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UCCPOST OFFICE BOX P
OAK RIDGE, TENNESSEE **ORNL**
CENTRAL FILES NUMBERCF-57-6-51

External Transmittal Authorized

COPY NO. 153

DATE: June 13, 1957

SUBJECT: Fuel Exposures in Heterogeneous Thorium
Breeder Reactors

TO: Distribution

FROM: B. E. Prince and S. Jaye

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Continued on page 2

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INTRODUCTION

In some countries large deposits of fertile material are available, but fissile material must be imported from some other country. If the nuclear energy capabilities of these countries are to be independent of outside sources of fuel (after a first loading), a reactor must be built which can produce power and maintain at least a constant inventory of fissile material. Further, economic power from heterogeneous-type reactors depends to a large extent upon achieving large fuel exposures in order to minimize the cost of fabrication and chemical processing.

This report summarizes some preliminary calculations of fuel exposures attainable in heterogeneous reactors, fueled with a mixture of Thorium and U^{233} , moderated with D_2O and operated with no net loss in fissionable fuel.

SUMMARY

The results of this survey indicate that heterogeneous Thorium-U²³³ systems can, under specialized assumptions, achieve exposures of 10,000 MWD/ton and above with retainment of the initial fuel inventory. These calculations are based on a single-region spherical reactor, containing U²³³-Thorium rods on an 8-in. lattice pitch and moderated with D₂O. The assumptions are made of operation with no structural poisons, no Pa²³³ losses, uniform fuel exposure, and control by fuel concentration. Graphs of the initial breeding ratio (IBR) and the fuel exposure as functions of Thorium concentration are presented in Fig. (1) and (2), respectively. Size was treated as a parameter, and the calculations were performed for reactor diameters of 12 ft and 14 ft. For diameters less than about 12 ft and practical Thorium concentrations, the IBR drops below unity. For a given diameter, the IBR and exposure are expected to be between the curves labeled $x = 0.03$ and 0.05 , which are the assumed limits of fraction of "constant" fission product poisons. It is concluded that, even with the special assumptions made, the results appear to warrant further investigation of these systems. This would include improvement of the calculation in the resonance region and extension of the parameter study.

Fig. 1 Initial Breeding Ratio Vs. Thorium Concentration

Fig. 2 Fuel Exposure Vs. Thorium Concentration

DISCUSSION

Fuel exposures of 5,000 to 10,000 MWD/ton can be set as a reasonable goal in economical fuel cycles. Thus, the heterogeneous Thorium-U²³³-D₂O reactor appears promising as an economical power reactor operated with no net breeding loss.

It should be pointed out that, in practice, the lifetime of the fuel elements before reprocessing will also be dependent on the radiation damage to the element. It has generally been considered that this damage forms the present day limitation in achieving large exposures. However, in recent experiments at Chalk River, fuel elements of thorium oxide clad with zirconium and cooled with water have been irradiated to 10,000 MWD/ton with substantially no damage to the element.⁽¹⁾

The physical assumptions and mathematical analysis used are discussed in the following section. It is expected that the analysis is inadequate in the following respects:

- (a) Nonuniformity of fuel burnup and poison formation has not been accounted for. The actual exposure of a section of a fuel rod will vary with position within the reactor and will also be a function of any rod movement during its irradiation history. This type of correction may reduce the above estimates of exposure.
- (b) Neglect of poisoning by structural material makes the above results optimistic. This was omitted to make the calculations independent of particular reactor types.
- (c) Resonance absorption in U²³³, which was neglected in this calculation, should tend to improve the breeding ratio, provided η is substantially independent of neutron energy. The effect is to bypass the thermal region, reducing thermal leakage and parasitic capture. This energy independence of η for U²³³ has been reported in the literature;⁽²⁾ however, there is evidence that $\eta_{\text{res}} < \eta_{\text{th}}$, under which circumstance the breeding ratio would decrease.⁽³⁾

