THERMAL NEUTRON ABSORBER BUILDUP IN IRRADIATED BERYLLIUM

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The differential equations which describe the buildup of thermal neutron absorbers in beryllium have been solved. The solutions were used to calculate the change in the macroscopic absorption cross section of the Plum Brook Reactor as a result of the buildup of these poisons. This calculated value agrees within the experimental error of an actual observed change. The effects of variations in the factors on which the poison is dependent were studied and are presented in the form of graphs.
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SUMMARY

The differential equations which describe the buildup of thermal neutron absorbers in beryllium have been solved. The solutions were used to calculate the change in the macroscopic absorption cross section of the Plum Brook Reactor as a result of the buildup of these poisons. This calculated value agrees within the experimental error of an actual observed change. The effects of variations in the factors on which the poison is dependent were studied and are presented in the form of graphs.

INTRODUCTION

When beryllium is exposed to fast neutrons, a chain of reactions takes place which results in the buildup of isotopes with high thermal neutron cross sections. For thermal reactors using beryllium as a reflector or moderator material these thermal neutron poisons can be significant, especially if the reactor must be shut down for several months or more. For example, after being shut down for approximately 100 days the PBR (Plum Brook Reactor) critical rod bank height at restart was about 0.8 inch higher than normal.

The buildup of poison in beryllium has been analyzed before by Morris (ref. 1) for continuous reactor operation. However, his solutions for the equations governing the poison buildup are not general because of the use of boundary conditions for continuous operation.

In this work the differential equations governing the reactions were solved for more general boundary conditions. The change in the core absorption cross section was then calculated for cyclic reactor operation in the PBR. By using perturbation theory the corresponding change in reactivity was calculated. The analytical model was also used to study the effects of operation time, shutdown time, power, and fast to thermal flux ratio on the buildup of thermal neutron poisons in beryllium.
SYMBOLS

$\Delta K/K$  reactivity  
$N_1$  number of $^{9}$Be nuclei per cm$^3$  
$N_2$  number of $^6$Li nuclei per cm$^3$  
$N_3$  number of $^3$He nuclei per cm$^3$  
$N_4$  number of $^3$He nuclei per cm$^3$  
$N_i^0$  value of $N_i$ at beginning of time interval  
$\delta \Sigma_a$  change in macroscopic absorption cross section  
$\lambda$  decay constant for $^3$He, sec$^{-1}$  
$\sigma_1$  cross section for $^{9}$Be$^9(n_f, \alpha)$  
$\sigma_2$  cross section for $^6$Li$^6(n_s, \alpha)$  
$\sigma_4$  cross section for $^3$He$^3(n_s, p)$  
$\phi_f$  fast neutron flux $> 0.82$ MeV, $n/(cm^2)(sec)$  
$\phi_s$  thermal neutron flux, $n/(cm^2)(sec)$  
$\phi_s^*$  adjoint thermal neutron flux

ANALYSIS

Defining Equations

The chain of reactions occurring in beryllium when in the presence of fast and thermal neutrons is as follows:

$$Be^9(n_f, \alpha) \rightarrow He^6(\beta^-)^{0.84S} \rightarrow Li^6(n_s, \alpha)^{910b} \rightarrow H^3 \frac{(\beta^-)12.5Y}{(n_s, p)5400b} He^3$$

Two of the isotopes formed, $^6$Li and $^3$He, have large thermal neutron absorption cross sections. To determine the concentration of these poisons, it was necessary to solve the set of simultaneous differential equations defining the reaction chain. These equations are
The above equations do not consider loss of hydrogen or helium by diffusion; however, for thick sections of beryllium (~1 in.) this is a negligible effect. The $N_1$ burnup is nearly zero, so for this analysis $N_1$ was assumed constant. The above equations were solved by using the Laplace transform and the complex inversion integral. The solutions give $N_2, N_3,$ and $N_4$ as functions of time:

