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SUBJECT: Experimental Determination of HRE-3 Breeding Ratio

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ABSTRACT

The accuracy with which the breeding ratio of HRE-3 could be determined after a period of reactor operation was investigated. Inaccuracies in measurement of the core U^{233} inventory and blanket U^{233} and Pa^{233} inventories appear to be the major sources of error. Appreciable errors could result from attempting to determine these inventories by sampling the reactor contents. For example, if generalized attack on stainless steel is at a rate of 1.0 mpy and if the associated film of corrosion products is 1% uranium, failure to account for this fuel in evaluation of the core inventory would cause an error of about 5% in the breeding ratio.

NOTICE

DISCLAIMER

EXPERIMENTAL DETERMINATION OF HRE-3 BREEDING RATIO

M. W. Rosenthal

INTRODUCTION

A major purpose in building HRE-3 would be to demonstrate that an aqueous homogeneous power reactor is capable of producing more fuel than it consumes. To accomplish this objective, it must be possible to determine the breeding ratio with sufficient accuracy to show that there is a net breeding gain. An examination has therefore been made of methods by which the breeding ratio of HRE-3 could be ascertained. In all cases the reactor was assumed to be started clean and operated for some period of time, after which the average breeding ratio during that period is evaluated.

METHOD OF DETERMINING BREEDING RATIO

The mean breeding ratio over a period of time, represented by $BR_m(o \rightarrow t)$, is defined as the ratio of the fissionable material produced to that which is destroyed; the amount produced is the algebraic sum of that destroyed, or "burn-up" (BU), and the change in the inventory of the system, ΔI. Mathematically, this can be expressed by

$$BR_m(o \rightarrow t) = \frac{BU + \Delta I}{BU} = 1 + \frac{\Delta I}{BU} \tag{1}$$

The burn-up is equal to the sum of the amounts of U^{233} and U^{235} destroyed in the core and the blanket

$$BU = BU_c^{23} + BU_c^{25} + BU_b^{23} + BU_b^{25} \tag{2}$$

where the superscripts "23" and "25" refer to U^{233} and U^{235}, respectively, and where the subscripts "c" and "b" refer to the core and blanket regions, respectively. The change in inventory is equal to the system inventory at time t, plus any withdrawals made during the period considered, and minus the fuel added to the system (the initial fuel inventory is considered as a fuel addition). This can be expressed by

$$\Delta I = I_c^{23} + I_c^{25} + I_b^{23+13} + I_b^{25} + W^{23} + W^{25} - F^{23} - F^{25} \qquad (3)$$

where I is the inventory, W the fuel withdrawal, and F the fuel addition. The superscript "13" refers to Pa^{233}. The quantities in the preceding equations can be determined by the methods discussed below.

Determination of F^{23}, F^{25}: The amount of uranium added to the system can be measured as accurately as needed. The isotopic composition can be determined by mass-spectrometric means, and equipment giving analyses as accurately as desired will probably be available.

Determination of W^{23}, W^{25}: The most accurate method of obtaining these quantities is to dissolve all material withdrawn from the system (to insure homogeneity), and then determine the uranium content by analysis.

Determination of I_c^{23}, I_c^{25}, I_b^{23+13}, I_b^{25}: If the fuel inventories are to be ascertained without interrupting the operation of the reactor, it must be done by sampling and analyzing the reactor contents. The danger in this procedure is, of course, that the sample may not be representative. This could easily result from the system not being homogeneous, due to possible precipitation of some uranium, or there may be fuel incorporated in a film of corrosion products. The HRT experience in determining fuel inventory illustrates the difficulties associated with this approach. It may be possible to determine inventory by always adding fuel containing a given percentage of U^{238}; determination of the uranium-isotope ratios based on U^{238} concentration would enable the inventory to be calculated. However, use of natural uranium in pretreatment and shakedown would make the U^{238} inventory uncertain. Determination of U^{233} in slurry by the ratio of U^{233} to thorium has the hazard that part of the thorium might accumulate in a stagnant region and not attain the same fuel concentration as the rest. Probably the most dependable way of obtaining the inventories is to drain the blanket and core systems, flush each with reagents that will remove any deposited fuel, and then dissolve the complete inventories. The systems would then be homogeneous and the contents could be determined accurately by analysis. The solutions might even be processed by solvent extraction and the fuel inventories recovered as metallic uranium.

<u>Determination of BU_c^{23}:</u> The core U^{233} burn-up could be determined by the net change in U^{233} inventory, given by

$$BU_c^{23} = F_c^{23} - I_c^{23} - W_c^{23} \tag{4}$$

<u>Determination of BU_c^{25}:</u> This quantity might be determined by using $\bar{\alpha}_{25}$, the average ratio of neutron captures to fissions in U^{235}, and the change in U^{236} inventory. This can be written as

$$BU_c^{25} = \left[I_c^{26} + W_c^{26} - F_c^{26} \right] \times \frac{1 + \bar{\alpha}_{25}}{\bar{\alpha}_{25}} \tag{5}$$

The correct average α_{25} would be estimated by multigroup calculations to properly weight the data for energy dependence. This procedure for determining BU_c^{25} ignores the loss of U^{236} by neutron absorption, but this would be insignificant for reasonable periods of operation if the feed were essentially free of U^{236}.