PROCEDURE

Calculations were based upon a single-region spherical reactor containing U^{233} -Th- D_2O at $250^\circ C$. The reactor was assumed to operate at constant power density and to be maintained critical by controlling fuel concentration. The basic working equations represent (1) the condition of criticality; (2) buildup of stable fission products during operation; and, (3) definition of the breeding ratio for this system:*

$$(1) \left(\frac{\eta^{23} \epsilon_p}{1 + B^2 \tau} - 1 - x \right) \Sigma^{23}(t) = \Sigma^P(t) + \Sigma^{O2} + \Sigma^{Mod} (1 + L^2 B^2)$$

$$\left(B^2 = \left(\frac{\pi}{R} \right)^2 \right)$$

$$(2) \frac{dN^P}{dt} = \Sigma_f^{23}(t) \phi(t) - \Sigma^P(t) \phi(t) \quad \left(\Sigma^P = N^P \sigma_p \right)$$

$$(3) B.R. = \Sigma^{O2} / \Sigma^{23}(t) + \frac{\eta^{23} \epsilon (1 - p)}{1 + B^2 \tau}$$

In the above equations the assumptions are made that the neutron flux is spatially uniform and that depletion of Thorium and D_2O atoms is negligible. The former assumption means, of course, that fuel burnout and poison formation will be independent of position within the reactor.

Fuel exposure times (t_o) were determined from the criterion that the total U^{233} burnup during a cycle be equal to the amount bred from Thorium, i.e.:

$$(4) \int_0^{t_o} (B.R. - 1) dt = 0$$

* See appendix for definitions of symbols.

Solution of equations (1) through (4) for the fuel exposure is outlined in the Appendix of this report. The following simple formulas express the initial breeding ratio (IBR) and the fuel exposure, the latter written in terms of the integrated specific power (MWD/ton);

$$(5) \quad \text{IBR} = F k_1 + k_2$$

$$(6) \quad \text{Exposure} \left[\text{MWD/ton} \right] = 1.94 \times 10^6 \frac{\sigma_{02}}{\sigma_p} \frac{k_1}{1 - k_2} \left\{ \frac{F k_1}{1 - k_2} - 1 \right\}$$

where:

$$F \equiv \frac{\Sigma^{02}}{\Sigma^{02} + \Sigma^{\text{mod}} (1 + L^2 B^2)}$$

$$k_1 \equiv \frac{\eta^{23} \epsilon_p}{1 + B^2 \tau} + 1 - x$$

$$k_2 \equiv \frac{\eta^{23} \epsilon (1 - p)}{1 + B^2 \tau}$$

Values used for fundamental nuclear constants are tabulated in the appendix.

Resonance escape probabilities (p) were determined for thorium rods in an 8-in. square lattice with a D_2O moderator. In this calculation, use was made of experimental values of the effective resonance integral for thorium rods as a function of rod size.⁽⁴⁾ An example of the calculation of p is given in the appendix.

The value of the fast effect (ϵ) was assumed to be 1.02, independent of rod size.

In equations (1) and (2), buildup of fission product poisons during operation is accounted for by separating the poisons into two groups.⁽⁵⁾ The large cross section poisons Xe^{135} and Sm^{149} , which saturate after short exposures, were treated as a constant fraction (x) of the U^{233} absorption cross section. The remaining group of "stable" fission products were then treated as a single poison with an effective thermal cross section (σ_p). Estimates from data presented in reference (5) gave σ_p as 60 barns per fission product pair and x within the range 0.03 to 0.05.

APPENDIXA. Nuclear Constants

The values of nuclear constants used in the computations are listed in Tables (1) and (2), below:

Table (1) Thermal Cross Sections and Fission
Parameters at 250°C*

<u>Element</u>	<u>σ_a (barns)</u>
U^{233}	394 ($\eta = 2.25$; $\nu = 2.50$ (<u>2</u>))
Th^{232}	4.66
D_2O	1.37×10^{-3} (<u>7</u>)
Fission Products	60 barns/F.P. pair (<u>5</u>)

* Except where otherwise noted, values taken from ref. (6) and corrected for thermal distribution at 250°C.

Table (2) Constants for Heavy Water at 250°C⁺

Density, gm/cm ³	0.88 (<u>8</u>)
τ , cm ²	197
L^2 , cm ²	35,440
$\xi \sigma_s$ (resonance), barns	5.009

⁺Values taken from reference (7), except where otherwise noted.

