THE UNIVERSITY OF MICHIGAN
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THE USE OF COMPUTERS TO DETERMINE TIME-DEPENDENT FISSION PRODUCT DISTRIBUTION

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ABSTRACT

In this paper are presented the assumptions, methods, and summary results of calculations employed to estimate the time-dependent radiation characteristics of fission product nuclides produced in the operation of a 500 megawatt (heat) nuclear reactor.

An electronic analog computer was used to estimate time-dependent activities in multiple-member decay chains. The use of this computer greatly eased the task of numerical computation inherent in such a program of calculations.

A knowledge of fission product activities is of importance in nuclear reactor operation, in the handling and processing of irradiated nuclear fuels, in the fabrication of incompletely decontaminated fuels, and in the handling, treatment, and disposal of fission products. Typical problems in these operations requiring knowledge of fission product activities are: 1) the prediction of radiation levels for shielding calculations; 2) the prediction of rates of heat release for the estimation of cooling requirements or thermal energy resources; and 3) the design of separation processes.

The work upon which this paper is based included the calculation of the time-dependent activities of each of the fission product nuclides by chemical elements with fission yields of 0.01% or greater, and with half-lives of 1 hour or greater.

This paper presents a summary of the following calculations:

1. The mass and activity (at 0, 1, 2, 4, 8, and 16 days after shutdown) of fission product nuclides present in the irradiated fuel of a 500 megawatt (heat) reactor operating on a 42-day cycle. The quantities are expressed in terms of both grams and curies per 10 kilograms of fissioned material.

2. The dosage rate at one meter from a point source of gamma photons, the activity of which corresponds to the total activity of each particular nuclide per 10 kilograms of fissioned material.

3. The rate of heat production in BTU's per hour from the total absorption of beta particles emitted by each particular nuclide.

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THE USE OF COMPUTERS TO DETERMINE TIME-DEPENDENT FISSION PRODUCT DISTRIBUTION

INTRODUCTION

In any installation designed to facilitate reactor operation, process spent reactor fuel, or separate fission products, consideration must be given to the amount of radioactivity present, by element, in the fission products resulting from reactor operation. The primary interest is in predicting radiation levels for shielding calculations and the rate of formation of heat at each stage of operations. It is also of interest to know the mass of each element present, including stable nuclides, in designing processes for the separation of fission products from reactor fuels or from each other.

In the course of operation of a nuclear reactor, the nuclear fuels undergo the process of fission. Each atom of fuel taking part in the fission process generally divides into two smaller atoms, usually unequal in size and each of a mass varying in probability in a certain manner about a value approximately half the mass of the original fuel atom. This process of division, known as fission, results in the release of neutrons and gamma radiation, as well as the production of the smaller atoms, known as fragment nuclides, and also as fission products.

Most of the neutrons resulting from fission are released at the instant of fission, but a small fraction, known as delayed neutrons, are released some seconds later from the fragment nuclides. The production of neutrons is

of only incidental interest to the objectives of this paper, but it does serve as an interesting parallel process to the study of the radioactivity of the fragment nuclides, or fission products. The basis of this statement is that most of the gamma radiation resulting from fission is released at the instant of fission, but a smaller fraction is released from the fragment nuclides at times varying from a fraction of a second to years following the fission process. In addition, particulate emissions, chiefly electrons, are released from the fragment nuclides after fission at rates varying widely with the identity of the fragment nuclides and in succession with, simultaneously with, or independently of the emission of gamma radiation. Consequently, the fragment nuclides display the phenomenon of radioactivity, emitting delayed gamma photons and electrons (known as beta radiation) at rates decreasing exponentially with time measured from the instant of fission. The magnitude of the rate of radioactive decay depends upon the characteristics of each fragment nuclide. Some of the fragment nuclides are stable as produced in fission, and the remainder decay to stable states by the emission of beta particles and gamma radiation.

The purpose of this paper is to examine the rates of release of gamma radiation and of beta radiation from the aggregate mass of fission products resulting from the operation of a nuclear reactor at times up to 16 days following the shutdown of the reactor. The specific activity as a function of time after reactor shutdown of the aggregate mass of all fission products (known as "gross fission products," as contrasted with the situation in which the activity of a single nuclide is examined) is independent of total reactor power, but is influenced strongly by the time elapsed during the formation of the fission products. If the fission products are formed during an interval

of a few seconds, the gross activity will fall off rapidly with time, but if the fission products are formed over a longer interval, the rate of gross decay will be smaller, because the short-lived nuclides will have decayed to some extent during the period of reactor operation. In the example studied in this paper, the length of reactor operation was 42 days.

In the work upon which this paper is based, the time-dependent decay characteristics of each individual fission product were examined, and these effects were then added together to obtain the total effects of gamma and beta activities reported here. Much less time-consuming methods, such as that of Way and Wigner[10], of approximate nature, are available for the estimation of the activity of gross fission products as a function of the time elapsed since reactor shutdown and of the duration of reactor operation. The results of these calculations have checked fairly closely with a Way-Wigner approximation for periods of 1 to 16 days following shutdown for a reactor operating on a 42-day cycle.