Determination of BU_b^{23}: Two methods of estimating the blanket U^{233} destruction appear feasible, although neither could be depended on to be very accurate: (1) isolate some long-lived radioactive fission products and determine their concentrations by counting techniques, using a system calibrated for the isotopes used; (2) use multigroup estimates of reactor conditions to compute the blanket burn-up from the measured core burn-up.

Determination of BU_b^{25}: This quantity is so small for reasonable periods of operation that it can be completely neglected ($BU_b^{25}/BU_b^{23} \approx 0.01\%$ for 480 days at 50 Mw core power).

ACCURACY OF BREEDING RATIO DETERMINATION

The maximum fractional error in the breeding ratio, $\delta BR_m/BR_m$, can be obtained from the estimated limits of error in the various quantities that are involved, and can be represented by

$$\frac{\delta BR_m}{BR_m} = \pm \sum_i \left| \left(\frac{\partial BR_m/BR_m}{\partial X/X} \right)_i \left(\frac{\delta X}{X} \right)_i \right| \tag{6}$$

where $X \equiv$ I's, F's, W's, etc., and the summation over i represents the summation over the various values of X. The equations for the partial derivatives appearing in equation (6) have been formulated and are given in the second column of Table 2 divided by ΔI/)BU + ΔI), which appears in every term.

The values of the various quantities of interest are given in Table 1 for a reactor having a core diameter of 4 ft, pressure-vessel I.D. of 9 ft, 1000 g Th/liter in the blanket, and a core power of 50 Mw (ht). Operating periods of 100 days and of one year were considered. These values were estimated using an ORACLE routine* which predicts the time-dependent behavior of two-region reactors. The feed was assumed to be pure U^{233}, the initial blanket material was free of uranium, and there were no fuel withdrawals over the operating period.

Using the values from Table 1, the errors introduced into BR_m are given in Table 2 for various assumed errors in the individual measurements. In Case I of Table 2, all of the inventories are assumed to be in error by ± 0.5%; the uncertainty in $\bar{\alpha}_{25}$ is taken as ± 20% and the error in BU_b^{23} as ± 30%. These inaccuracies would lead to a maximum error in BR_m of 1.6% after 100 days of operation and 1.3% after one year.

The consequences of failure to account for uranium in the corrosion film are treated in Cases II and III. The generalized corrosive attack on the stainless-steel surfaces is assumed to occur at a rate of 1.0 mpy, and all of the iron and chromium is considered to deposit as oxide in a film of corrosion product. If this film contains 1% uranium, the fuel associated with the corrosion products in the core system would amount to 0.39 kg after 100 days of operation; this represents 2.7% of the core inventory. For the same conditions, after one year the values would be 1.44 kg and 9.2%. The errors in inventories used in Cases II and III correspond to these conditions. For other corrosion rates and uranium concentrations the errors would be proportionate. The values in Table 2 indicate that if uranium accumulated in the corrosion film at the rate assumed, the breeding ratio could be in error by over 5% from this effect alone.

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*Melvin Tobias, M. W. Rosenthal, and T. B. Fowler, "Time-Dependent Studies of the Nuclear Characteristics of HRE-3," ORNL-CF-57-12-1 (Dec. 31, 1957).

DISCUSSION

For the assumed conditions of operation, the major sources of error would be the core inventory of U^{233}, the blanket inventory of U^{233} and Pa^{233}, and for a longer period of operation, the burn-up of U^{233} in the blanket might be important. Accumulations of uranium in the reactor that are not detected by sampling probably would produce much larger errors than those from other sources. Hence, a breeding ratio determined by draining and flushing the system is likely to be much more dependable than one obtained by sampling.

In Table 2 there appears to be little difference in accuracy between operating periods of 100 days and 1 year. However, if the rate of uranium deposition decreased with the passage of time (rather than staying constant as assumed), the error from this source would be reduced by using a longer period. This improvement would result from the uranium loss representing a smaller fraction of ΔI, the net change in the inventory of the system.

The sums of the absolute values in each column of Table 2 represent the maximum errors in BR_m which could occur with the assumed inaccuracies in the measurements. If the measurement errors were random, such as those resulting from imprecision in chemical analyses, some of these would produce positive errors in BR_m and some negative, and the net error would be less than the total. However, the errors resulting from loss of uranium are additive (except that associated with U^{236}), and no improvement in accuracy is obtained from compensating effects.

