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PHYSICS RESEARCH QUARTERLY REPORT APRIL, MAY, JUNE, 1966

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BNWL-315
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The Staffs
of
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FIRST UNRESTRICTED DISTRIBUTION MADE NOV 15 '66

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PHYSICS RESEARCH QUARTERLY REPORT
APRIL, MAY, JUNE, 1966

CRITICALITY OF PLUTONIUM COMPOUNDS IN THE UNDERMODERATED RANGE, H:Pu $\leq$ 20—

L. E. Hansen, Critical Mass Physics Section

A variety of plutonium compounds are known to exist, some of which are frequently encountered during plutonium processing and fabrication operations. In the process of setting criticality safety limits for handling these compounds, one is often hampered by the limited amount, or complete lack, of criticality data available. Assumptions are frequently made which lead to quite conservative estimates of the mass required for criticality. Allowances are seldom made for departures from theoretical compound concentrations, and the critical mass is frequently based on the assumption of a hypothetical mixture of plutonium atoms in water.

Calculations were made to determine the bare and water reflected spherical critical masses of twelve of the most frequently encountered compounds, in the undermoderated range (H/Pu $\lesssim$ 20). The critical masses of plutonium atoms in water were also calculated for undermoderated systems.

Since these compounds are rarely encountered at their theoretical densities, the variation of the critical mass with changes in the concentration of the compounds was also calculated. For unreflected systems the critical mass may be expressed as $M_c \alpha$ (density)$^{-2}$, and for reflected systems $M_c \alpha$(core density)$^{-m}$ (reflector density)$^{-n}$. For reflected systems the equation is subject to the condition that m + n = 2.(1) If $\rho_{o\ core}$ and $\rho_{o\ refl.}$ are the theoretical densities of the core and reflector, and M_{c_o} the critical mass at those densities, the critical mass (M_c) for other densities may be computed from the equation

$$M_c = M_{c_o} \left(\frac{\rho_{o\ core}}{\rho_{core}}\right)^m \left(\frac{\rho_{o\ refl.}}{\rho_{refl.}}\right)^n .$$

For the water reflector at full density (ρ = 1 g/cm^3), the core density exponent (m) can easily be determined by computing the critical mass at two concentrations of compound in the core and making use of the equation $\log M_c = -m \log \rho_{core} + \log A$.

To determine the core density exponent, the spherical critical mass was computed at core densities ranging from $0.1 \lesssim \rho/\rho_o \lesssim 1.0$, where ρ_o is the theoretical concentration of the compound at a specific H:Pu ratio. The theoretical concentration was computed by volume substitution of water into the plutonium compound which was assumed to exist when there was no water present.* The spherical critical radii were computed by means of the DTF multigroup, multiregion transport theory code using an S_4 approximation in the solution of the transport equation.(2) The anisotropic scattering was defined through the first Legendre moment of the scattering kernel. The multigroup constants (18 energy groups) used in the DTF code were obtained from the GAMTEC II computer code.(3) The unreflected and fully water reflected spherical critical masses are given in Figures 1, 2, 3, and 4. The core density exponents were computed using values of ρ/ρ_0 greater than 0.2, since at $\rho/\rho_0 = 0.1$, a plot of log M_c versus log ρ/ρ_0 was no longer linear. This deviation is also shown by Paxton.(4) The calculated values of the core density exponents and the theoretical densities of the compounds are given in Table I.

The core density exponents for homogeneous mixtures of Plutonium atoms in water were also computed for various water reflector thicknesses (see Figure 5). They indicate that reflector thicknesses of 15 to 20 cm are essentially infinite. This corresponds with 6 to 8 in. water thicknesses which are generally accepted