The calculations reported here are a summary of the mass and the activity of each fission product nuclide present in the spent fuel of a 500 megawatt (heat) reactor operating on a 42-day cycle, at the time of removal from the reactor and as a function of time thereafter. These quantities have been expressed in terms of grams and curies per 10 kilograms of fissioned material, respectively. The calculations represent some modification and extension of the work of Moteff[7] and Engelder[2], and relied also upon the work of Coryell et al.[1], Engelkemeir et al.[3], Glendenin et al.[4], Katcoff et al.[5], and Wiles et al.[11]. The major modifications are summarized in Table I.

The effect of the calculated activities, in terms of roentgens per hour at one meter from a point source, and B.T.U's per hour of heat generated

Table I

Modifications and Extensions of Engelder's Work

Incorporated in This Report

Modification	Engelder	Nehemias
Minimum fission yield	1%	0.01%
Minimum half life	5 hr	1 hr
Reactor operation time	1000 hr	42 d
Steady state power	550 Mw	500 Mw
Time after shutdown	1 d	0,1,2,4, 8,16 d
Calculation of decay chains	none past 2nd generation	all chains complete

from beta particle absorption, has also been calculated. Because of its dependence upon absorber material, absorber thickness and geometry, calculation of heat generation from gamma absorption has been omitted. The several calculations discussed above appear in a summary in Fig. 1.

The calculation of the number of grams and curies present is a separate problem for each nuclide. To each mass number corresponds a decay chain, although in some cases the decay chain consists merely of a single stable nuclide. It will be assumed that the first nuclide in each decay chain is formed at the total yield rate for that mass number, that its daughter products are formed only through the radioactive decay of the parent nuclide.

Many of these results can be obtained by various methods. In these calculations, a particular format has been selected and maintained throughout for ease of calculation.

ASSUMPTIONS

Reactor operation time, T = 42 days.

Steady state reactor power = 500 Mw. (heat)

Time after removal from reactor, t = 0, 1, 2, 4, 8, 16 days.

Radioactive decay schemes taken from "Nuclear Data", NBS Circular - 499[9], or reasonable values assumed when no data is available.

Fission yields greater than 0.01% are used.

Species with half lives less than one hour which have precursors with half lives less than one hour are not considered.

Fission yields for fast fission of U-235 are not well known. The curve of fast fission yields for U-235, estimated by Engelder, has been used

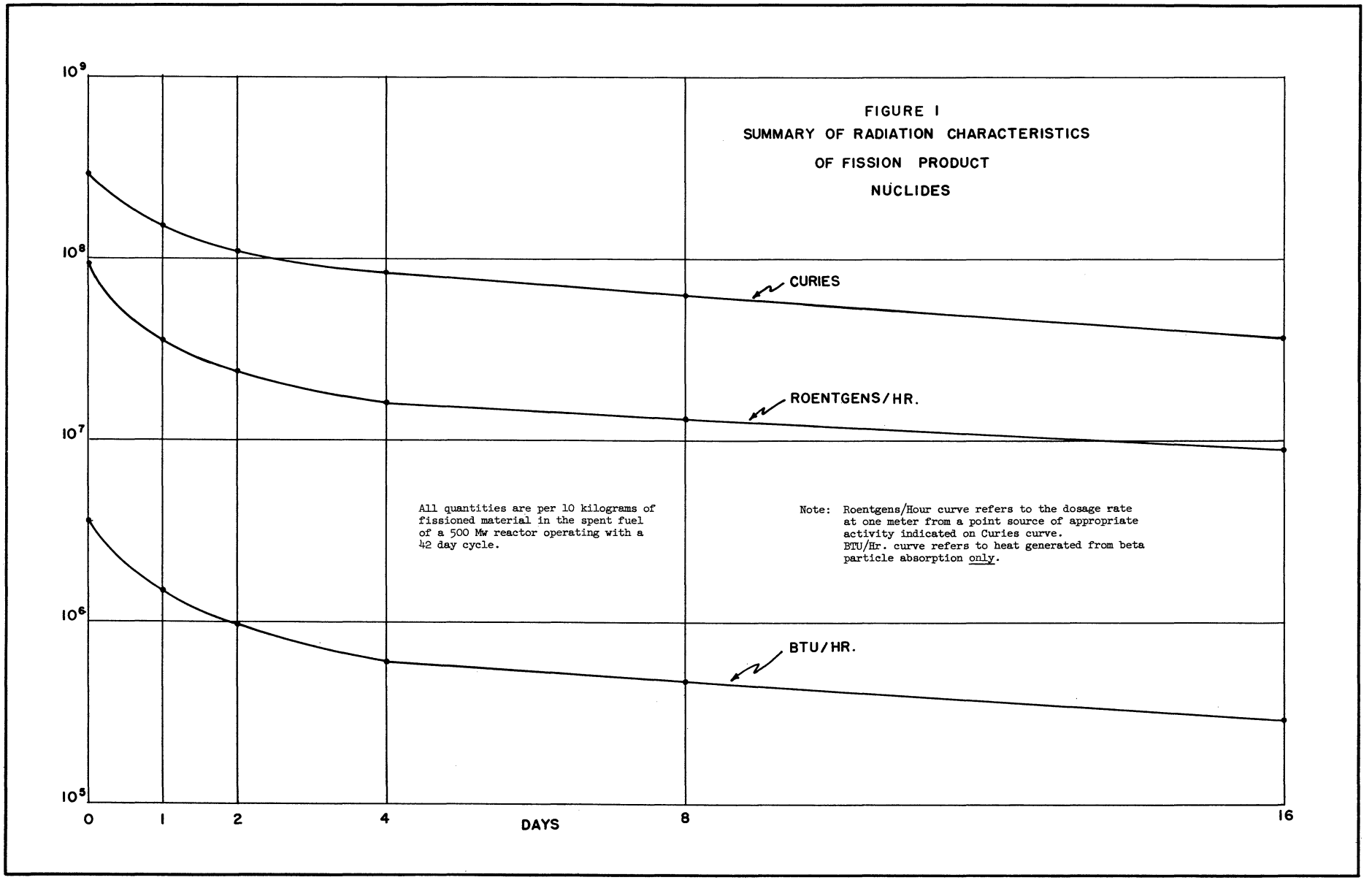
FIGURE I
SUMMARY OF RADIATION CHARACTERISTICS
OF FISSION PRODUCT
NUCLIDES
10^9
10^8
10^7
10^6
10^5
CURIES
ROENTGENS/HR.
BTU/HR.
All quantities are per 10 kilograms of fissioned material in the spent fuel of a 500 Mw reactor operating with a 42 day cycle.
Note: Roentgens/Hour curve refers to the dosage rate at one meter from a point source of appropriate activity indicated on Curies curve.
BTU/Hr. curve refers to heat generated from beta particle absorption only.
0
1
2
4
8
16
DAYS