$$
\frac{dN_2}{dt} = \frac{N_1 \varphi S_1}{\varphi S_2} - N_2 \varphi S_2
$$

$$
\frac{dN_3}{dt} = N_2 \varphi S_2 - N_3 \lambda + N_4 \varphi S_4
$$

$$
\frac{dN_4}{dt} = N_3 \lambda - N_4 \varphi S_4
$$

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$$
N_2 = \frac{N_1 \varphi S_1}{\varphi S_2} \left( 1 - e^{-\varphi S_2 t} \right) + N_2^0 e^{-\varphi S_2 t}
$$

$$
N_3 = e^{-(\lambda + \varphi S_4) t} \left[ N_3^0 - \frac{N_3^0 + N_4^0}{\lambda + \varphi S_4} \left( \frac{N_1 \varphi S_1 + N_2 \varphi S_4}{\lambda + \varphi S (\sigma_4 - \sigma_2)} \right) \right] + e^{-\varphi S_2 t} \left[ \frac{N_1 \varphi S_1}{\varphi S_2 (\lambda + \varphi S (\sigma_4 - \sigma_2))} - \frac{N_1 \varphi S_1 + N_2 \varphi S (\sigma_4 - \sigma_2)}{\lambda + \varphi S (\sigma_4 - \sigma_2)} \right] + N_4^0 \varphi S_4 t
$$

$$
N_4 = \frac{N_1 \varphi S_1}{\varphi S_4} \left[ \frac{1}{\varphi S (\lambda + \varphi S_4)} + \frac{1}{\lambda + \varphi S_4} \right]
$$
With \( N_2 \) and \( N_4 \) determined, the increase in the macroscopic absorption cross section in the beryllium is given by

\[
\delta \Sigma_a = N_2 \sigma_2 + N_4 \sigma_4
\]
Application to Plum Brook Reactor

To determine the poison buildup in the PBR the beryllium reflector which surrounds the core on all four sides was broken up into seven regions. This was to account for the variation in flux around the reflector. Average fluxes for the seven regions were obtained from a two-dimensional neutron diffusion calculation. The value of $\sigma_1$ was determined by flux-weighting the cross section over the interval 0.82 MeV to 10 MeV. An average operation time and shutdown time per cycle was determined for the 5 years of operation prior to the 100 day shutdown. This information was input to the poison buildup computer program to solve for the poison atom densities. The reactivity corresponding to the change in absorption cross section was obtained from equation (8). The adjoint fluxes required to solve equation (8) were obtained from the neutron diffusion calculation. Table I gives some of the physical constants used for the PBR.

### Table I. - Values of Constants Used for PBR Calculation

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_1^0$, atoms/cm$^3$</td>
<td>$1.12 \times 10^{23}$</td>
</tr>
<tr>
<td>$N_2^0$, atoms/cm$^3$</td>
<td>0</td>
</tr>
<tr>
<td>$N_3^0$, atoms/cm$^3$</td>
<td>0</td>
</tr>
<tr>
<td>$N_4^0$, atoms/cm$^3$</td>
<td>0</td>
</tr>
<tr>
<td>$\sigma_1$, b</td>
<td>0.0214</td>
</tr>
<tr>
<td>$\sigma_2$, b</td>
<td>950</td>
</tr>
<tr>
<td>$\sigma_3$, b</td>
<td>5400</td>
</tr>
<tr>
<td>$\lambda$, sec$^{-1}$</td>
<td>$1.76 \times 10^{-9}$</td>
</tr>
<tr>
<td>$\phi_s$, N/(cm$^2$)(sec)</td>
<td>$\sim 5 \times 10^{14}$</td>
</tr>
<tr>
<td>$\phi_{s2}$ (&gt;0.82 MeV), N/(cm$^2$)(sec)</td>
<td>$\sim 1 \times 10^{14}$</td>
</tr>
<tr>
<td>$t_{on}$, days</td>
<td>9.45</td>
</tr>
<tr>
<td>$t_{off}$, days</td>
<td>14.95</td>
</tr>
</tbody>
</table>

