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HYDROGEN SLOWING-DOWN METHOD FOR
CRITICALITY CALCULATIONS

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ABSTRACT

A criticality equation is derived assuming slowing-down only by hydrogen. The use of criticality experiments as a basis for calculation is described. Approximate methods are given for handling inhomogeneities and multi-region reactors. Some criticality data on solutions of UO_3 dissolved in H_3PO_4 are listed in Appendix I, and the Goertzel-Selengut equations are derived in Appendix II.

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INTRODUCTION

The present report is concerned with criticality calculations for liquid reactors, the principal constituents of which are water and enriched uranium (93.5% U-235). The liquid is assumed contained in a steel vessel, which has some tamping effect. Slowing-down is due almost entirely to H, and fast neutron absorption is due almost entirely to epithermal capture in U.

The equations derived in this report have the following features:

- (1) The criticality equation contains a "first flight correction."
- (2) Account is taken of epithermal fission.
- (3) Account is taken of neutron absorption and leakage in the slowing-down integral.
- (4) The slowing-down integral is conveniently transformed into a sum, with different fast neutron cross sections used in different energy intervals.

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SECTION I. DERIVATION OF CRITICALITY EQUATION

When a neutron of energy E' is scattered by H, all final energies from zero to E' have equal probability $P(E)$: $P(E)dE = dE/E'$. The hydrogen slowing-down (HSD) method differs from age theory by taking account of this spectrum of final energies following a collision with H.

The following assumptions are made:

- (1) The fission spectrum is disregarded. It is assumed that $Q_0(\bar{r})$ fast neutrons are born in fission per cubic centimeter per second at energy E_0 and position \bar{r} .
- (2) Slowing-down is due only to collisions with H, and not to collisions with heavier elements (justification: in H_2O , oxygen contributes only 2.5% to the slowing-down cross section).
- (3) Capture of fast neutrons is due entirely to uranium.
- (4) The flux $\phi(E, \bar{r})$ is separable into a function of energy E and a function of position \bar{r} . The space dependence of $Q_0(\bar{r})$ and of the flux at all energies is considered to be the same. Hence the buckling B^2 of the assembly is the same for all energy neutrons, and the extrapolation distance δ beyond the physical surface of the core is the same for all energy neutrons.

If the fission spectrum is taken into account, and also slowing-down by heavy elements (changing assumptions 1 and 2), then the Goertzel-Selengut equations⁽¹⁾ are obtained. These equations, derived in Appendix II, require more elaborate numerical methods for solution than the criticality equation derived below.

The basic equation of diffusion theory is $\bar{J} = -D \text{grad } \phi$, where \bar{J} is the net neutron current. In diffusion theory, $D = 1/3 \Sigma_s$ where Σ_s is the macroscopic scattering cross section. A first-order transport correction is preferred for D

$$D = \frac{1}{3} \left[\Sigma_{tr} + \Sigma_a \left(\frac{1}{5} + \frac{4}{5} \cdot \frac{2}{3A} \right) \right]^{-1}$$

where the transport cross section $\Sigma_{tr} = \Sigma_s (1 - \overline{\cos \theta})$, $\overline{\cos \theta} = 2/3A$, θ is the scattering angle, A is the atomic weight of the scattering nucleus, and Σ_a is the macroscopic absorption cross section. A good approximation for D is

$$D = \frac{1}{3 \Sigma_{tr} + 2.2 \Sigma_a(H) + 0.6 \Sigma_a(\text{heavy elements})}$$

For each energy interval dE the following equation is written

$$D \nabla^2 \phi(E) - (\Sigma_a + \Sigma_{sh}) \phi(E) + S(E) = 0 \quad (1)$$

(1) G. Goertzel, TAB-53 (1950).

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where Σ_{sh} is the macroscopic hydrogen scattering cross section, and $S(E)$ is the number of neutrons scattered into the energy interval dE per second per cubic centimeter. The spatial dependence of $\phi(E)$ is given by $\nabla^2 \phi + B^2 \phi = 0$, so (1) can be written

$$-\Sigma_L \phi(E) + S(E) = 0 \tag{2}$$

where the "loss" cross section $\Sigma_L = \Sigma_a + \Sigma_{sh} + DB^2$.

