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SUBJECT: Time Behavior of Fuel Concentrations in Single-Region
Reactors Containing U-233, U-235, Th-232 and Fission
Product Poisons

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FROM: Nathan Gilbert and Paul R. Kasten

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Abstract

Analytical expressions were obtained for the time behavior of fuel concentrations and fuel-feed rates in single-region, spherical, $\text{UO}_3\text{-ThO}_2\text{-D}_2\text{O}$ reactors.

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ThO₂-UO₃-D₂O Reactors

The following equations were used to express the conditions of criticality and material balance in an aqueous homogeneous, single-region reactor, initially fueled with U-235 and using Th-232 as a fertile material to produce U-233:

$$\frac{dN(25)}{dt} = F(25) - \sigma'(25) N(25) \phi \quad (1)$$

$$\begin{aligned} \frac{dN(13)}{dt} = & \sigma(02) N(02) \phi + \frac{1 - p(02)}{1 + B^2 \tau} \left[\eta(25) \sigma(25) N(25) \right. \\ & \left. + \eta(23) \sigma(23) N(23) \right] \phi - \sigma'(13) N(13) \phi \end{aligned} \quad (2)$$

$$\frac{dN(23)}{dt} = \lambda(13) N(13) + F(23) - \sigma'(23) N(23) \phi \quad (3)$$

$$\bar{\eta} \in pf = (1 + B^2 \tau) (1 + B^2 L^2) \quad (4)$$

$$\Sigma(p) = \Sigma(p_1) + \Sigma(p_2) \quad (5)$$

$$\frac{dN(p_1)}{dt} = c_1 \left[\sigma_f(25) N(25) + \sigma_f(23) N(23) \right] \phi - \sigma'(p_1) N(p_1) \phi \quad (6)$$

$$\frac{dN(p_2)}{dt} = c_2 \left[\sigma_f(25) N(25) + \sigma_f(23) N(23) \right] \phi - \sigma'(p_2) N(p_2) \phi \quad (7)$$

The primed values of absorption cross-sections include the effect of removal of fuel solution for processing and, where appropriate, the radioactive decay of Pa-233:

$$\begin{aligned}
 \sigma'(25) &= \sigma(25) + \frac{k_1}{T_2 \phi} \\
 \sigma'(13) &= \sigma(13) + \frac{\lambda(13)}{\phi} + \frac{k_2}{T_2 \phi} \\
 \sigma'(23) &= \sigma(23) + \frac{k_3}{T_2 \phi} \\
 \sigma'(p_1) &= \sigma(p_1) + \frac{k_4}{T_2 \phi} \\
 \sigma'(p_2) &= \sigma(p_2) + \frac{k_5}{T_2 \phi}
 \end{aligned} \tag{8}$$

where T_2 is the effective fuel removal cycle time and k_i 's are the fractions of respective atoms removed in passing through the fuel-processing plant.

Although ϕ will vary with time, it will be considered constant here insofar as the $\sigma'(i)$'s are concerned. This will be adequate for most cases.

Equations 1 - 7 can be rewritten in terms of a new variable

$$T \equiv \int_0^t \phi \, dt \text{ (after manipulation of the critical equation):}$$

$$\frac{dN(25)}{dT} = \frac{F(25)}{\phi} - \sigma'(25) N(25) \tag{9}$$

$$\frac{dN(23)}{dT} = \frac{\lambda(13)}{\phi} N(13) + \frac{F(23)}{\phi} - \sigma'(23) N(23) \tag{10}$$

$$\begin{aligned}
 \frac{dN(13)}{dT} &= \sigma(02) N(02) + \frac{1 - p(02)}{1 + B^2 \tau} [\eta(25) \sigma(25) N(25) + \eta(23) \sigma(23) N(23)] \\
 &\quad - \sigma'(13) N(13)
 \end{aligned} \tag{11}$$

$$\frac{dN(p_1)}{dT} = c_1 [\sigma_f(23) N(23) + \sigma_f(25) N(25)] - \sigma'(p_1) N(p_1) \tag{12}$$

$$\frac{dN(p_2)}{dT} = c_2 [\sigma_f(23) N(23) + \sigma_f(25) N(25)] - \sigma'(p_2) N(p_2) \tag{13}$$