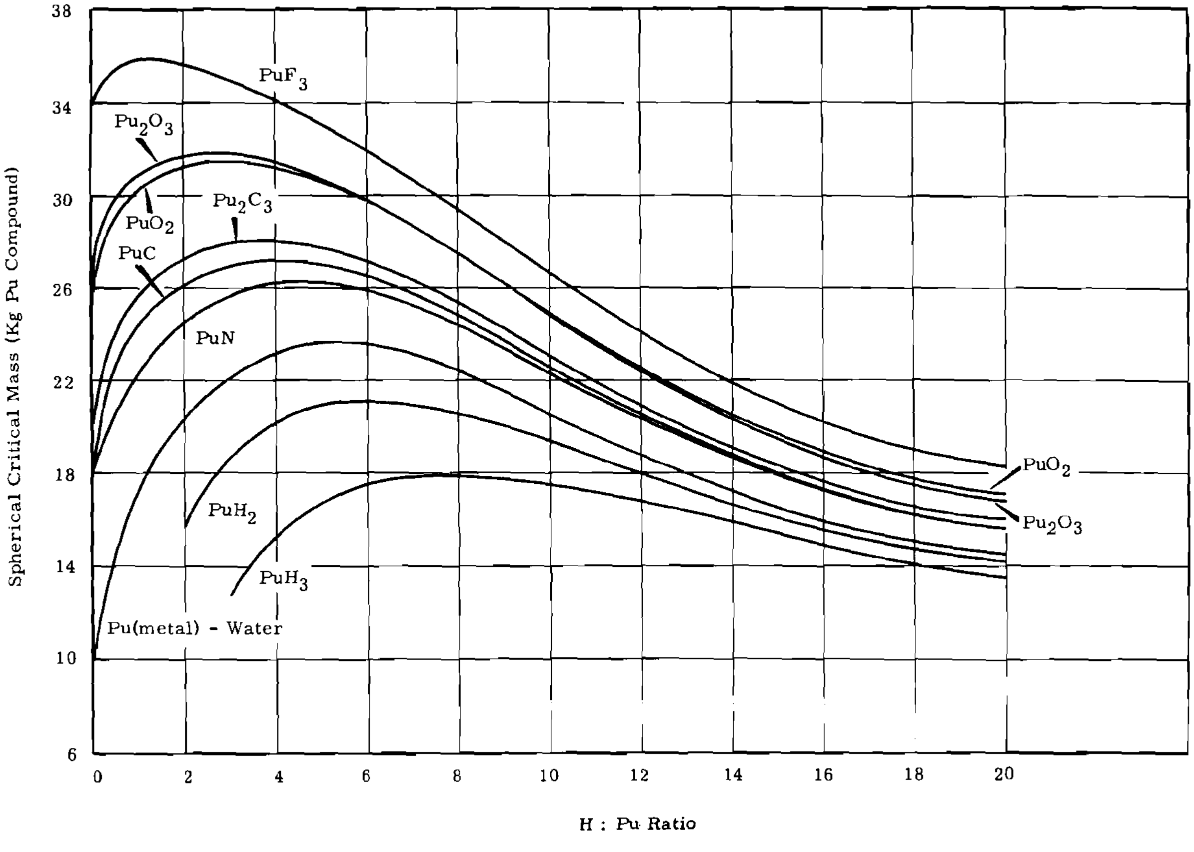

FIGURE 1. Unreflected Spherical Critical Mass for Homogeneous ^{239}Pu Compound–Water Mixtures

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* *$Pu(C_2O_4)_2$ and $Pu(NO_3)_4$ are unstable compounds and do not exist in a "dry" state.*

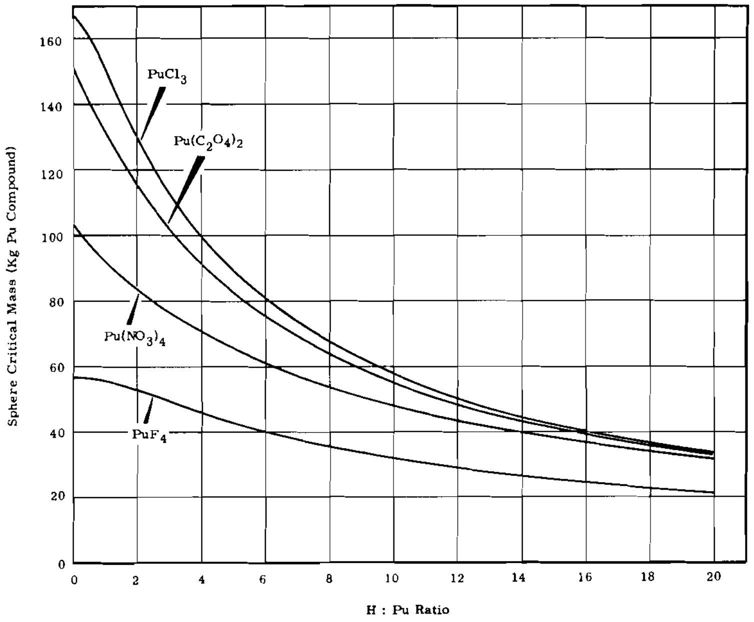

FIGURE 2. Unreflected Spherical Critical Mass for Homogeneous ^{239}Pu Compound—Water Mixtures