The source term $S(E)$ is given by

$$S(E)dE = Q_0 \frac{\Sigma_{sh}(E_0)}{\Sigma_L(E_0)} \frac{dE}{E_0} + \int_E^{E_0} \Sigma_{sh}(E') \phi(E') \frac{dE'}{E'} \tag{3}$$

The first term gives the neutrons scattered into dE on their first collision with H; the integral gives the neutrons scattered into dE from dE' . The symbol ξ will be used for the first collision probability $\Sigma_{sh}(E_0)/\Sigma_L(E_0)$. Differentiating $S(E)$,

$$\frac{dS}{dE} = -\frac{\Sigma_{sh}(E)\phi(E)}{E} = -\frac{\Sigma_{sh}(E)}{E} \cdot \frac{\phi(E)\Sigma_L(E)}{\Sigma_L(E)} = -\frac{\Sigma_{sh}(E)}{E} \cdot \frac{S(E)}{\Sigma_L(E)} \tag{4}$$

where a substitution has been made using equation (2). This integrates to

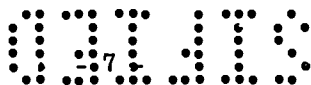
$$\ln \frac{S(E_t)}{S(E_0)} = \int_{E_t}^{E_0} \frac{\Sigma_{sh}(E)}{\Sigma_L(E)} \frac{dE}{E} \tag{5}$$

where E_t will be considered the upper limit of the thermal energy distribution. From equation (3), $S(E_0) = Q_0 \xi / E_0$. Since

$$\frac{\Sigma_{sh}}{\Sigma_L} = 1 - \frac{\Sigma_a}{\Sigma_L} - \frac{DB^2}{\Sigma_L}$$

we have

$$S(E_t) = \frac{Q_0 \xi}{E_0} \exp \left[\int_{E_t}^{E_0} \frac{dE}{E} - \int_{E_t}^{E_0} \frac{\Sigma_a}{\Sigma_L} \frac{dE}{E} - B^2 \int_{E_t}^{E_0} \frac{D}{\Sigma_L} \frac{dE}{E} \right] \tag{6}$$



It will be noted that equation (6) has omitted neutrons scattered to energies below E_t on the first collision. Since these are of the order of $Q_0 \times 10^{-7}$, this neglect is justified.

For the source term for the thermal group of neutrons, we want not only the neutrons scattered into an interval dE at E_t , but all neutrons scattered below the energy E_t . We therefore replace $S(E)dE$ with $S(E_t) \cdot E_t$, and write for the thermal group of neutrons

$$D\nabla^2 \phi_t - \Sigma_a \phi_t + S(E_t) \cdot E_t = 0 \quad (7)$$

where

$$\phi_t = \int_0^{E_t} \phi(E) dE$$

Multiplying equation (6) by E_t , and noting

$$\exp \int_{E_t}^{E_0} \frac{dE}{E} = E_0/E_t$$

we have

$$S(E_t) \cdot E_t = Q_0 \xi \exp \left[- \int_{E_t}^{E_0} \frac{\Sigma_a}{\Sigma_L} \frac{dE}{E} \right] \exp \left[- B^2 \int_{E_t}^{E_0} \frac{D}{\Sigma_L} \frac{dE}{E} \right] \quad (8)$$

The first exponential is the resonance escape probability p_a , and the second exponential is the fast neutron non-leakage probability p_f .

For steady state, Q_0 is given by

$$Q_0 = \phi_t \Sigma_a \eta + Q_0 \xi p_f (1 - p_a) (\eta f) \quad (9)$$

The first term is the number of fast neutrons born per second per cubic centimeter due to capture of thermal neutrons. $\eta = \Sigma_f \nu / \Sigma_a$ where Σ_f is the macroscopic thermal fission cross section and ν is the number of neutrons per fission ($\nu = 2.5$). The second term is the contribution of epithermal fissions. The expression assumes that absorption $(1 - p_a)$ takes place below some energy E_1 , and that the leakage $(1 - p_f)$ takes place above E_1 . The number of neutrons produced per epithermal neutron capture is ηf , where f is a number between 0.86 and 1.00.

