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SUBJECT: Na^{24} Activity in the HFIR Primary Coolant Water

TO: W. R. Gall

FROM: H. A. McLain

Summary

The Na^{24} activity in the HFIR primary coolant water is calculated to be normally in the range of 1.4×10^5 to 6×10^5 dis/min·ml.

Introduction

Sodium-24 has been found in the water coolant of reactors with aluminum clad fuel elements, and it must be considered for the shielding of the primary water system.[1,2] This isotope, which has a half-life of 15.06 hr, is produced in the reactor by the Al^{27} (n,α) Na^{24} reaction. For each disintegration of Na^{24}, a 2.76 Mev gamma photon and a 1.38 gamma photon are released.[3]

Method of Calculation and Results

Longtin indicated that the rate of addition of Na^{24} to the primary water of a reactor with aluminum clad fuel elements is approximately equal to the rate of Na^{24} production in the oxide films on these elements.[4,5] It is assumed for the purposes of these calculations that the Na^{24} is removed from the oxide film immediately after its formation.

The rate of production of Na^{24} in the oxide film may be computed by the use of effective threshold energies and effective cross sections.[6,7] This method assumes that the rate of reaction between aluminum and fast neutrons can be approximated by the relation:

$$\text{Reaction rate} = \int_0^\infty \Sigma(E)\phi(E)dE \approx \Sigma\ (8.7\ \text{Mev}) \int_{8.7\ \text{Mev}}^\infty \phi(E)dE,$$

where

$$\begin{aligned} 8.7\ \text{Mev} &= \text{effective threshold energy} \\ \Sigma(8.7\ \text{Mev}) &= \text{effective cross section} \\ &= 0 \text{ for } E < 8.7\ \text{Mev} \\ &= N\sigma\ (8.7\ \text{Mev}) \text{ for } E \geqslant 8.7\ \text{Mev} \end{aligned}$$

Roys and Shure[6] states that the effective cross section $\sigma(8.7\ \text{Mev})$ for Al^{27} is 177 mb.

Griess estimates that the thickness of the oxide film on the HFIR fuel plates will build up after 10 days operation at 100 Mw to approximately ½ mil for a water pH of 5.0 and 2 mil for water pH of 7.0. Assuming the density of this film to be 4 g/cc and the composition of this film to be $Al_2O_3 \cdot H_2O$, the effective cross section for the $A^{27}(n,\alpha)\ N^{24}$ reaction are:

Water pH = 5.0 $$\Sigma(8.7\ \text{Mev}) = 1.05 \times 10^{-6}\ \frac{\text{cm}^2}{\text{cm}^2\ \text{surface}}$$

Water pH = 7.0 $$\Sigma(8.7\ \text{Mev}) = 4.2 \times 10^{-6}\ \frac{\text{cm}^2}{\text{cm}^2\ \text{surface}}$$

To calculate the effective neutron flux in the fuel region of the reactor, use was made of the results computed by Cheverton[9,10] using 16 region, 34 group, diffusion theory (GNU code). The effective neutron flux was extrapolated from the flux in the 4 to 10 Mev energy range (the high energy range considered by the GNU code) by the relation,[9]

$$\int_{8.7}^{\infty} \phi(E)dE = \left[\int_{4}^{10} \phi(E)dE\right] \frac{\left[\int_{8.7}^{\infty} e^{-(\sqrt{2E}\ -\ E)}\, dE\right]}{\left[\int_{4}^{\infty} e^{-(\sqrt{2E}\ -\ E)}\, dE\right]}$$

where E is the energy in Mev. The value of the effective neutron flux

$$\int_{8.7}^{\infty} \phi(E)dE$$

computed is $4.61 \times 10^{12}\ \frac{\text{neuts}}{\text{sec-cm}^2}$.

The HFIR core is assumed to contain 190 fuel plates which are 22 in. long and 3.65 in. wide and 380 fuel plates which are 22 in. long and 3.11 in. wide. Assuming both sides of these plates are exposed to the cooling water, the aluminum surface area is 8.24×10^4 in.^{2} or 5.32×10^5 cm^2.

With the above values, the reaction rate or the rate of addition of Na^{24} to the HFIR primary coolant is:

	Reaction Rate
Water pH = 5.0	2.56×10^{12} atoms/sec
Water pH = 7.0	1.024×10^{13} atoms/sec

Equilibrium conditions can be assumed for the Na^{24} concentration in the primary coolant since the half-life of Na^{24} is relatively long (15.06 hr). Writing a material balance for the Na^{24} concentration,

$$\frac{dN}{dt} = 0 = \frac{RR}{V} - \alpha N - \lambda N$$

or

$$N = \frac{RR}{V(\alpha + \lambda)}$$

where

N = Na^{24} concentration

RR = reaction rate

V = volume of the coolant system

α = cleanup constant of system

$$= \frac{\text{flow rate through the demineralizer}}{\text{system volume}}$$

λ = decay constant of Na^{24} .