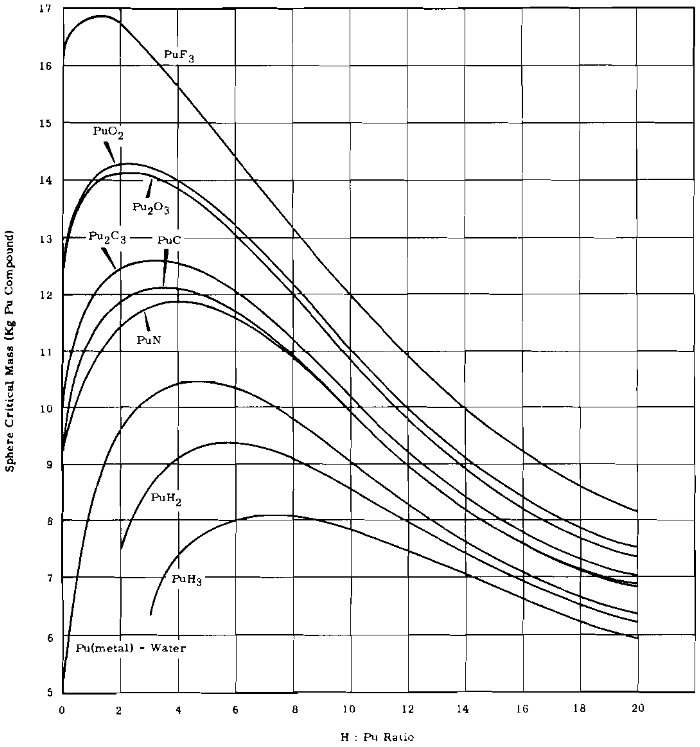

FIGURE 3. Water Reflected Spherical Mass of Homogeneous ^{239}Pu Compound—Water Mixtures

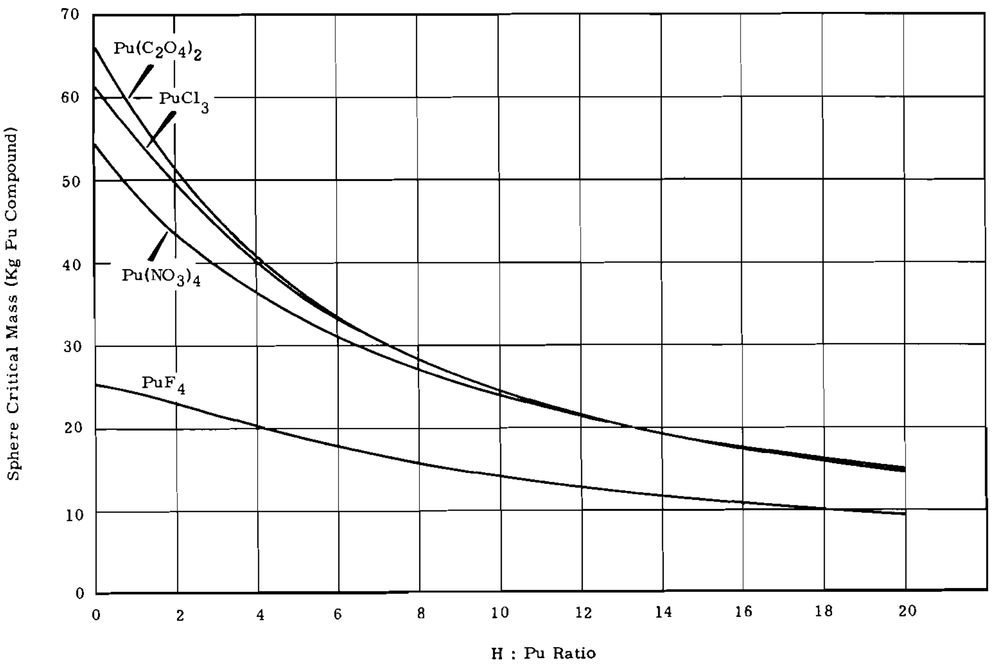

FIGURE 4. Water Reflected Spherical Critical Mass of Homogeneous ^{239}Pu Compound—Water Mixtures