$$\beta(25) N(25) + \beta(23) N(23) = m + \sigma(p_1) N(p_1) + \sigma(p_2) N(p_2) \quad (14)$$

$$\text{where } \beta(25) = \left[\frac{\epsilon p(02)}{1 + B^2 \tau} \eta(25) - 1 - (fp)_0 \right] \sigma(25)$$

$$\beta(23) = \left[\frac{\epsilon p(02)}{1 + B^2 \tau} \eta(23) - 1 - (fp)_0 \right] \sigma(23)$$

$$m = \Sigma(02) + \Sigma(\text{Mod}) + B^2 D.$$

Note that $\Sigma(13)$ is omitted from the formulation of m ; its contribution to m is small; it may be included in the $(fp)_0$ term.

If $F(25) = 0$, equation (9) becomes

$$\frac{dN(25)}{dT} = -\sigma'(25) N(25), \text{ which can be solved directly to give}$$

$$N(25) = N_0(25) e^{-\sigma'(25) T} \quad (15)$$

$N(23)$ is eliminated from equations (11), (12), and (13) by substituting its value from the critical equation (14) and using equation (15):

$$N(23) = \frac{1}{\beta(23)} \left[m + \sigma(p_1) N(p_1) + \sigma(p_2) N(p_2) \right] - \frac{\beta(25)}{\beta(23)} N_0(25) e^{-\sigma'(25) T}$$

The following set of equations is obtained upon collection of terms and introduction of appropriate symbols (see Nomenclature) for groups of constants:

$$\frac{dN(13)}{dT} + x_1 N(13) = x_2 + x_3 e^{-\sigma'(25) T} + x_4 N(p_1) + x_5 N(p_2) \quad (16)$$

$$\frac{dN(p_1)}{dT} + y_1 N(p_1) = y_2 + y_3 e^{-\sigma'(25) T} + y_4 N(p_2) \quad (17)$$

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$$\frac{dN(p_2)}{dt} + z_1 N(p_2) = z_2 + z_3 e^{-\sigma'(25)T} + z_4 N(p_1) \quad (18)$$

The set of simultaneous differential equations (16) - (18) in $N(13)$, $N(p_1)$ and $N(p_2)$ is solved by using the Laplace transforms; the corresponding equations (19) - (21) are obtained:

$$[s \text{ is the transform parameter; } n = \mathcal{L}\{N\}]$$

$$(s + x_1) n(13) - x_4 n(p_1) - x_5 n(p_2) = N_0(13) + \frac{x_2}{s} + \frac{x_3}{s + \sigma'(25)} \quad (19)$$

$$(s + y_1) n(p_1) - y_4 n(p_2) = N_0(p_1) + \frac{y_2}{s} + \frac{y_3}{s + \sigma'(25)} \quad (20)$$

$$- z_4 n(p_1) + (s + z_1) n(p_2) = N_0(p_2) + \frac{z_2}{s} + \frac{z_3}{s + \sigma'(25)} \quad (21)$$

and the solutions will be given by:

$$\left. \begin{aligned} N(13) &= \mathcal{L}^{-1} \left\{ \frac{D(13)}{D_1} \right\} \\ N(p_1) &= \mathcal{L}^{-1} \left\{ \frac{D(p_1)}{D_1} \right\} \\ N(p_2) &= \mathcal{L}^{-1} \left\{ \frac{D(p_2)}{D_1} \right\} \end{aligned} \right\} \quad (22)$$

where

$$D_1 = \begin{vmatrix} s + x_1 & -x_4 & -x_5 \\ 0 & s + y_1 & -y_4 \\ 0 & -z_4 & s + z_1 \end{vmatrix} = (s - r_2)(s - r_3)(s - r_4)$$