in these calculations (see Fig. 2). Modification of the results may be undertaken when better, more detailed data become available. The calculations presented here are somewhat more sensitive to such modifications than Engelder's because yields down to 0.01% are used. Thus, yield values are assumed throughout the highly conjectural central trough region.

Neutron capture by fission products while residing in the reactor has not been considered because of the extreme paucity of cross section data at the neutron energies involved.

Secondary processes, such as the low energy spectrum of bremstrahlung produced during the absorption of beta particles in matter, and the production of internal conversion and secondary electrons during the absorption of gamma radiation, have not been taken into account. In calculations of detailed shielding requirements, these effects should be considered as they become necessary.

SAMPLE CALCULATION

For every mass number, including those which yield only stable nuclides, the rate of formation in grams per second and the total yield in grams have been calculated. To calculate the rate of formation for each mass number one uses the relationship:

$$\text{grams/second} = \frac{yWfA}{N},$$

where,

y = fission yield,

W = reactor power level in watts,

f = fission rate, in fissions per watt-second ($f = 3.25 \times 10^{10}$, assuming 192 Mev/fission),

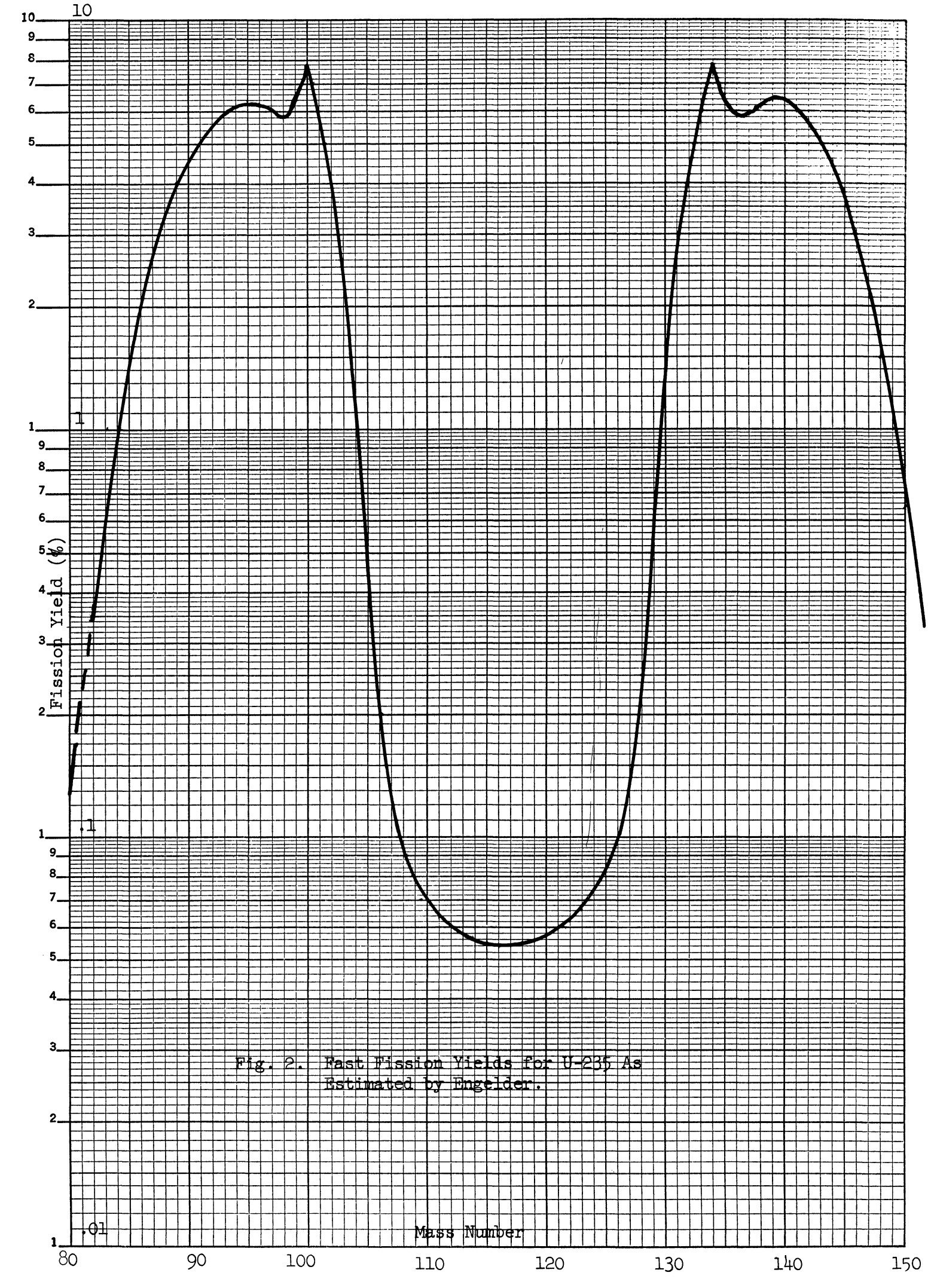

Fig. 2. Fast Fission Yields for U-235 As Estimated by Engelder.

A = atomic number,

N = Avogadro's number, 6.023×10^{23}.

For mass number 127,

$$\text{grams/second} = \frac{0.0015(5\times10^{8})(3.25\times10^{10})127}{6.023\times10^{23}} = 0.0515\times10^{-4}$$