TABLE I. Core Density Exponents for Plutonium Compounds in Water

		H:Pu							
Compound	ρ_o, g/cm^3	0	1	2	3	5	10	15	20
		Density Exponents							
Pu(Metal)	19.60	1.441	1.466			1.525	1.560		1.594
PuH$_2$	10.40$^{(a)}$			1.455	1.478	1.517	1.554		1.612
PuH$_3$	9.61$^{(a)}$				1.465	1.504	1.563	1.589	1.605
PuN	14.25$^{(a)}$	1.454	1.478			1.552	1.592		1.622
PuC	13.60$^{(a)}$	1.453	1.476			1.548	1.603		1.671
PuO$_2$	11.46$^{(a)}$	1.486	1.504			1.567	1.599		1.625
Pu$_2$C$_3$	12.70$^{(a)}$	1.466	1.486			1.553	1.591		1.621
Pu$_2$O$_3$	11.47$^{(a)}$	1.484	1.493			1.558	1.595		1.625
PuF$_3$	9.32$^{(b)}$	1.525	1.535			1.587	1.613		1.633
PuF$_4$	7.00$^{(b)}$	1.558	1.567			1.605	1.622		1.641
PuCl$_3$	5.70$^{(b)}$	1.623	1.622			1.659	1.677		1.692
Pu(NO$_3$)$_4$	6.20$^{(c)}$	1.697	1.699			1.693	1.688		1.681
Pu(C$_2$O$_4$)$_2$	4.50$^{(d)}$	1.674	1.675			1.676	1.676		1.660

(a) *J. J. Katz and G. T. Seaborg, "Chemistry of the Actinide Elements, Wiley, New York. 1957.*

(b) *R. K. Steunenberg and R. C. Vogel. "Fluoride and Other Halide Volatility Processes," Reactor Handbook, Fuel Reprocessing, Interscience Publishers, Inc., New York, 1961, Second Edition, vol. II, pp. 254-268.*

(c) *Density computed by a least squares fit of experimental data on Pu(NO$_3$)$_4$ solutions with excess nitrate.*

(d) *Density computed by assuming the maximum density of Pu(C$_2$O$_4$)$_2$·6H$_2$O to be 3 g/cm^3 and that the water can be replaced by an equal volume of Pu(C$_2$O$_4$)$_2$.*