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where r_2, r_3 , and r_4 are roots of

$$(s + x_1) \left[(s + y_1) (s + z_1) - z_4 y_4 \right] = 0 \quad (22a)$$

and $D(i)$ is a characteristic determinant. The determinant $D(13)$ can be shown to be equal to

$$D(13) = \frac{A_4 s^4 + A_3 s^3 + A_2 s^2 + A_1 s + A_0}{(s) [s + \sigma'(25)]}$$

so that

$$N(13) = \frac{A_4 s^4 + A_3 s^3 + A_2 s^2 + A_1 s + A_0}{s [s + \sigma'(25)] (s - r_2)(s - r_3)(s - r_4)}$$

and the desired function $N(13)$ will be given by

$$N(13) = \sum_{n=0}^4 H_n e^{r_n T} \quad (23)$$

$$\text{where } H_0 = \frac{A_0}{r_1 r_2 r_3 r_4}$$

$$H_1 = \frac{\sum_{i=0}^4 A_i (r_1)^i}{r_1 (r_1 - r_2)(r_1 - r_3)(r_1 - r_4)}$$

$$H_2 = \frac{\sum_{i=0}^4 A_i (r_2)^i}{r_2 (r_2 - r_1)(r_2 - r_3)(r_2 - r_4)}$$

$$H_3 = \frac{\sum_{i=0}^4 A_i (r_3)^i}{r_3 (r_3 - r_1)(r_3 - r_2)(r_3 - r_4)}$$

$$H_4 = \frac{\sum_{i=0}^4 A_i (r_4)^i}{r_4 (r_4 - r_1)(r_4 - r_2)(r_4 - r_3)}$$

(24)

and

$$\begin{aligned} r_0 &= 0 \\ r_1 &= -\sigma'(25) \end{aligned} \quad (25)$$

Functions $N(p_1)$ and $N(p_2)$ can be obtained in a similar manner, if required.

$N(23)$ is obtained by substitution of $N(13)$, and $\frac{dN(13)}{dT}$ into equation (11), namely:

$$\begin{aligned} N(23) &= \frac{1}{a_1 \eta(23) \sigma(23)} \left\{ -\Sigma(02) - a_1 \eta(25) \sigma(25) N_0(25) e^{r_1 T} \right. \\ &\quad \left. + \sum_{n=0}^4 \left[\sigma'(13) + r_n \right] H_n e^{r_n T} \right\} \end{aligned} \quad (26)$$

An expression for $\frac{F(23)}{\phi}$ is obtained by substituting appropriate expressions for $\frac{dN(23)}{dT}$, $N(13)$ and $N(23)$ from equations (24) and (26) into equation (10). Solving for $\frac{F(23)}{\phi}$ and combining terms, the following equation is obtained:

$$\begin{aligned} \frac{F(23)}{\phi} &= - \frac{\sigma'(23) \Sigma(02)}{a_1 \eta(23) \sigma(23)} - \frac{\eta(25) \sigma(25) [\sigma'(23) + r_1]}{\eta(23) \sigma(23)} N_0(25) e^{r_1 T} \\ &\quad + \sum_{n=0}^4 I_n e^{r_n T} \end{aligned} \quad (27)$$

where

$$I_n = \left\{ \frac{[\sigma'(13) + r_n] [\sigma'(23) + r_n]}{a_1 \eta(23) \sigma(23)} - \frac{\lambda(13)}{\phi_0} \right\} H_n$$

The cumulative feed requirements of U-233 for period of time, t ,

$$\int_0^t F(23) dt = \int_0^T \frac{F(23)}{\phi} dT \quad (28)$$

is obtained by substituting $\frac{F(23)}{\phi}$ given by equation (27) into (28) and performing the indicated integration:

$$F(23)_{\text{cumulative}} = \left\{ \frac{\sigma'(23) [\sigma'(13) H_0 - \Sigma(02)]}{a_1 \eta(23) \sigma(23)} - \frac{\lambda(13)}{\phi_0} H_0 \right\} T - \frac{\eta(25) \sigma(25) [\sigma'(23) + r_1]}{\eta(23) \sigma(23)} N_0(25) \frac{(e^{r_1 T} - 1)}{r_1} + \sum_{n=1}^4 J_n (e^{r_n T} - 1) \quad (29)$$

$$\text{where } J_n = \left\{ \frac{[\sigma'(13) + r_n] [\sigma'(23) + r_n]}{a_1 \eta(23) \sigma(23)} - \frac{\lambda(13)}{\phi_0} \right\} \frac{H_n}{r_n}$$