The total number of grams of mass number 127 produced in the reactor core after 42 days of steady state operation at 500 Mw is simply:

$$= 0.0515\times10^{-4}(42)(24)(3600)$$

$$= 18.5$$

This quantity and those that follow in this sample calculation may be expressed as quantity present per 10 kilograms of fissioned material by dividing by 2.31, since total mass of fissioned material after 42 days

$$= \frac{(5\times10^{8})(3.25\times10^{10})(235)(42)(24)(3600)}{6.023\times10^{23}}$$

$$= 23{,}100 \text{ grams.}$$

The decay chain for mass number 127 is shown in Fig. 3.

At the end of 42 days of steady state reactor operation, the buildup of this parent nuclide (Sb-127) approaches saturation; i.e., the rate of decay has increased to approximately equal the rate of formation. The fraction of saturation for a parent nuclide after T days of reactor operation is (Moteff)[7]:

$$F_1 = (1-e^{-\lambda_1 T}),$$

where λ_1 is the decay constant in reciprocal days and T is the reactor operation time in days.

For Sb-127 at 42 days,

$$F_1 = (1-e^{-0.179(42)}),$$

$$= (1-0.00054),$$

$$= 0.99946$$

$$\cong 1.$$

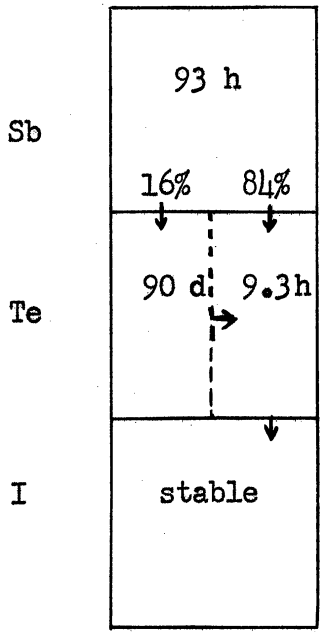

Fig. 3. Decay Chain for Mass Number 127

The activity at saturation is calculated according to the relationship

$$\text{curies} = \frac{yWf}{3.7\times10^{10}},$$

where,

y = fission yield

W = reactor power level

and

f = fission rate, in fission/watt-second

= $(3.25\times10^{10}$ assuming 192 Mev/fission)

For mass number 127,

$$\text{curies} = \frac{0.0015(5\times10^{8})(3.25\times10^{10})}{3.7\times10^{10}}$$

$= 0.638\times10^{6}$ curies in the entire reactor core after 42 days of operation.