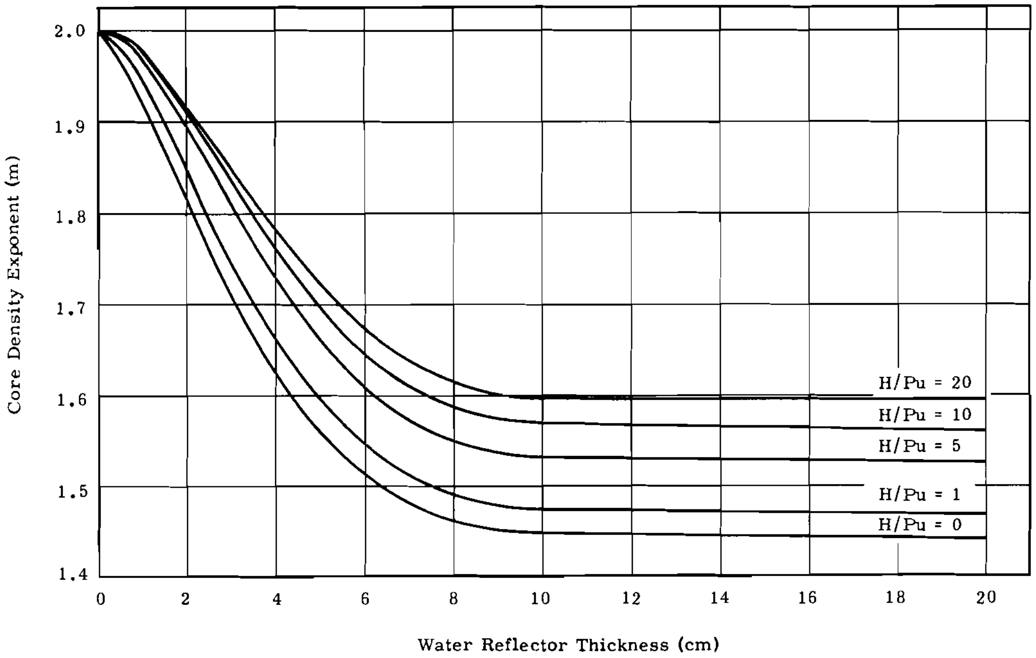

FIGURE 5. Core Density Exponents for ^{239}Pu(metal)–Water Systems as a Function of Water Reflector Thickness

as effectively infinite reflectors.

Experiments have been performed to determine the critical mass of under-moderated (H:Pu = 15) PuO_2-polystyrene assemblies both bare and Plexiglas reflected.(5) For the bare PuO_2-polystyrene assembly, 2.2 wt% Pu^{240}, the critical radius in spherical geometry was derived from experiments to be 18.58 ± 0.22 cm. The calculated critical radius, using the DTF code with multigroup constants obtained from the GAMTEC-II code, was 18.31 cm. For the Plexiglas reflected sphere, the experimentally derived critical radius was 13.54 ± 0.22 cm., which also compares favorably with the computed value of 13.46 cm. The critical height of a bare parallelepiped whose base was a 30.68 cm square was measured to be 32.84 ± 0.52 cm. The value computed using the DTF code was 33.95 cm.

By making use of the computed critical masses and core density exponents, the critical masses for these compounds may be predicted for any concentration down to 0.2 of the theoretical concentration at a specified H:Pu ratio. These data provide a simple method of obtaining estimates of the spherical critical mass of some of the most frequently encountered plutonium compounds by using density relationships.

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OPTIMUM SHIELDING—H. S. Zwibel, Theoretical Physics Unit, Nuclear Analysis Section

INTRODUCTION

Among several reasons for studying an elementary problem are that its solution can represent trends that are qualitatively similar to the unsolved real problem and that an exact solution of the simple system can be used as a proving ground for approximation (or numerical) methods that are to be applied to a complex problem. Both of these reasons motivated the following research in optimum shielding.

The system studied is an infinitesimally thin rod. It is designed so that particles can leave only at the ends; hence only two directions of motion need be specified, either right or left. This rod is uniformally composed of a material that isotropically scatters the particles. In addition, a fixed amount of a pure absorber can be embedded in the rod. The question, "How shall the absorber be distributed throughout the rod in order that the transmission coefficient be as small as possible?" is answered theoretically by variational methods later in this paper under Analytical Solution and studied numerically under Numerical Method.

THEORY

The equations satisfied by the particle flux in the rod are

$$\frac{dR(x)}{dx} + (\sigma(x) + 1/2)\ R(x) = 1/2\ L(x) \tag{1a}$$

$$\frac{-dL(x)}{dx} + (\sigma(x) + 1/2)\ L(x) = 1/2\ R(x) \tag{1b}$$

with the following boundary conditions

$$R(o) = 1 \tag{2a}$$

$$L(x_o) = 0 \tag{2b}$$

R(x) is the right moving flux, L(x) is the left moving flux, and x is the distance in the rod (total length x_o) measured in units of the scattering mean free path of the scattering medium. σ(x) is the absorption cross section and is to be chosen such that $R(x_o)$, mean free path of the scattering medium. σ(x) is the absorption cross section and is to be chosen such that R(x), the transmitted flux, is as small as possible, subject to the constraint,

$$\int_o^{x_o} \sigma(x)dx = a, \tag{3}$$

where a is the total opacity.

The problem would be straightforward if Equation 1 could be solved directly for $R(x_o)$ as a function of σ(x). This, however, is not possible in general and recourse to another method is necessary.