To obtain a relationship between t and T , the defining equation, $dT = \phi dt$, is used in combination with the assumption of constant power generation. Thus,

$$\begin{aligned} \Sigma_f \phi &= K \text{ and } \frac{1}{\phi} = \frac{1}{K} \Sigma_f = \frac{1}{K} [\sigma_f(25) N(25) + \sigma_f(23) N(23)] \\ dt &= \frac{1}{\phi} dT \\ &= \frac{1}{K} [\sigma_f(25) N(25) + \sigma_f(23) N(23)] dT \end{aligned} \quad (29a)$$

where K is a constant related to the reactor power.

It follows from Eq. (29a) that

$$t = \int_0^t dt = \int_0^T \frac{1}{K} [\sigma_f(25) N(25) + \sigma_f(23) N(23)] dT \quad (29b)$$

Substituting for $N(25)$ and $N(23)$ the appropriate expressions, equations (15) and (26), respectively, and performing the indicated integration, the following relationship between t and T is obtained:

$$\begin{aligned} t = & \frac{\sigma_f(23) [\sigma'(13) H_0 - \Sigma(02)] T}{a_1 \eta(23) \sigma(23) K} + \\ & + \left[\sigma_f(25) - \frac{\eta(25) \sigma(25) \sigma_f(23)}{\eta(23) \sigma(23)} \right] \frac{N_0(25)}{K r_1} \left(e^{r_1 T} - 1 \right) \\ & + \sum_{n=1}^4 \left[\frac{\sigma'(13) + r_n}{r_n} \right] H_n \left(e^{r_n T} - 1 \right) \left[\frac{\sigma_f(23)}{a_1 \eta(23) \sigma(23) K} \right] \end{aligned} \quad (30)$$

Equation (30) permits a computation of t for any desired T , or vice versa; this equation combined with the expression for $F(23)$ permits calculation of the fuel feed requirements of the system for a specified time.

The poison concentrations in the system, $N(p_1)$ and $N(p_2)$, can be obtained by substituting the expressions for $N(25)$ and $N(23)$, into equations (12) and (13) and solving the resultant differential equations.

If $F(23)$ is set equal to zero, Equations 9-14 can be solved in a manner similar to the above; however, under this condition a fourth-order algebraic equation has to be solved in order to obtain characteristic

roots. If $\lambda(13)$ is assumed to be infinite (equivalent to instantaneous decay of $\text{Pa}^{233} \rightarrow \text{U}^{233}$) and $F(23)$ is again set equal to zero, the resulting mathematical system is equivalent to one solved previously for a $\text{U}^{235} - \text{U}^{238} - \text{Pu}^{239}$ system.*

*P. R. Kasten and R. E. Aven, "Fuel Costs in Batch- and Continually Processed Homogeneous Reactors," CF memo to be issued.

CF-57-2-36 ←

NOMENCLATURE

- $a_1 = \frac{1 - p(02)}{1 + B^2 \tau}$
- $A_0 = x_{11} w_2 + y_{11} w_3 + z_{11} w_4$
- $A_1 = x_{10} w_2 + x_{11} w_1 + y_{10} w_3 + y_{11} w_4 + z_{10} w_4 + x_5 z_{11}$
- $A_2 = w_2 N_0(13) + x_{10} w_1 + x_{11} w_3 + w_3 N_0(p_1) + x_4 y_{10} + w_4 N_0(p_2) + x_5 z_{10}$
- $A_3 = w_1 N_0(13) + x_{10} w_1 + x_4 N_0(p_1) + x_5 N_0(p_2)$
- $A_4 = N_0(13)$
- $B^2 = \text{buckling of reactor; } B^2 = \left[\frac{\pi}{R} \right]^2 \text{ where } R \text{ is the physical reactor radius plus an effective extrapolation distance; cm}^{-2}$
- $B_L^2 = \text{ratio of thermal neutron leakage to absorption; dimensionless}$
- $B^2 \tau = \text{ratio of fast neutron leakage to moderation; dimensionless}$
- $C_1 = \text{fraction of atoms produced having effective cross section } \sigma(p_1) \text{ per fission}$
- $C_2 = \text{fraction of atoms produced having effective cross section } \sigma(p_2) \text{ per fission}$
- $D = \text{thermal diffusion coefficient for reactor core, cm}$
- $D_1 = (s - r_2)(s - r_3)(s - r_4)$
- $D(13) = \frac{A_0 + A_1 s + A_2 s^2 + A_3 s^3 + A_4 s^4}{s [s + \sigma'(25)]}$
- $f = \text{thermal utilization, } \frac{\Sigma(\text{fuel})}{\Sigma_T}, \text{ dimensionless}$
- $(fp)_0 = \text{fraction poisons due to high cross section poisons;}$