The decay rate is directly proportional to the amount of material, M, present; i.e.,

$$\frac{dM}{dt} = -\lambda M.$$

As M is desired in grams, and dM/dt has been computed in grams per second (at saturation),

$$M(\text{grams}) = \frac{\text{grams/second}}{\lambda}$$

$$= 0.015\times10^{-4}\ \frac{3600(93)}{0.693},$$

$$= 2.46.$$

The fraction of saturation for a daughter nuclide after T days of operation is (Moteff)

$$F_2 = \frac{\lambda_1\ \lambda_2}{\lambda_1 - \lambda_2}\left[\frac{1-e^{-\lambda_2 T}}{\lambda_2} - \frac{1-e^{-\lambda_1 T}}{\lambda_1}\right].$$

For 90-day Te-127,

$$F_2 = \frac{0.179(0.0077)}{0.179-0.0077}\left[\frac{1-e^{-0.0077(42)}}{0.0077} - \frac{1-e^{-0.179(42)}}{0.179}\right],$$

$$= 0.263.$$

The number of curies of 90-day Te-127 present after 42 days of reactor operation is thus

$$= 0.638\times10^{6}\ (0.263)0.16 = 0.027\times10^{6} \text{ curies},$$

and, as before, the number of grams

$$= \frac{0.0515\times10^{-4}\ (90)24(3600)0.263(0.16)}{0.693}$$

$$= 2.43 \text{ grams}.$$

A total of $(18.5-2.46)(0.16) = 2.56$ grams of the 90-day isomer of Te-127 has been formed. Of this, only 0.13 grams has decayed to the 9.3 hour isomer in the entire 42-day period. On this basis, one may consider that essentially the only source of the 9.3-hour isomer is direct decay of Sb-127. From this, calculating as before for a daughter nuclide,

$$F_2 \cong 1,$$

$$\text{curies} = 0.638\times10^{6}\ (0.84) = 0.546\times10^{6}$$

and,

$$\text{grams} = \frac{0.0515\times10^{-4}\ (9.3)3600(0.84)}{0.693} = 0.206$$

The total number of grams formed at mass number 127 has been computed (18.5 grams). The mass of each of the radioactive nuclides has also been computed at the time of removal from the reactor. It follows that the mass of stable I-127 at 42 days is:

$$18.5-(2.46+2.43+0.206) = 13.4 \text{ grams}$$

The fold-in chart (Fig. 4) presents the results of these calculations of mass and activity level for each nuclide, per 10 kilograms of fissioned material, at the time of removal from the reactor.

GAMMA RADIATION LEVELS

Marinelli, et al.[6], have published a curve of roentgens per hour per millicurie point source at one centimeter distance as a function of photon energy as shown in Fig. 5. This assumes one photon, at the energy in question, per disintegration. If the disintegration scheme is known, the radiation level at a particular position may be calculated.

The disintegration scheme used for mass number 127 is shown in Fig. 6.

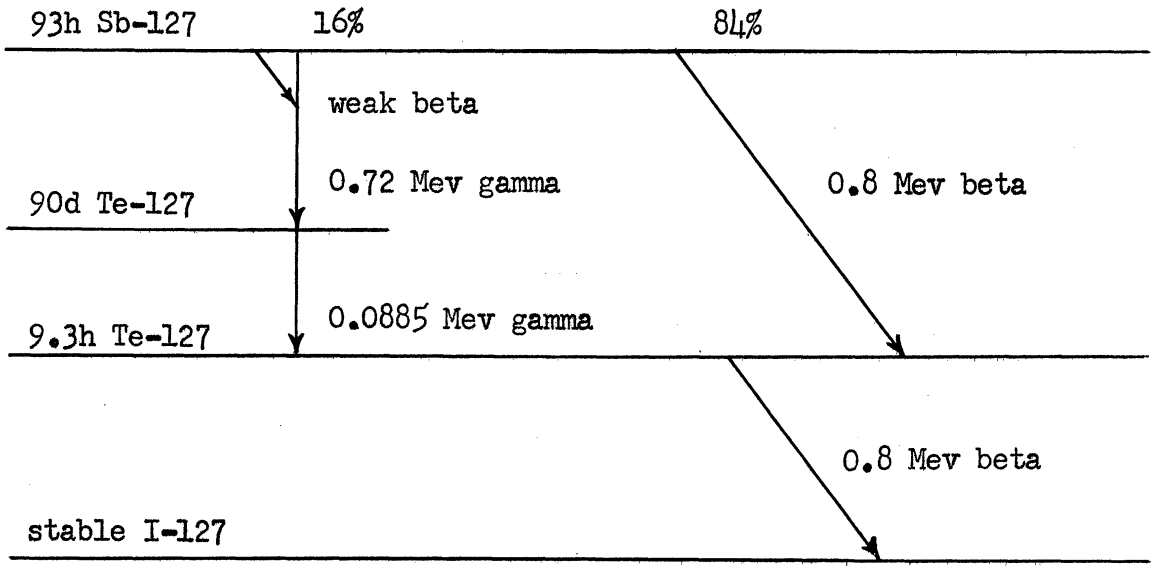

Fig. 6. Disintegration Scheme for Mass Number 127.