Rearranging equation (9) and substituting in equation (8) gives

$$S(E_t) \cdot E_t = \phi_t \Sigma_a \cdot \frac{\eta \xi p_a p_f}{1 - \eta \xi (1 - p_a) p_f} = \phi_t \Sigma_a \cdot \chi \quad (10)$$

Equation (7) becomes

$$D \nabla^2 \phi_t + \Sigma_a (\chi - 1) \phi_t = 0 \quad (11)$$

$$\frac{\nabla^2 \phi_t}{\phi_t} = -B^2 = -\frac{\Sigma_a}{D} (\chi - 1) = -\frac{(\chi - 1)}{L^2}$$

where L is the diffusion distance for thermal neutrons. The criticality condition is $\chi = 1 + L^2 B^2$. It can be shown⁽²⁾ that the reproduction constant k is given by $k = \chi(1 + L^2 B^2)^{-1}$. The non-leakage probability p_t of thermal neutrons is $(1 + L^2 B^2)^{-1}$.

The expression

$$1 + L^2 B^2 = \chi = \frac{\eta \xi p_a p_f}{1 - \eta \xi (1 - p_a) p_f}$$

can be rearranged to

$$1 = \left(\frac{\xi p_f p_a}{1 + L^2 B^2} \right) \eta + \xi p_f (1 - p_a) \eta f \quad (12)$$

The first term on the right may be identified with neutrons born in thermal fission and the second term with neutrons born in epithermal fission. The form of equation (12) [and also of equation (9)] depends on $k = 1$, and therefore (12) is the steady state criticality equation.

(2) S. Glasstone and M. C. Edlund, "The Elements of Nuclear Reactor Theory," Van Nostrand, New York, 1952.

SECTION II. SOLVING THE CRITICALITY EQUATION

Equation (12) can be rearranged to

$$1 = \xi \eta p_f p_t \left[1 - (1 - p_a)(1 - f/p_t) \right] \quad (13)$$

where $p_t = (1 + L^2 B^2)^{-1}$. For a given class of reactors, $(1 - p_a)(1 - f/p_t)$ can be estimated sufficiently accurately, and treated as a small correction.

Valerino and Yeager⁽³⁾ have made extensive calculations on H₂O and enriched uranium (93% U-235) systems with different H/U ratios, and from their results, the following values of f (Table 1) are deduced corresponding to various values of η for these systems.

TABLE 1

THE VALUE OF f TO USE WITH η

η	f
1.0	1.000
1.1	0.990
1.2	0.980
1.3	0.970
1.4	0.960
1.5	0.949
1.6	0.939
1.7	0.928
1.8	0.917
1.9	0.903
2.00	0.887
2.05	0.876
2.10	0.860

As an example, for the LASL DIR-P reactor, $(1 - p_a) \approx 0.3$, and $(1 - f/p_t) \approx 0.03$, so $(1 - (1 - p_a)(1 - f/p_t))$ is taken to be 0.99. This leads to

$$\frac{1 + L^2 B^2}{0.99 \eta \xi} = \exp - B^2 \int_{E_t}^{E_0} \frac{D}{\Sigma_a + \Sigma_{sh} + DB^2} \frac{dE}{E} \quad (14)$$

Equation (14) is used to determine B^2 , which in turn determines the dimensions of a just-critical reactor. The method of solving equation (14) has been to use an approximate value B_a^2 in place of B^2 inside the integral and in the expression $(1 + L^2 B^2)$. B^2 in front of the

(3) M. F. Valerino and P. Yeager, "Brief Summary of NACA Work on Criticality Calculations of Enriched Hydrogen Moderated Reactors," to be published as an NACA report.

integral is the important quantity, since here it determines the fast neutron leakage. After a first solution of the equation, a better value for B_a^2 may be used, the final value of B^2 being obtained by iteration.