B. Derivation of IBR and Fuel Exposure:

The system of equations describing the operation of the reactor are given as follows:

$$\text{Criticality:} \quad k_1 \Sigma^{23}(t) = \Sigma^P(t) + \Sigma^c \quad (1)$$

Buildup of "Stable" fission product poisons:

$$\frac{dN^P}{dt} = \beta \Sigma^{23}(t) \phi(t) - \Sigma^P(t) \phi(t) \quad (2)$$

Definition of Breeding Ratio:

$$\text{B. R.} = \frac{\Sigma^{02}}{\Sigma^{23}(t)} + k_2 \quad (3)$$

where the following groups have been defined:

$$k_1 \equiv \frac{\eta \epsilon p}{1 + \beta^2 \tau} - 1 - x$$

$$k_2 \equiv \frac{\eta \epsilon (1 - p)}{1 + \beta^2 \tau}$$

$$\Sigma^c \equiv \Sigma^{02} + \Sigma^{\text{mod}} + DB^2$$

The reactor is assumed to operate at constant power density:

$$P = K \beta \Sigma^{23}(t) \phi(t) = \text{constant} \quad (4)$$

$$\left(\frac{1}{K} = 3.1 \times 10^{10} \text{ fission/watt sec} \right)$$

From equation (1)

$$\Sigma^{23}(t) = \frac{\Sigma^P(t)}{k_1} + \frac{\Sigma^c}{k_1} \quad (5)$$

Substituting (5) into (2) and changing the variable to flux time;

$$\text{i.e., } T = \int_0^t \phi(t) dt ;$$

$$\frac{1}{\sigma_p} \frac{d\Sigma^D(T)}{dT} = \left(\frac{\beta}{k_1} - 1 \right) \Sigma^P(T) + \frac{\beta}{k_1} \Sigma^c \quad (6)$$

The solution of equation (6), with the initial condition that $\Sigma^P(0) = 0$ yields:

$$\Sigma^P(T) = \frac{\beta}{k_1 \left(1 - \frac{\beta}{k_1} \right)} \left[1 - e^{-\left(1 - \frac{\beta}{k_1} \right) \sigma_p T} \right] \quad (7)$$

and from (5):

$$\Sigma^{23}(T) = \frac{\Sigma^c}{k_1 \left(1 - \frac{\beta}{k_1} \right)} \left[1 - \frac{\beta}{k_1} e^{-\left(1 - \frac{\beta}{k_1} \right) \sigma_p T} \right] \quad (8)$$

The IBR is now obtained from equation (3):

$$\begin{aligned} \text{IBR} &= \frac{\Sigma^{02}}{\Sigma^{23}(0)} + k_2 \\ &= \Sigma^{02} \frac{k_1}{\Sigma^c} + k_2 \\ &= F k_1 + k_2 \end{aligned} \quad (9)$$

Combining equation (3) with the limiting condition that the total burnup of U^{233} during the fuel irradiation cycle be equal to the bred fuel:

$$\begin{aligned}
 \int_0^{t_0} (B.R. - 1) dt &= \int_0^{T_0} (B.R. - 1) \frac{dT}{\phi} \\
 &= \frac{K\beta}{P} \int_0^{T_0} \left[\Sigma^{02} + (k_2 - 1) \Sigma^{23}(T) \right] dT = 0
 \end{aligned} \tag{10}$$

Thus:

$$\Sigma^{02} T_0 + (k_2 - 1) \int_0^{T_0} \Sigma^{23}(T) dT = 0 \tag{11}$$

From (8):

$$\begin{aligned}
 \int_0^{T_0} \Sigma^{23}(T) dT &= \frac{\Sigma^c}{k_1 \left(1 - \frac{\beta}{k_1}\right)} \left[T_0 + \frac{\frac{\beta}{k_1} \left(e^{-\left(1 - \frac{\beta}{k_1}\right) \sigma_p T_0} - 1 \right)}{\left(1 - \frac{\beta}{k_1}\right) \sigma_p} \right] \\
 &= \frac{P}{K\beta} t_0
 \end{aligned} \tag{12}$$

By trial and error equations (11) and (12) may be solved for the maximum exposure (t_0). However, numerical estimates show that the exponential term in (12) can be approximated with sufficient accuracy by the first three terms in the series expansion:

$$e^{-\left(1 - \frac{\beta}{k_1}\right) \sigma_p T_0} \simeq 1 - \left(1 - \frac{\beta}{k_1}\right) \sigma_p T_0 + \frac{\left(1 - \frac{\beta}{k_1}\right)^2 \sigma_p^2 T_0^2}{2} \tag{13}$$