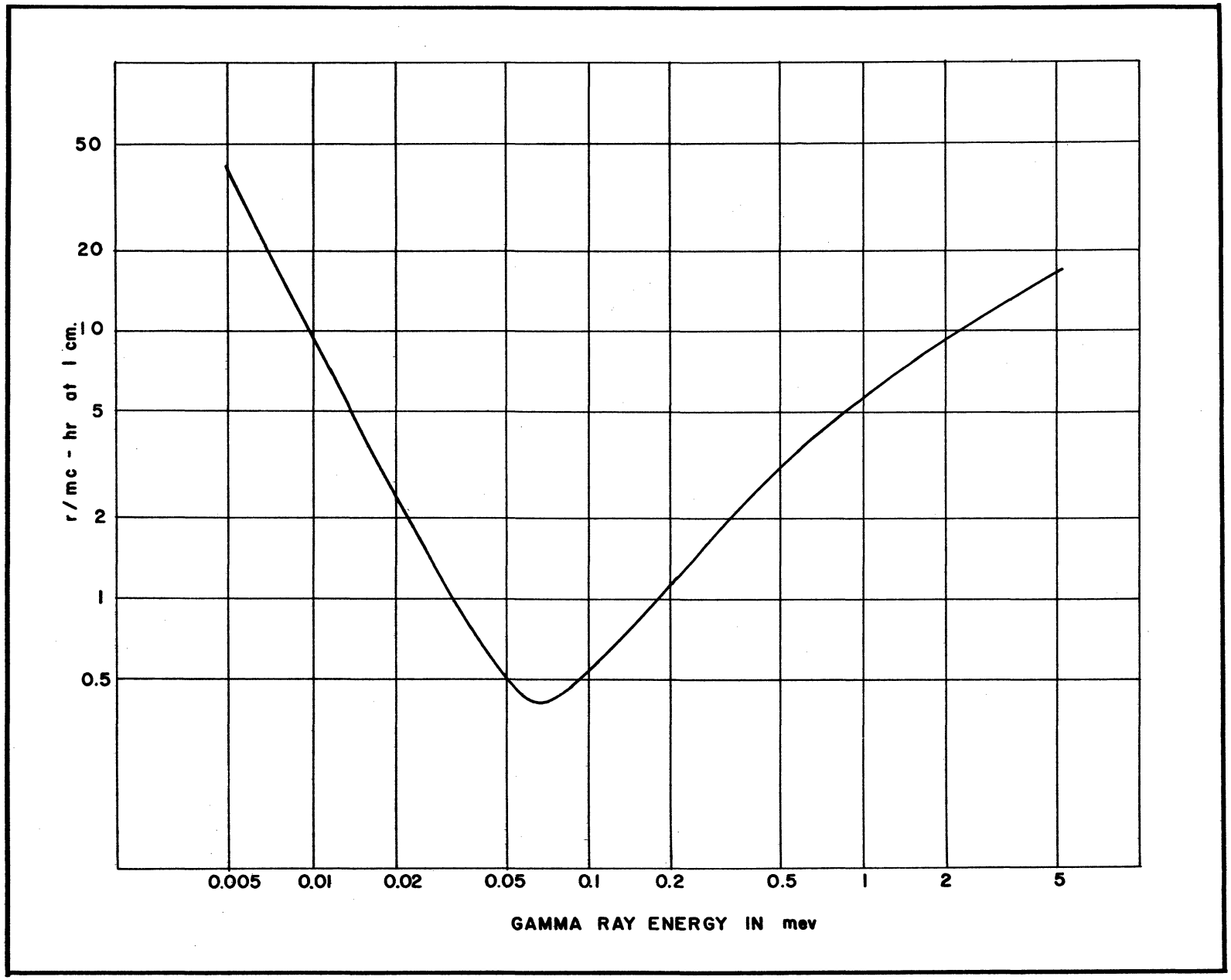

Fig. 5. Roentgens per hour per millicurie at one centimeter from a point gamma emitter as a function of photon energy, assuming one photon per disintegration.

In the decay of Sb-127, there are 0.16 photons per disintegration of 0.72 Mev energy. As shown in Fig. 5, such a photon yields 0.42 roentgens per hour per curie at one meter. Therefore, for Sb-127,

$$\frac{(0.16)0.638\times10^{6}\ (0.42)}{2.31} = 1.85\times10^{4}$$

roentgens per hour at one meter for 10 kilograms of fissioned material. Similarly, for 90-day Te-127,

$$\frac{0.027\times10^{6}\ (0.045)}{2.31} = 5.26\times10^{2}\ .$$

BETA PARTICLE ABSORPTION

As discussed by Engelder[2], it is probable that beta particles emitted by fission product nuclides within a typical piece of equipment will be totally absorbed; therefore, all the emitted energy would be absorbed within the equipment as thermal energy.

