14} nucl/cc
Calcium	0.01 ppm	2.33×10^{14} nucl/cc
Cobalt	Less than 0.001 ppm	9.08×10^{13} nucl/cc
Lithium	0.7 to 1.3 ppm	5.98×10^{16} nucl/cc
Sodium	0.2 ppm	5.09×10^{15} nucl/cc
Zirconium	0.01 ppm	4.30×10^{14} nucl/cc

Anions

Chlorides	Less than 0.05 ppm	1.28×10^{15} nucl/cc (Cl)
Sulfates	Less than 1.0 ppm	2.30×10^{15} nucl/cc (S)
Phosphates	Less than 0.02 ppm	4.64×10^{15} nucl/cc (P)

Dissolved Gases

Argon	Less than 0.10 ppm	1.38×10^{14} nucl/cc
Carbon dioxide	Less than 0.05 ppm	The remaining gases
Nitrogen	Less than 1.0 ppm	yield isotopes whose
Carbon monoxide	Less than 0.02 ppm	A(t)'s are insignificant.
Methane	Less than 0.02 ppm	
Helium	Normally less than 0.02 ppm could be high at times.	
Hydrogen	1.3 to 2.3 ppm	
Oxygen	Less than 0.005 ppm	
Ammonia	Normally less than 1.0 ppm could be high at times.	

Other Materials

Hydrogen peroxide	Less than 0.10 ppm	
Silicon dioxide	Less than 0.05 ppm	3.83×10^{14} nucl/cc (Si)
Crud**	0.025 to 0.050 ppm	

* Personal communication, W. D. Baird.

** Crud consists primarily (70-80%) of Fe_3O_4 with other oxides. The nucl/cc value given for Fe includes contribution from Fe_3O_4 .

Figure 1 is a plot of specific activity for different gamma energy ranges expressed in Mev/cc-sec versus time after shutdown. Plot I is a total of the respective energy ranges. The curves in Figure 1 and Figures 3 through 13 can also be used for determining the specific activity after a given transit time from the active zone on the rear face has elapsed. Time zero occurs when the coolant just leaves the active zone on the rear face.

For the first 80 sec., N^{16} controls the plot of the total in Figure 1; but as time increases from this point, the longer half-life isotopes control Plot I.

The following constants were used in evaluating the specific activity:

Reactor transit time	1.65 sec
Primary loop transit time	58.7 sec
Average thermal flux	$3.0 \times 10^{13} \text{ n/cm}^2 \cdot \text{sec}$
Fission spectrum flux	$1.88 \times 10^{13} \text{ n/cm}^2 \cdot \text{sec}$
Coolant water density (in reactor)	0.834 g/cc
Reactor flow rate	$1.03 \times 10^7 \text{ cc/sec}$ ($71.4 \times 10^6 \text{ lb/hr}$)
Bleed rate	$3.47 \times 10^4 \text{ cc/sec}$
Reactor power level	4000 MW

III. (γ, n) Reaction of Deuterium

As the threshold energy for the (γ, n) reaction of deuterium is only 2.23 Mev, the abundance of the 6.13 Mev gamma from N^{16} decay presents the primary source for the (γ, n) reaction. This preceding factor coupled with the relatively large concentration of deuterium yields an additional neutron source in the primary loop coolant.

Figure 2 is a plot of the number of neutrons/cc-sec derived from (γ, n) reaction versus time lapse after leaving the active zone on the rear face. The major pipe sizes are considered on this plot because the number of n/cc-sec through photo-disintegration is dependent on pipe size. Based on the 6.13 Mev gamma, the approximate energy of the resulting neutron is 2 Mev.

The source strength of the effluent at the middle of the rear face connector is 6.5 n/cc-sec and $5.37 \times 10^{-2} \text{ n/cc-sec}$ for the front face connector.

IV. Decay Curves (Figures 3-13)

Individual decay curves for isotopes that contribute significantly to coolant activity are illustrated in Figures 3 through 13. These plots are specific activity versus time after shutdown or transit time after leaving the active zone on rear face.

The following definitions apply to Figures 3-13:

$A(t)$ - specific activity in dis/cc·sec

t - time after shutdown or transit time from active zone on rear face in sec β^- and γ energies are in Mev. The superscript digits of β^- and γ energies represent the number of occurrences in 100 disintegrations.

To change dis/cc·sec to curies/cc multiply by 2.7×10^{-11} .

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FIGURE 1

SPECIFIC ACTIVITY (Mev/cc-sec) FOR DIFFERENT
 GAMMA ENERGY RANGES VERSUS TIME AFTER SHUTDOWN
 OR TRANSIT TIME FROM ACTIVE ZONE OF REAR FACE

$A(t)$ - Mev/cc-sec

t - sec

Plot I is the total of II through VIII.

Plot	Gamma Energy Range (Mev)
II	6.13 - 7.12 (^{16}Mg)
III	2.5 - 3.0 (^{24}Na)
IV	2.0 - 2.5
V	1.5 - 2.0
VI	1.0 - 1.5
VII	0.5 - 1.0
VIII	0.01 - 0.5

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FIGURE 2

PHOTO DISINTEGRATION NEUTRON SOURCE
STRENGTH VERSUS TIME FOR DIFFERENT SIZE
PIPE.

Pipe dimensions are in inches and are
outside diameters.

$A(t)n$ - neutron/cc·sec
 t - sec

FIGURE 5

0.19

$\tau = 29$ sec

θ^-

3.2558 4.6042 γ 0.2096 1.3654

FIGURE 2

A^4 $\tau = 1.83h$

1.20⁹⁹ 2.49¹ 1.29⁹⁹

FIGURE 10

M_n^{56}
 $\tau = 2.58n$
 θ^-
 2.8660
 1.0524
 0.7515
 0.8499
 1.8123
 2.1114

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