### RESULTS AND DISCUSSION

Comparison of Measured and Calculated Reactivity Change

When the PBR was restarted following a 100-day shutdown, the critical rod bank height (RBH) was about 0.8 inch higher than expected. In terms of reactivity this is about $1.35$. The reactivity associated with the buildup of poisons in the beryllium reflector was calculated to be worth $1.21$. The estimated probable error in the predicted
or expected reactivity (RBH), and hence in the measured reactivity change, is ±30 percent. The estimated probable error in the calculated reactivity change is believed to be about 20 percent.

Effects of Variations in the Independent Variables

Since there was a noticeable effect as a result of a 100-day shutdown, the question arose as to how serious the effect might be for longer shutdown periods. Figures 1 and 2 show the poison buildup as a function of shutdown time following 5 years of cyclic operation. For this period, which was the first 5 years of operation for the PBR, the average on time was about 10 days and the average down time was about 15 days. (Since that time this average has changed to about 20 days on and 5 days off.)

![Graph showing poison buildup as a function of shutdown time](image)
The present PBR core and fuel loading are such that the maximum excess reactivity in the control rods is about $15.00. Thus from figure 2 it is seen that had the PBR been shut down for a 3 year period, it could not have been restarted without major core changes.

Figure 3 shows the effect of operation time for 20-day cycles. The longer the on time per cycle, the more quickly the poison level rises. However, after about 1 year of operation the poison level for 5 days operation out of 20 and 15 days operation out of 20 is about the same.

Figure 4 shows the poison buildup against operation time for several flux or power levels. (Figs. 4 to 6 are for continuous operation; however, the effects would be similar for cyclic operation.) The higher the reactor power, the steeper the rate of poison....
Figure 4. - Poison buildup as function of operation time for neutron flux ratio $\varphi_I/\varphi_s = 1$.

Figure 5. - Poison buildup as function of operation time with fast neutron flux $\varphi_I$ constant and thermal neutron flux $\varphi_s$ a parameter.
buildup. However, after several years the poison level is independent of the power. After 10 years of operation the poison levels for the three different powers are within 3 percent of each other.

To examine the effect of the fast-to-thermal flux ratio, several combinations of the fast and thermal flux were calculated. The results are plotted in figures 5 and 6. Figure 5 shows that, for a given fast flux, changing the thermal flux causes a change in the poison level and the rate of poison buildup. Figure 6 shows that, for a given thermal flux, increasing the fast flux by a factor of 10 causes an increase in poison by a factor of 10.

CONCLUDING REMARKS

Following a long shutdown of the PBR there was an unexpected change in the core reactivity. This change can be accounted for by the buildup of Li$^6$ and He$^3$ in the beryllium reflector.
Although good agreement was obtained between the nominal measured and calculated reactivity change for the PBR, there was considerable uncertainty in both the measured value (because of uncertainty in the predicted critical rod position) and the calculated value. Chemical analyses are planned which will determine the atom densities of the reaction products in beryllium from the PBR core. This will give a second and more direct experimental check on the calculation of poison buildup in beryllium.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, April 20, 1970,
120-27.
APPENDIX - IBM-1620 COMPUTER CODE FOR POISON BUILDUP IN BERYLLIUM FOR CYCLIC REACTOR OPERATION