F = feed rate of element to reactor system, $\frac{\text{atoms}}{\text{barn} - \text{cm} - \text{sec}}$

$$F(25) = U^{235} \text{ feed rate; } F(25)_{\text{cumulative}} = \int_0^t F(25) dt$$

$$F(23) = U^{233} \text{ feed rate } F(23)_{\text{cumulative}} = \int_0^t F(23) dt$$

$$H_0 = \frac{A_0}{r_1 r_2 r_3 r_4}$$

$$H_1 = \frac{\sum_{i=0}^4 A_i r_1^i}{(r_1)(r_1 - r_2)(r_1 - r_3)(r_1 - r_4)}$$

$$H_2 = \frac{\sum_{i=0}^4 A_i r_2^i}{(r_2)(r_2 - r_1)(r_2 - r_3)(r_2 - r_4)}$$

$$H_3 = \frac{\sum_{i=0}^4 A_i r_3^i}{(r_3)(r_3 - r_1)(r_3 - r_2)(r_3 - r_4)}$$

$$H_4 = \frac{\sum_{i=0}^4 A_i r_4^i}{(r_4)(r_4 - r_1)(r_4 - r_2)(r_4 - r_3)}$$

$$J_n = \left\{ \frac{[\sigma'(13) + r_n][\sigma'(23) + r_n]}{a_1 \eta(23) \sigma(23)} - \frac{\lambda(13)}{\phi_0} \right\} \frac{H_n}{r_n}$$

k_1, k_2, k_3, k_4, k_5 = fractions of respective atoms of (25), (13), (23), (p₁) and (p₂) removed from the system in passing through the fuel processing plant.

$K = \Sigma_f \phi$ at constant power density

$m = \Sigma(02) + \Sigma(\text{mod.}) + B^2 D$

$n = \mathcal{L}\{N\}$

$N =$ concentration of particular element; $\frac{\text{atoms}}{\text{barn-cm}}$

$N(02)$ refers to Th^{232} concentration

$N(13)$ refers to Pa^{233} concentration

$N(23)$ refers to U^{233} concentration

$N(25)$ refers to U^{235} concentration

$N(p_1)$ refers to concentration of poisons having an effective cross section of $\sigma(p_1)$

$N(p_2)$ refers to concentration of poisons having an effective cross section of $\sigma(p_2)$

$N_0 =$ concentration of particular element at zero time; atoms/barn - cm

$p(02) = p =$ resonance escape probability as determined by Th^{232} concentration

$r_0 = 0$

$r_1 = -\sigma'(25)$

$r_2 =$
 $r_3 =$
 $r_4 =$

} characteristic roots defined by Eq. 22a

$s =$ transform parameter

$t =$ time, sec or yrs.

$T = \int_0^t \phi dt$

$T_2 =$ fuel processing cycle time, days

$$w_1 = y_1 + z_1$$

$$w_2 = y_1 z_1 - y_4 z_4$$

$$w_3 = x_4 z_1 + x_5 z_4$$

$$w_4 = x_4 y_4 + x_5 y_1$$

$$x_1 = \sigma'(13)$$

$$x_2 = \frac{a_1 \eta(23) \sigma(23)}{\beta(23)} m + \Sigma(02)$$

$$x_3 = a_1 \left[\eta(25) \sigma(25) - \frac{\eta(23) \sigma(23)}{\beta(23)} \beta(25) \right] N_o(25)$$