TABLE 1

CONDITIONS OF HRE-3 AFTER 100 DAYS AND ONE YEAR OF OPERATION

Quantity	Symbol	Initial	After 100 d.	After 1 year
Core U^{233} Inventory	I_c^{23}	13.50 kg	13.60 kg	13.53 kg
Core U^{234} Inventory	I_c^{24}	0	0.62	2.01
Core U^{235} Inventory	I_c^{25}	0	0.017	0.15
Core U^{236} Inventory	I_c^{26}	0	< 0.001	0.016
Blanket U^{233} Inventory	I_b^{23+13}	0	7.36	26.13
Blanket U^{234} Inventory	I_b^{24}	0	0.026	0.25
Blanket U^{235} Inventory	I_b^{25}	0	< 0.001	0.0019
Blanket U^{236} Inventory	I_b^{26}	0	< 0.001	< 0.001
Cumulative U^{233} Additions	F_c^{23}	13.50	19.94	36.10
Other Fuel Addition		0	0	0
U^{233} + U^{235} Inventory Change	ΔI	0	1.04	3.71
Burn-up of Core U^{233}	BU_c^{23}	0	6.34	22.57
Burn-up of Core U^{235}	BU_c^{25}	0	0.0028	0.11
Burn-up of Blanket U^{233}	BU_b^{23}	0	0.083	1.61
Burn-up of Blanket U^{235}	BU_b^{25}	0	< 0.001	< 0.001
Total U^{233} + U^{235} Burn-up	BU	0	6.42	24.29
Integrated Core Power		0	5,050 Mwd	18,050 Mwd
Integrated Blanket Power		0	66	1,285

TABLE 2

EFFECT OF MEASUREMENT ERRORS ON BREEDING RATIO DETERMINATION IN HRE-3

Quantity, X	$\frac{\partial BR_m/Br_m}{\partial X/X} \div \frac{\Delta I}{BU + \Delta I}$	Case I			Case II		Case III	
		$\frac{\delta X}{X}$	$\left(\frac{\delta BR_m}{BR_m}\right)_x$ for 100 days	$\left(\frac{\delta BR_m}{BR_m}\right)_x$ for 1 year	$\frac{\delta X}{X}$	$\left(\frac{\delta BR_m}{BR_m}\right)_x$ for 100 days	$\frac{\delta X}{X}$	$\left(\frac{\delta BR_m}{BR_m}\right)_x$ for 1 year
I_c^{23}	$\frac{I_c^{23}}{\Delta I} + \frac{I_c^{23}}{BU}$	± 0.5%	± 1.05%	± 0.55%	± 2.7%	± 5.66%	± 9.2%	± 5.04%
I_c^{25}	$\frac{I_c^{25}}{\Delta I}$	0.5	0.0011	0.0026	2.7	0.0060	9.2	0.048
I_c^{26}	$-\frac{BU_c^{25}}{BU} \times \frac{I_c^{26}}{I^{26} + W^{26} - F^{26}}$	0.5	< 0.001	< 0.001	2.7	< 0.001	9.2	0.0054
I_b^{23+13}	$\frac{I_b^{23+13}}{\Delta I}$	0.5	0.49	0.46				
I_b^{25}	$\frac{I_b^{25}}{\Delta I}$	0.5	< 0.001	< 0.001				
W_c^{23}	$\frac{W_c^{23}}{\Delta I} + \frac{W_c^{23}}{BU}$	0	0	0				
W_c^{25}	$\frac{W_c^{25}}{\Delta I}$	0	0	0				
W_c^{26}	$-\frac{BU_c^{25}}{BU} \times \frac{W_c^{26}}{I_c^{26} + W_c^{26} - F_c^{26}}$	0	0	0				

TABLE 2 (Cont'd)

EFFECT OF MEASUREMENT ERRORS ON BREEDING RATIO DETERMINATION IN HRE-3

Quantity, X	$\frac{\partial BR_m/Br_m}{\partial X/X} \div \frac{\Delta I}{BU + \Delta I}$	Case I			Case II		Case III	
		$\frac{\delta X}{X}$	$\left(\frac{\delta BR_m}{BR_m}\right)_x$ for 100 days	$\left(\frac{\delta BR_m}{BR_m}\right)_x$ for 1 year	$\frac{\delta X}{X}$	$\left(\frac{\delta BR_m}{BR_m}\right)_x$ for 100 days	$\frac{\delta X}{X}$	$\left(\frac{\delta BR_m}{BR_m}\right)_x$ for 1 year
F_c^{23}	$-\frac{F_c^{23}}{\Delta I} - \frac{F_c^{23}}{BU}$	0	0	0				
F_c^{25}	$-\frac{F_c^{25}}{\Delta I}$	0	0	0				
F_c^{26}	$\frac{BU_c^{25}}{BU} \times \frac{F_c^{26}}{I_c^{26} + W_c^{26} - F_c^{26}}$	0	0	0				
$\bar{\alpha}_{25}$	$-\frac{BU_c^{25}}{BU}$	17	0.0010	0.010				
BU_b^{23}	$-\frac{BU_b^{23}}{BU}$	30	0.054	0.26				
BU_b^{25}	$-\frac{BU^{25}}{BU}$	0	0	0				
TOTAL			± 1.6%	± 1.3%		± 5.7%		± 5.1%

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