If the integral is replaced by a sum, the equation can be written

$$\ln \left(\frac{0.99 \eta \xi}{1 + L^2 B_a^2} \right) = B^2 \left[\sum_{E_t}^{E_o} \frac{\Delta E/E}{B_a^2 + (\Sigma_a + \Sigma_{sh})/D} \right] \quad (15)$$

The interval ΔE has been taken to be a decade (for example, 10^5 to 10^6 ev), because this is convenient for determining average values of Σ_a , Σ_{sh} , and D for an interval from fast neutron cross section curves. $\Delta E/E$ is then 2.3. E_t was taken to be 0.0316 ev, and a half decade was used for the lowest energy interval. For a solution of 0.5 M UO_3 dissolved in 4.26 M H_3PO_4 in a critical assembly, the sum in equation (15) is found to have the approximate numerical value of 12.4 cm^2 . This value is about a third the expected "neutron age" in this solution because (1) it measures a slowing-down migration area after the first flight, and (2) absorption and leakage appear in the denominator of the integral in equation (14), in addition to slowing-down.

SECTION III. USE OF CRITICALITY MEASUREMENTS

The assumption that B^2 is the same for all energy neutrons implies an untamped assembly. Since small homogeneous reactors are usually contained in a steel vessel and tamped by the steel walls, calculations using HSD theory are made for the "equivalent" untamped reactor. The appropriate extrapolation distance δ into the steel walls surrounding the core is best determined from criticality experiments. If the reactor is cylindrical, δ is related to B^2 through the equation

$$B^2 = \left(\frac{2.405}{R + \delta_R} \right)^2 + \left(\frac{\pi}{H + 2\delta_H} \right)^2$$

where R and H are the just-critical radius and height of the core. δ_R and δ_H may be different and in principle can be determined separately from critical measurements on the same solution contained in cylinders of different radii. In the following discussion it is assumed that δ_R and δ_H are equal.

The first collision probability ξ is also very difficult to calculate accurately. The four criticality measurements (without stainless steel grids) reported in Appendix I were used to determine both δ and ξ for solutions of UO_3 in H_3PO_4 at room temperature, in a cylindrical container with 3" iron on all sides. During the calculations, a large trend in δ was found for the four solutions, and the direction of the trend (increasing or decreasing δ with increasing H/U ratio) depended on the value chosen for ξ . Since the transport mean-free-path λ_{tr} is very nearly the same for the four solutions, that value of ξ ($\xi = 0.88$) was chosen for which δ had no trend, but only a small random variation within the experimental uncertainty of the measurements. Table 2 gives calculations made on these four solutions as an example of the use of equation (15). The average δ for the four solutions is 3.29 cm.

Also listed in Table 2 are calculations on a solution which may be used in the DIR-P reactor: 0.6 M UO_3 in 5.6 M H_3PO_4 at 430°C. The vessel filling at room temperature (58%) is such that the solution at 430°C is expected to have expanded by a factor 1.43, and to have lost to the vapor phase 2.09 moles of H_2O per liter of expanded liquid. Because of this reduced H concentration, λ_{tr} and δ are expected to be larger by the factor 1.61 than they are for the room temperature solutions.

TABLE 2
 CALCULATIONS ON SOLUTIONS OF UO_3 IN H_3PO_4 USING EQUATION (15)
 $\nu = 2.5, \xi = 0.88$

Quantity	Units	Sol. 1	Sol. 2	Sol. 3	Sol. 4	<u>0.6 M UO_3 5.6 M H_3PO_4 430°C</u>
η	Dimensionless	1.903	1.885	1.857	1.816	1.947
L^2	cm ²	1.79	2.00	2.23	2.60	6.08
B_a^2	cm ⁻²	0.0352	0.0335	0.0324	0.0309	0.0150
$\frac{0.99 \eta \xi}{1 + L^2 B_a^2}$	Dimensionless	1.536	1.515	1.487	1.443	1.531
$\ln\left(\frac{0.99 \eta \xi}{1 + L^2 B_a^2}\right)$	Dimensionless	0.4292	0.4154	0.3968	0.3667	0.4258
$\sum\left(\frac{\Delta E/E}{B_a^2 + (\Sigma_a + \Sigma_{sh})/D}\right)$	Dimensionless	12.40	12.40	12.18	12.10	28.29
B^2	cm ⁻²	0.03461	0.03350	0.03258	0.03031	0.01505
Core radius	cm	15.75	15.75	15.75	15.75	17.75
Core height	cm	16.4 ^x	17.2 ^x	18.0 ^x	19.5 ^x	38.02*
δ	cm	3.30*	3.28*	3.26*	3.32*	5.30

* This number was the unknown.

x This number determined by experiment; see Appendix I.