$$x_4 = \frac{a_1 \eta(23) \sigma(23)}{\beta(23)} \sigma(p_1)$$

$$x_5 = \frac{a_1 \eta(23) \sigma(23)}{\beta(23)} \sigma(p_2) = \frac{\sigma(p_2)}{\sigma(p_1)} x_4$$

$$x_{10} = \sigma'(25) N_o(13) + x_2 + x_3$$

$$x_{11} = x_2 \sigma'(25)$$

$$y_1 = \sigma'(p_1) - \frac{c_1 \sigma_f(23)}{\beta(23)} \sigma(p_1)$$

$$y_2 = \frac{c_1 \sigma_f(23)}{\beta(23)} m$$

$$y_3 = c_1 \left[\sigma_f(25) - \frac{\sigma_f(23)}{\beta(23)} \beta(25) \right] N_o(25)$$

$$y_4 = \frac{c_1 \sigma_f(23)}{\beta(23)} \sigma(p_2)$$

$$y_{10} = \sigma'(25) N_o(p_1) + y_2 + y_3$$

$$y_{11} = y_2 \sigma'(25)$$

$$z_1 = \sigma'(p_2) - \frac{c_2 \sigma_f(23)}{\beta(23)} \sigma(p_2)$$

$$z_2 = \frac{c_2 \sigma_f(23)}{\beta(23)} m$$

$$z_3 = c_2 \left[\sigma_f(25) - \frac{\sigma_f(23) \beta(25)}{\beta(23)} \right] N_o(25) = \frac{c_2}{c_1} y_3$$

$$z_4 = \frac{c_2 \sigma_f(23)}{\beta(23)} \sigma(p_1)$$

$$z_{10} = \sigma'(25) N_o(p_2) + z_2 + z_3$$

$$z_{11} = z_2 \sigma'(25)$$

$$\beta(23) = \left[\frac{\epsilon p(02)}{1 + B^2 \tau} \eta(23) - 1 - (fp)_o \right] \sigma(23)$$

$$\beta(25) = \left[\frac{\epsilon p(02)}{1 + B^2 \tau} \eta(25) - 1 - (fp)_o \right] \sigma(25)$$

$$\epsilon = \text{fast effect; taken to be 1}$$

$$\bar{\eta} = \text{average number of fast neutrons produced per thermal neutron absorbed in fuel}$$

$$\lambda(13) = \text{decay constant of Pa}^{233} = \frac{0.693}{\text{half-life}} ; \text{sec}^{-1}$$

σ = microscopic absorption cross section, barns

$\sigma(02)$ refers to absorption cross section of Th^{232}

$\sigma(13)$ refers to absorption cross section of Pa^{233}

$\sigma(23)$ refers to absorption cross section of U^{233}

$\sigma(25)$ refers to absorption cross section of U^{235}

$\sigma(p_1)$ refers to absorption cross section of effective fission product poison p_1

σ_f = microscopic fission cross section of fuel; barns

σ' = effective removal cross section

$$\sigma'(25) = \sigma(25) + \frac{k_1}{T_2 \phi_0}$$

$$\sigma'(13) = \sigma(13) + \frac{k_2}{T_2 \phi_0} + \frac{\lambda(13)}{\phi_0}$$

$$\sigma'(23) = \sigma(23) + \frac{k_3}{T_2 \phi_0}$$

$$\sigma'(p_1) = \sigma(p_1) + \frac{k_4}{T_2 \phi_0}$$

$$\sigma'(p_2) = \sigma(p_2) + \frac{k_5}{T_2 \phi_0}$$

Σ = macroscopic absorption cross section; cm^{-1}

Σ_f = macroscopic fission cross section; cm^{-1}

$$\Sigma_f(23) = \sigma_f(23) N(23)$$

$$\Sigma_f(25) = \sigma_f(25) N(25)$$

$$\Sigma_f = \Sigma_f(23) + \Sigma_f(25)$$

$\Sigma(\text{mod})$ = macroscopic absorption cross section of moderator; cm^{-1}

Σ_{T} = total macroscopic absorption cross section; cm^{-1}

τ = Fermi Age; cm^2

ϕ = neutron flux; neutrons/barn-sec.

ϕ_0 = average value of ϕ