Assuming the coolant system volume V to be 30,000 gal, the demineralizer flowrate to be 200 gpm, and the decay constant λ to be 0.000767 min^{-1}, the concentrations and activities of Na^{24} in this system are:

Demineralizer Flow Rate	Water pH	Concentration N	Activity Nλ
200 gpm	5.0	1.82×10^{8} $\frac{\text{atoms}}{\text{ml}}$	1.40×10^{5} $\frac{\text{dis}}{\text{min ml}}$
200	7.0	7.3×10^{8}	5.6×10^{5}
0	5.0	1.77×10^{9}	1.36×10^{6}
0	7.0	7.1×10^{9}	5.4×10^{6}

Comparison with the Extrapolation of the ORR and LITR Data

Cole and Cox found that the Na^{24} activity in the ORR and the LITR cooling waters could be correlated by the relation[11,12]

$$C = \frac{K\,P\,N}{F(\alpha + \lambda t)}$$

where

C = Na^{24} activity in coolant, dis/min·ml

K = constant of proportionality

P = power level of the reactor, Mw

F = flow rate of coolant, gpm $\times 10^{-3}$

α = fraction of coolant flow rate through the demineralizer

λ = decay constant of Na^{24}, min^{-1}

t = coolant circuit time, min

From measurements on the ORR, the constant K in this relation has a value of 11.7.[12]

Normally, the number of fuel elements in the ORR is 24.[13] The quantity N in the above relation can be assumed proportional to the aluminum exposed to the cooling water in the reactor core. This is considered a good assumption in view of the method mentioned above. Assuming that the ORR contains 24 fuel elements with 19 plates each which are 24-5/8 in. long and 28 in. wide,[13] the aluminum surface area in the ORR core is 6.29×10^{4} $in.^{2}$.. Therefore, a psuedo N value for the HFIR core is $24 \times 8.25 \times 10^{4}/6.29 \times 10^{4} = 31.5$.

Assuming the following parameters for the HFIR,

$P = 100$ Mw

$F = 15{,}250$ gpm

$\alpha = 200/15{,}250 = 0.01311$

$\lambda = 1/(15.06)(60) = 0.000737\ \text{min}^{-1}$

$t = 30{,}000/15{,}250 = 1.967$

the Na^{24} in the primary coolant is 1.66×10^{5} dis/min·ml. If there is no flow through the demineralizers ($\alpha = 0$), the Na^{24} activity in the primary coolant is 1.67×10^{6} dis/min·ml. Cole[11] performed a similar analysis for the HFIR assuming a 200 gpm clean-up rate and calculated a Na^{24} activity of 1.94×10^{5} dis/min·ml.

These values agree well with those calculated above since both of these methods are quite approximate. The pH of the ORR cooling water is usually between 5.5 and 6.0.

Comparison with Calculation Assuming a Recoil Mechanism

The rate of addition of Na^{24} to the primary coolant from a clean fuel element by recoil mechanism is given by ORNL-963[14] to be,

$$\text{Addition rate} = \frac{(\text{reaction rate in Al})(\text{area})}{4\alpha} = \frac{\left[\Sigma(8.7\ \text{Mev}) \int_{8.7\ \text{Mev}}^{\infty} \phi(E)\,dE\right](\text{area})}{4\alpha}$$

where

$$\alpha = \frac{(\text{atomic weight of } Na^{24})}{(\text{atomic weight of } Al^{27})(\text{recoil range})}$$

Assuming the density of aluminum to be 2.70 g/cc and the recoil range to be 2.4×10^{-4} cm[14], the rate of addition of Na^{24} to the coolant is,

$$= \frac{(1.77 \times 10^{-26})(2.7)(6.025 \times 10^{23})(4.61 \times 10^{12})(5.32 \times 10^{5})}{\dfrac{(27)(4)(23)}{(27)(2.4 \times 10^{-4})}} = 1.84 \times 10^{11} \frac{\text{atoms}}{\text{sec}} .$$

Using the material balance for steady state conditions shown above, this is equivalent to an activity level in the coolant of 1.0×10^{4} dis/min·ml.

This value is somewhat lower than the values calculated by the above methods. This indicates that possibly Na^{24} may rapidly diffuse out of the oxide film after formation, and/or it may be rapidly leached out of the oxide by the cooling water. Another possible mechanism is that formation of the oxide increases the effective, exposed, surface area of the aluminum which permits a greater escape rate of the Na^{24} by the recoil mechanism.

Conclusions

The Na^{24} activity in the HFIR primary coolant water is normally in the range 1.4×10^{5} to 6×10^{5} dis/min·ml.

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