In the decay of Sb-127, there are 0.84 beta particles per disintegration with a maximum energy of 0.8 Mev. A satisfactory conversion factor from maximum energy to average energy over the range of energies involved is 0.4. The conversion factor from Mev to BTU is 1.517×10^{-16}, and 3600 (3.7×10^{10}) converts curies into particles per hour. Thus, for Sb-127,

$$\frac{(1.517\times10^{-16})3600(3.7\times10^{10})0.84(0.8)0.4(0.638\times10^{6})}{2.31} = 1.50\times10^{3}$$

BTU per hour per 10 kilograms of fissioned material.

Similarly for 9.3-hour Te-127,

$$\frac{(1.517\times10^{-16})3600(3.7\times10^{10})0.8(0.4)0.546\times10^{6}}{2.31} = 1.52\times10^{3}$$

DECAY CHARACTERISTICS

The number of grams and curies of each nuclide, and the resulting rates of beta energy absorption and gamma radiation levels have been calculated at the time of removal from the reactor, for each mass number, according to the processes just outlined. It is now of interest to evaluate the variation in these quantities with time after removal from the reactor. For purposes of this calculation, the various nuclides were partitioned into three groups: first generation (parent) nuclides, second generation (daughter) nuclides, and third generation (grand-daughter) nuclides.

Rutherford[8] has developed explicit expressions directly applicable to this problem. As stated by Rutherford, the situation is as follows: ". . . a primary source has supplied (a particular) matter A at a constant rate for any time T and is then suddenly removed. Required the amount of A, B, C at any subsequent time," the following nomenclature is pertinent:

1) B and C are daughter and grand-daughter of A, respectively.
2) λ_1, λ_2, λ_3 are the decay constants of A, B, C respectively.
3) T is the operation time of the reactor, in this case, 42 days.
4) t is the independent variable, time.
5) P_T, P; Q_T, Q; R_T, R refer to the amounts of A, B, and C present at time of shutdown, t = 0, and after an arbitrary time t, respectively.

The ratios P/P_T, Q/Q_T, R/R_T, i.e., the ratios of the amount present at time t after removal from the reactor to the amount present at shutdown, are given by Equations 1, 2, and 3, respectively. These ratios shall henceforth be referred to as survival ratios.

$$1)\quad P/P_T = e^{-\lambda_1 t};$$

$$2)\quad Q/Q_T = \frac{ae^{-\lambda_2 t} - be^{-\lambda_1 t}}{a-b}$$

where

$$a = \frac{1-e^{-\lambda_2 T}}{\lambda_2}$$

$$b = \frac{1-e^{-\lambda_1 T}}{\lambda_1}$$

and,

$$3)\quad R/R_T = \frac{fe^{-\lambda_1 t} + ge^{-\lambda_2 t} + he^{-\lambda_3 t}}{f+g+h}$$

where

$$f = \frac{\lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}\ (1-e^{-\lambda_1 T}),$$

$$g = \frac{\lambda_1}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)}\ (1-e^{-\lambda_2 T}),$$

$$h = \frac{\lambda_1 \lambda_2}{\lambda_3(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}\ (1-e^{-\lambda_3 T}).$$

USE OF ANALOG COMPUTER

These three cases can be easily solved by means of an electronic differential analyzer. In each case, the equivalent system of simultaneous ordinary differential equations can be formulated as follows.

Case I:

$$\frac{dP}{dt} = -\lambda_1 P,$$

with the condition,

$$P(o) = P_T .$$

Case II:

$$\frac{dP}{dt} = -\lambda_1 P$$

$$\frac{dQ}{dt} = -\lambda_2 + \lambda_1 P,$$

with the conditions,

$$P(o) = P_T, \qquad Q(o) = Q_T .$$

Case III:

$$\frac{dP}{dt} = -\lambda_1 P,$$

$$\frac{dQ}{dt} = -\lambda_2 Q + \lambda_1 P,$$

$$\frac{dR}{dt} = -\lambda_3 R + \lambda_2 Q,$$

with the conditions,

$$P(o) = P_T, \quad Q(o) = Q_T, \quad R(o) = R_T .$$

Choosing suitable transformations, the machine equations corresponding to these three cases can be programmed as shown in Fig. 7. Here first-generation cases are performed with the topmost circuit; second-generation with the simultaneous solution of the top and middle circuits; while third-generation cases require all three. By judicious planning, many of the first-generation cases can be solved with the same computer setup used for some daughter