A tractable method is provided by variational approach. Consider the functional, $T\left\{\sigma(x)\right\}$, given by

$$T\left\{\sigma(x)\right\} = \int_0^{x_0} R(x) \quad \delta(x-x_0)\ dx$$

$$- \int_0^{x_0} R^\dagger(x) \left[\frac{dR(x)}{dx} + (\sigma(x) + 1/2)R(x) - \frac{L(x)}{2} \right] dx$$

$$- \int_0^{x_0} L^\dagger(x) \left[\frac{-dL(x)}{dx} + (\sigma(x) + 1/2)\ L(x) - \frac{R(x)}{2} \right] dx$$

$$+ \lambda \left[\int_0^{x_0} \sigma(x)\ dx - a \right] . \tag{4}$$

$R^\dagger(x)$ $\left(L^\dagger(x)\right)$ are the adjoint functions--often called importance--and represent the probability that a right (left) moving particle at x be transmitted. They satisfy the adjoint equations:

$$\frac{-dR^\dagger(x)}{dx} + (\sigma(x) + 1/2)\ R^\dagger\ (x) = 1/2\ L^\dagger(x) \tag{5a}$$

$$\frac{dL^\dagger(x)}{dx} + (\sigma(x) + 1/2)\ L^\dagger(x) = \frac{R^\dagger(x)}{2} \tag{5b}$$

with the boundary conditions

$$R^\dagger(x_0) = 1 \tag{6a}$$

$$L^\dagger(o) = 0. \tag{6b}$$

λ is an unknown constant (Lagrange multiplier) that takes account of the constraint on $\sigma(x)$.

The usefulness of this functional is that it is stationary with respect to small variations of $\sigma(x)$ about the optimum, $\sigma^*(x)$. This yields an equation connecting the fluxes, adjoints, λ, and $\sigma(x)$ which, together with the other equations (1, 3, 5), provide a solvable system (i.e., six equations for six unknowns).

To see this in more detail, vary $\sigma(x)$ by an arbitrary (but small) amount in Equation 4. This yields for

$$\delta T = T\left\{\sigma(x) + \epsilon g(x)\right\} - T\left\{\sigma(x)\right\},$$

$$\delta T = \int_0^{x_0} dx\ \delta R(x)\ \delta(x-x_0) - \int_0^{x_0} \delta R^\dagger(x) \left[\frac{dR}{dx} + (\sigma(x) + 1/2)\ R(x) - 1/2\ L(x)\right] dx$$

$$- \int_0^{x_0} R^\dagger(x) \left[\frac{d\ \delta R(x)}{dx} + (\sigma(x) + 1/2)\ \delta R(x) - 1/2\ \delta L(x) \right] dx$$

$$- \int_0^{x_0} \delta L^\dagger(x) \left[\frac{-dL}{dx} + (\sigma(x) + 1/2)\ L(x) - 1/2\ R(x) \right] dx$$

$$- \int_0^{x_0} L^\dagger(x) \left[\frac{-d\delta L}{dx} + (\sigma(x) + 1/2)\delta L(x) - 1/2\ \delta R(x) \right] dx - \int_0^{x_0} \left[R^\dagger(x)\ R(x) + L^\dagger(x)\ L(x) - \lambda \right] \epsilon g(x)\ dx$$

where only terms linear in the variation, $\epsilon g(x)$. have been retained. The variation of the fluxes, e.g., $\delta(R(x))$, is the difference between the exact fluxes for $\sigma(x)$ given by $\sigma(x) + \epsilon g(x)$, and $\sigma(x)$. Due to Equations 1 and 5 this reduces to

$$\delta T = - \int_0^{x_0} \left[R^\dagger(x)\ R(x) + L^\dagger(x)\ L(x) - \lambda \right] \epsilon g(x)\ dx, \qquad (7)$$

which, of course, is the well known first order perturbation expression. This forms the basis for both the theoretical and numerical solutions that follow.