SECTION IV. APPROXIMATE TREATMENT OF INHOMOGENEITIES

Baffles, struts, control rod thimbles (but not control rods), and similar protuberances into the core can be treated approximately by making a correction to η . These inhomogeneities displace a solution containing U, as well as introduce a mild poison into the core. Their effectiveness depends on $\phi\phi^*$ at their location, where ϕ^* is the adjoint function of the flux. $\phi\phi^*$ is approximated by ϕ^2 , where, for simplicity, ϕ^2 is calculated assuming no inhomogeneity. ϕ is determined from the functions satisfying the space equation

$$\nabla^2 \phi + B^2 \phi = 0$$

Using the subscript i to identify inhomogeneities,

$$\eta \approx \frac{\sum_f \nu V_s \overline{\phi^2}}{\sum_a V_s \overline{\phi^2} + \sum_{ai} V_i \phi_i^2} \quad (16)$$

where V_s is the volume of solution in the core, V_i is the volume of the inhomogeneity, and $\overline{\phi^2}$ is the flux-squared averaged over the volume occupied by solution.

To overcome the corrosive nature of the phosphoric acid solution, the steel vessel of the DIR-P reactor may be lined with a few mils of gold. Equation (16) may be used to estimate the poisoning effect of this lining. An alternative procedure is to estimate the reduction in δ caused by this lining. δ may be expressed as follows⁽⁴⁾

$$\delta = C \cdot \lambda_{tr} \left(\frac{1 + \beta}{1 - \beta} \right)$$

where β is the albedo for steel and C is an experimentally determined constant. Since β is the ratio of neutron current back into the core divided by neutron current which leaks out of the core, and since both currents are attenuated by the transmission of the gold lining, the effect of the gold lining can be estimated by multiplying β by the square of its transmission.

⁽⁴⁾S. Glasstone and M. C. Edlund, loc cit, p. 134.

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SECTION V. CRITICALITY ESTIMATES FOR A MULTI-REGION REACTOR

A homogeneous reactor may have regions, such as the space where cooling coils are located, where η is much smaller than in the principal part of the reactor. Criticality for an assembly of distinct homogeneous regions can be calculated approximately by a method of flux mapping. For each region the "material buckling" B_m^2 must be calculated. For a number of rapid calculations, it is simplest to use a one-group equation with an appropriate migration area M^2 .

$$B_m^2 = \frac{\eta - 1}{M^2}$$

For illustration of the method, suppose the assembly consists of a stack of several right-cylindrical sections. We then write for each cylinder

$$B_m^2 = \frac{2.405^2}{(R + \delta)^2} + C^2 = G^2 + C^2$$

G^2 can be calculated at once and we have

$$C^2 = B_m^2 - G^2$$

The C^2 gives the axial curvature of the flux distribution in this cylinder of the stack. The flux mapping is done axially along the assembly, starting with the top cylinder, and matching values of ϕ and $D \frac{d\phi}{dz}$ at each interface between cylinders. At the first interface the equations in a typical case are as follows:

$$A \sin C_1(z_1 + \delta) = F e^{iC_2(z_1 - z_1)} + E e^{-iC_2(z_1 - z_1)}$$

$$D_1 C_1 A \cos C_1(z_1 + \delta) = D_2 i C_2 \left[F e^{iC_2(z_1 - z_1)} - E e^{-iC_2(z_1 - z_1)} \right]$$

where $(z_1 - z_1)$ is the expression $(z - z_1)$ evaluated at $z = z_1$. δ is the extrapolation distance above the top of the first region; z_1 is the axial coordinate of the interface. The functions on the right side of the equations imply that $G^2 > B_m^2$ in the second region, making C_2^2 a negative number; iC_2 is taken to be a positive real number. Coefficients F and E will be obtained in terms of the arbitrary amplitude A .