With this simplification, equation (11) can be solved for T_o :

$$T_o \approx \frac{2k_1}{\beta \sigma_p} \left[\frac{\Sigma^{02}}{\Sigma^c} \frac{k_1}{(1 - k_2)} - 1 \right] \quad (14)$$

Solving (12) for t_o :

$$t_o = \frac{\beta K}{P} \frac{\Sigma^c}{k_1} \left[T_o + \frac{\beta}{k_1} \frac{\sigma_p T_o^2}{2} \right] \quad (15)$$

Substituting (14) into (15);

$$t_o = \frac{2K}{P} \frac{\Sigma^{02}}{\sigma_p} \frac{k_1}{1 - k_2} \left[\frac{\Sigma^{02}}{\Sigma^c} \frac{k_1}{(1 - k_2)} - 1 \right] \quad (16)$$

From the relations:

$$\begin{aligned} \text{Exposure} \left(\frac{\text{MWD}}{\text{ton}} \right) &= t_o \text{ (sec)} \times \frac{1}{86,400} \left(\frac{\text{days}}{\text{sec}} \right) \times P \left(\frac{\text{watts}}{\text{cm}^3} \right) \times 10^{-6} \left(\frac{\text{Mw}}{\text{watt}} \right) \\ &\times 10^6 \left(\frac{\text{gms}}{\text{ton}} \right) \times \frac{1}{c_{02}} \left(\frac{\text{cm}^3}{\text{gm Th}} \right) \end{aligned}$$

$$\Sigma^{02} = \frac{0.602 \times 10^{24}}{232} c_{02} \sigma_{02}$$

$$\text{Exposure} \left(\frac{\text{MWD}}{\text{ton}} \right) = 1.94 \times 10^6 \left(\frac{\sigma_{02}}{\sigma_p} \right) \frac{k_1}{1 - k_2} \left[F \frac{k_1}{(1 - k_2)} - 1 \right] \quad (17)$$

C. Calculation of p:

The resonance escape probabilities were calculated from the empirical formula:

$$p = e^{-\frac{N_{02} R_a^{\text{eff}}}{\xi \Sigma_s}}$$

Values of R_a^{eff} , as a function of the surface-to-mass ratio of the fuel rod, were obtained from reference (4). An example is given below:

For $c_{O2} = 400$ gms/liter;

$$\text{Volume Fraction Thorium} = \frac{\pi r^2}{(8 \times 2.54)^2} = \frac{0.4}{\rho_{O2}}$$

Taking the density of Thorium as 11.7 gms/cm^3 (7);

Rod radius = $r = 2.1 \text{ cm}$

$$\frac{S}{M} = \frac{2 \pi r}{\rho_{O2} \pi r^2} = \frac{2}{11.7 \times 2.1} = 0.081$$

From reference (4), for $\frac{S}{M} = 0.081$, $R_a^{\text{eff}} \simeq 10.5$ barns

Thus, from Table (2) and the above equation:

$$p = \exp \left[- \frac{0.4 \times 10.5 \times 20}{232 \times 5.009 \times 0.88 \times 0.97} \right] = 0.92$$

D. Nomenclature:

B^2	Buckling (cm^{-2})
C_{02}	Concentration of Thorium in reactor (gms/cm^3)
L	Diffusion length (cm)
N^{02}	Atom concentration of Thorium in reactor (atoms/cm^3)
p	Resonance escape probability
r	Radius of Thorium rod (cm)
R_a^{eff}	Effective resonance integral for Thorium cylinders (cm^2)
t	Fuel exposure time (sec); t_0 is the maximum fuel exposure time for no net breeding loss
T	Flux time (cm^{-2})
x	Fraction of absorbtions in U^{233} which produce high cross section fission product poisons
ϵ	Fast fission factor
ρ_{02}	Density of Thorium rod (gms/cm^3)
η, ν	Fission parameters of U^{233} ; $\beta = \frac{\eta}{\nu} = \frac{\sigma_f^{23}}{\sigma^{23}}$
ξ	Average neutron lethargy increment per collision
σ	Microscopic thermal absorbtion cross section (cm^2)

D. Nomenclature (Cont'd):

Σ Macroscopic thermal absorbtion cross section (cm^{-1})

Superscripts: 23 = U^{233}

02 = Th^{232}

p = "Stable" fission products

mod = D_2O

Σ_s Macroscopic scattering cross section for D_2O in resonance region (cm^{-1})

τ Fermi Age (cm^2)

ϕ Thermal neutron flux

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