ANALYTICAL SOLUTION

A necessary condition that an optimum distribution must satisfy is that $\delta T = 0$ for sufficiently small variations of σ about the optimum. One has from Equation 7

$$\int_0^{x_0} \left[C(x) - \lambda \right] g(x)\ dx = 0 \qquad (8)$$

where

$$C(x) \equiv R^\dagger(x)\ R(x) + L^\dagger(x)\ L(x). \qquad (9)$$

If $g(x)$ is indeed arbitrary, then the integrand must vanish, which gives

$$C(x) - \lambda = 0 \qquad (10)$$

however, one must at this point be cautious. There are certain situations under which $g(x)$ is not arbitrary. One such case is at those x values for which $\sigma^*(x)$, the optimum distribution, is zero. Since σ must be greater than or equal to zero, only positive $g(x)$ is allowed at such x values. At such x values, Equation 10 need not be true.

Assuming the validity of Equation 10, differentiating, and using the transport equations to eliminate the derivatives yields

$$L^\dagger(x)\ R(x) = R^\dagger(x)\ L(x). \qquad (11)$$

It is easy to see, however, that this equation cannot hold at the ends of the rod. Consider the right end ($x = x_0$): since $L(x_0) = 0$, then $L^\dagger(x_0) = 0$. This, with the boundary condition $L^\dagger(o) = 0$, implies that $L^\dagger$ be identically zero. This condition can be true only if there is no back scattering (since than a left moving particle has zero probability of leaving the right end). Similar conclusions are obtained at $x = 0$, due to the fact that $L^\dagger(o) = 0$: this imples that $L(x)$ be identically zero, which again can be true only if there is no back scatter.**

The only way out of this dilemma is to demand that Equation 11 not be true at either end. Since Equation 11 is a direct consequence of Equation 10

** *In fact, if there is no back scatter, then it does not matter how the absorber is distributed.*

we must have

$$C(x) \neq \lambda$$

for x at the ends. Since Equation 10 need not hold if σ is zero, a possible solution is to have $\sigma(o) = \sigma(x_o) = 0$.[***] In fact, due to the continuity of $C(x)$, $\sigma^*(x)$ will have to be zero over a finite distance from the ends.

The simplest picture that emerges is the following: $\sigma^*(x) = 0$ for some distance z in from the left end. For $0 \leq x < z$, $C(x)$ will not be constant. A similar situation holds for some distance z' in from the right end of the rod. Again $C(x)$ will not be constant. For $z \leq x \leq x_o - z'$, however, Equation 10 obtains (this assumes that $\sigma \neq 0$ in this region).

With this assumption one now attempts to solve the coupled set of nonlinear equations. What is remarkable is that a solution is readily obtained. The optimum distribution is a step function located symmetrically about the midpoint of the rod. In other words

$$\begin{aligned} \sigma^*(x) &= 0 & 0 \leq x < z \\ &= \frac{a}{x_o - 2z} & z \leq x \leq x_o - z \\ &= 0 & x_o - z < x \leq x_o \quad (12) \end{aligned}$$

where z is given by

$$z = \sqrt{\left(\frac{1 + a}{a}\right)^2 + \frac{x_o}{a}} - \left(\frac{1 + a}{a}\right). \quad (13)$$

The value of T, when $\sigma^*(x)$ is used, is

$$T^* = \frac{1}{(z + 1)} \exp\left[- \frac{a\sqrt{\alpha(\alpha + 1)}}{\alpha}\right]$$

where $\alpha = a/(x_o - 2z)$. (14)

T^* is smaller than any other that has been calculated.[**] This means that σ^* at least yields a relative minimum (and not a maximum). The possibility exists that a more complicated distribution[***] might yield a smaller minimum. This has not been investigated.

NUMERICAL METHOD

The numerical approach is straight forward, and is based on Equation 7,

$$\delta T = -\int_0^{x_o} C(x) \; \varepsilon g(x) \; dx \qquad (7)$$

This gives the change in T when one goes from $\sigma(x)$ to $\sigma(x) + \varepsilon g(x)$. To make this change in T negative (smaller transmission), one need only make $g(x)$ large where $C(x)$ is large and make $g(x)$ small where $C(x)$ is small. Except for possible second o order corrections, this will always yield a smaller value for T.[****]

A computer code, SEARCH, has been written that automates the above

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[***] *This is true even if $\sigma(x)$ has delta function discontinuities.*