The condition of criticality is that the flux should go to zero at the extrapolated boundary at the bottom of the stack of cylinders. If the flux goes to zero inside the assembly, the

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assembly is then super-critical; if it goes to zero at some distance outside the assembly, the assembly is sub-critical.

Each cylinder of the assembly can be split up into a cylindrical core and concentric annular rings. The radial contribution G^2 to the buckling can then be calculated using Bessel functions and assuming the cylinder to be infinitely long. In this case, flux mapping is done first radially and then axially. In two dimensions, the method is only approximate, because the C^2 for the axial functions for a given cylinder of the assembly is an average over the different radial regions. The method is useful for treating problems which are extremely complex to solve exactly.

The Goertzel-Selengut Equations*

This method is an improvement on HSD theory, because account is taken of the fission spectrum and of slowing-down by heavy elements. The method is not widely known, and is therefore outlined below in essentially the form given in TAB-53.

The variable used is the logarithmic energy $u = \ln E$; $du = dE/E$. When a neutron of logarithmic energy u' is scattered by H, the probability of the final energy being between u and $u + du$ is

$$P(u) du = e^u e^{-u'} du$$

For heavier elements, age theory is used. Thus the expression for neutrons scattered into the interval du is

$$S(u) = e^u \int_u^\infty \phi(u') \Sigma_{sh}(u') e^{-u'} du' + \frac{\partial}{\partial u} \left[\zeta \Sigma_s(u) \phi(u) \right]$$

where ζ is the average logarithmic energy loss per collision, and $\zeta \Sigma_s$ is the sum of products $\zeta \times \Sigma_s$ for the heavy elements. To this expression may be added a source term $g(u)$, where $g(u)du$ is the normalized fission spectrum.

$$\text{If we let } \psi(u) = e^u \int_u^\infty \phi(u') \Sigma_{sh}(u') e^{-u'} du'$$

we find that $\psi(u)$ satisfies the differential equation (as may be verified by differentiation)

$$-\Sigma_{sh} \phi(u) = \psi(u) - \frac{\partial}{\partial u} \psi(u) \quad (A1)$$

For steady state we can write an equation which balances neutron loss against neutron sources for the interval du :

$$D\nabla^2 \phi(u) - (\Sigma_a + \Sigma_{sh}) \phi(u) = \psi(u) + g(u) + \frac{\partial}{\partial u} \left[\zeta \Sigma_s \phi(u) \right] \quad (A2)$$

Slowing-down is now described by the coupled equations (A1) and (A2). Subtraction of (A1) from (A2) gives

*Method by G. Goertzel, Nuclear Development Associates, based on an equation by D. Selengut, Aircraft Nuclear Propulsion Project, General Electric.

$$D\nabla^2 \phi(u) - \Sigma_a \phi(u) = \frac{\partial}{\partial u} \left[\xi \Sigma_s \phi(u) + \psi(u) \right] + g(u) \quad (\text{A3})$$

showing that the slowing-down density $q(u) = \left[\xi \Sigma_s \phi(u) + \psi(u) \right]$, since $\left[D\nabla^2 \phi(u) - \Sigma_a \phi(u) \right]$ is the sink for neutrons, and $g(u)$ is the source.

The critical equation is written

$$\int_{u_t}^{\infty} \phi(u) \Sigma_f \nu \, du + \left[\xi \Sigma_s \phi(u_t) + \psi(u_t) \right] \cdot \left[\frac{\Sigma_f \nu}{\Sigma_a + DB^2} \right]_{u = u_t} = k \quad (\text{A4})$$

where u_t is the log of the thermal energy E_t . The integral is the epithermal fission contribution. If $g(u)$ is normalized so that

$$\int_0^{\infty} g(u) \, du = 1$$

then $\left[\xi \Sigma_s \phi(u_t) + \psi(u_t) \right]$ is the probability that a neutron born fast becomes thermal, and this expression takes the place of the HSD theory expression $p_a p_f$.

$$\left[\frac{\Sigma_f \nu}{\Sigma_a + DB^2} \right]_{u = u_t} \quad \text{gives the number of fission neutrons produced per neu-}$$

tron slowed-down to thermal. The numerical methods required to determine $\phi(u_t)$ and $\psi(u_t)$ are more elaborate than those used with HSD theory.

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