C POISON BUILDUP IN BERYLLIUM FOR CYCLIC REACTOR OPERATION
PRINT 100
100 FORMAT(57HPOISON BUILDUP IN BERYLLIUM FOR CYCLIC REACTOR OPERATIO
1N,/) READ 200,AN1,AN2,AN3,AN4,PHI1,PHI4,NTS,TON,TOFF
200 FORMAT(4E9.4,2E9.2,14,2F6.0)
T=TON*86400.*
DK=1.76E-9
SIG1=.0214E-24
SIG2=950.E-24
SIG4=5400.E-24
PRINT 101,AN1
PRINT 102,AN2
PRINT 103,AN3
PRINT 104,AN4
PRINT 105,PHI1
PRINT 106,PHI4
101 FORMAT(23H INITIAL BE NO. DEN. IS,E10.2)
102 FORMAT(23H INITIAL LI NO. DEN. IS,E10.2)
103 FORMAT(23H INITIAL TR NO. DEN. IS,E10.2)
104 FORMAT(23H INITIAL HE NO. DEN. IS,E10.2/)
105 FORMAT(27H FAST (GRP. 1) FLUX USED IS,E10.2/)
106 FORMAT(27H SLOW (GRP. 4) FLUX USED IS,E10.2/)
PRINT 117,TON
PRINT 118,TOFF
117 FORMAT (30H IRRADIATION TIME PER CYCLE IS,F6.2,5H DAYS)
118 FORMAT (27H SHUTDOWN TIME PER CYCLE IS,F9.2,5H DAYS/)
PHIF=PHI1
PHIS=PHI4
I=0
M=0
1=I+1
A=AN1*PHIF*SIG1
B=PHIS*SIG2
C=PHIS*SIG4
D=DK+C
E=D-B
EB=EXP(-B*T)
ED=EXP(-D*T)
BN2=A/B*(1.-EB)+AN2*EB
BN3=D*(AN3-(AN3+AN4)*C/D+(A+AN2*C)*B/E/D-AN2*B/E-A*B*C/D/D/E)+EB*
1*(AN2*B/E-(A+AN2*C)/E+A*C/B/E)+A*C/D*T+(AN3+AN4)*C/D*(A+AN2*C)/D-A*
2C*(1./B/D+1./D/D)
BN4=E*(AN4+C/D+(AN2+AN3)*B*DK/D/E-AN3*DK/E-A*B*DK/D/D/E)+EB*(A/B-
1AN2)*DK/E+A*DK/D*T+(AN4+AN3+AN2-A*(1./B+1./D))*DK/D
AN2=BN2
AN3=BN3
AN4=BN4
PN2=AN2*SIG2
PN4=AN4*SIG4
DELSIG=PN2+PN4
PRINT 108,AN1
PRINT 109,AN2
PRINT 110,AN3
PRINT 111,AN4
PRINT 112,PN2
PRINT 113,PN4
PRINT 114,DELSIG
PRINT 115,1
108 FORMAT (18H BE NU. DENSITY IS,E10.2)
109 FORMAT (18H LI NU. DENSITY IS,E10.2)
110 FORMAT (18H TR NU. DENSITY IS,E10.2)
111 FORMAT (18H HE NU. DENSITY IS,E10.2)
112 FORMAT (25H DELTA SIGMA DUE TO LI IS,E10.2)
113 FORMAT (25H DELTA SIGMA DUE TO HE IS,E10.2)
114 FORMAT (25H THE TOTAL DELTA SIGMA IS,E10.2)
115 FORMAT (44H THE ABOVE VALUES ARE AFTER TIME STEP NUMBER,14,//)
   IF(M)2,2,3
   M=1
   PHIF=1.E-20
   PHIS=1.E-20
   T=TOFF*86400.
   IF(I-NTS)1,4,4
3   M=0
   PHIF=PHI1
   PHIS=PHI4
   T=TON*86400.
   IF(I-NTS)1,4,4
4   PRINT 116
   116 FORMAT (19H END OF CALCULATION)
   GO TO 5
END
REFERENCES


"The aeronautical and space activities of the United States shall be conducted so as to contribute . . . to the expansion of human knowledge of phenomena in the atmosphere and space. The Administration shall provide for the widest practicable and appropriate dissemination of information concerning its activities and the results thereof."

— National Aeronautics and Space Act of 1958

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