[**] *Calculations have been done for $\sigma(x) = a/x_o$, and $\sigma(x) = a\delta(x-\beta)$, with β variable. In all these cases T is larger than with Equation 12.*

[***] *For example $\sigma(x)$ might have one or more zeros in the interior of the rod.*

[****] *This, of course, is why C(x) must be constant for the optimum distribution. The exception occurs at those x values for which σ is zero. If, for those x, C(x) is smaller than average, one cannot take away from $\sigma(x)$, hence C(x) will remain low.*

scheme. An arbitrary $\sigma(x)$ is chosen and $C(x)$ is determined by solving the transport equations. A new $\sigma(x)$ is chosen by multiplying the old $\sigma(x)$ by $C(x)$ and then renormalizing. Using this $\sigma(x)$, a new $C(x)$ is obtained and the process is repeated. The results of a typical calculation are shown in Figure 1. Except near the discontinuity, the agreement is very good (the value for the optimum T is quite good).

CONCLUSION

Two points emerge from this study. First, the optimum distribution could not have been guessed by intuition. Perhaps, if the solution had been the uniform distribution or a delta function, one could have guessed the answer (with a posteriori "foresight" of course), but not the peculiar step that was obtained. Since one can not "feel" the answer even in this simple problem, the solution for a realistic problem must be even harder to guess.

This leads us to the second point, as illustrated by Program SEARCH. The numerical techniques it uses are simple, straightforward, and appear to work. For complex problems, a numerical approach is probably the only way, and SEARCH can serve as a prototype. At the present time work is underway to extend SEARCH to include multienergy detail. This, of course, will vastly enlarge the type of optimum problems that can be considered; and the results should further broaden our "intuition".

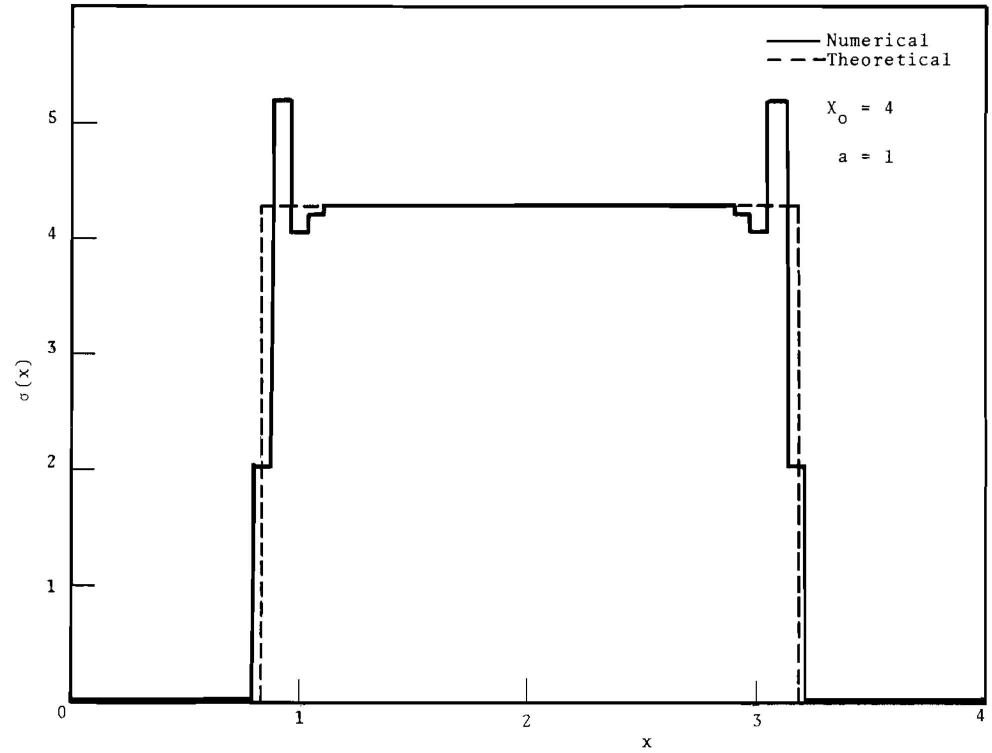

FIGURE 1. Plot of Numerical and Theoretical Values of Absorption Cross Section Versus the Distance in the Rod

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