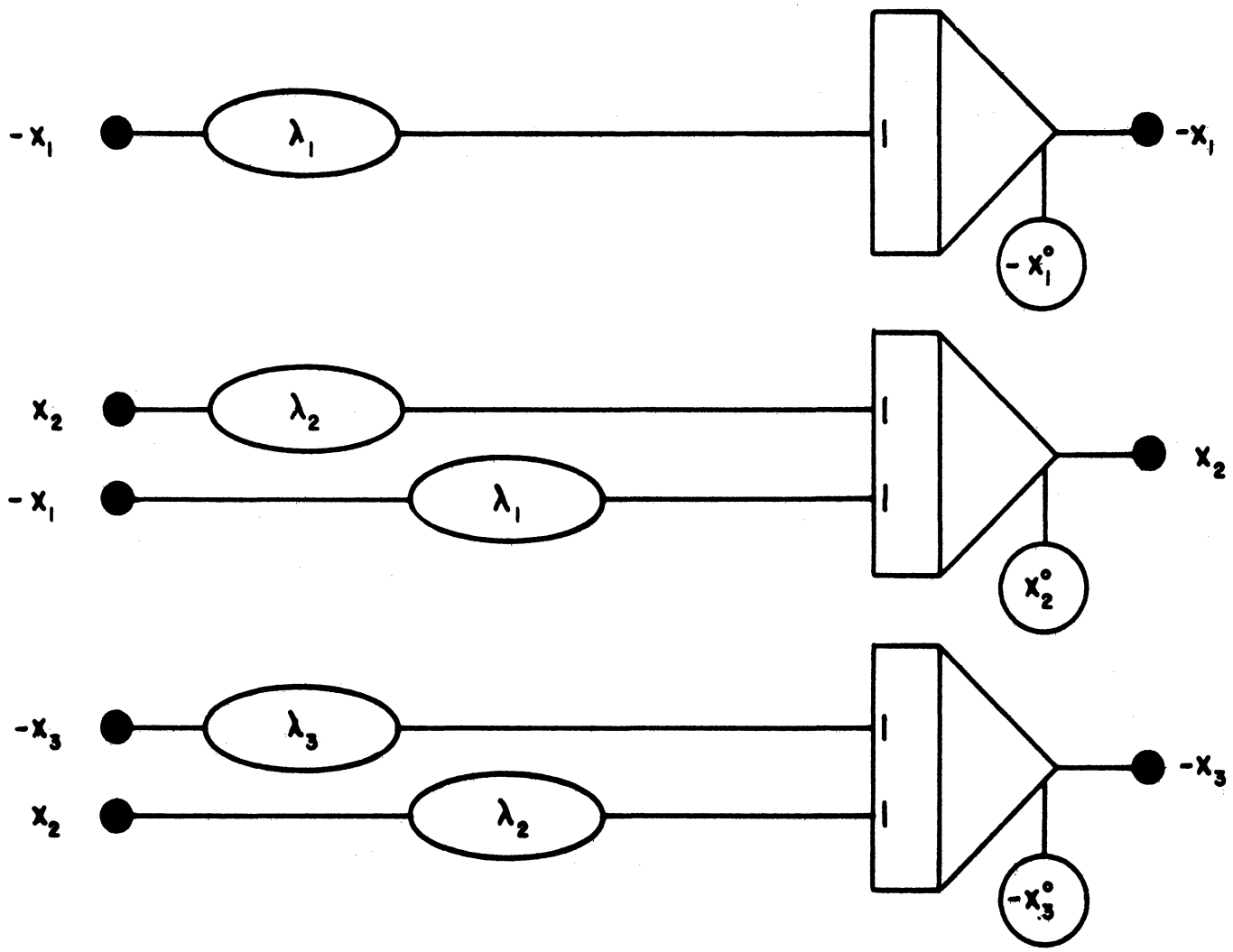

FIG. 7 SCHEMATIC DIAGRAM
OF NECESSARY COMPUTER PROGRAMS

calculations. By feeding the output of the computer directly to an x-y plotting board results can be read off the resulting graphs at any future time for any particular time after reactor shutdown.

In the case of the mass number 127, both isomers of Te-127 may be considered daughter products, as previously discussed. The survival ratios for the three radioactive members of the sample decay chain for 0, 1, 2, 4, 8, and 16 days after shutdown are presented in Table II. These multiplicative factors are then applied to the four quantities of interest as calculated at time of shutdown. The mass of the stable nuclide increases directly with the loss of mass of the radioactive members of the chain. The resultant information for mass number 127 is presented in Table III. Table IV contains these data for all the mass numbers.

Table II

Survival Ratios for Mass Number 127

The ratios of quantity present at a given time after shutdown to quantity present initially are tabulated for each radioactive nuclide (including isomers) of mass number 127, for t = 0, 1, 2, 4, 8, 16 days after shutdown.

Days after shutdown	Sb 127	90 d Te 127	9.3 h Te 127
0	1	1	1
1	0.836	1.062	0.905
2	0.700	1.062	0.759
4	0.489	1.110	0.538
8	0.249	1.128	0.261
16	0.057	1.099	0.063

Table III

Calculated Mass, Activity, Radiation Level and Rate
of Heat Formation of Each Nuclide
of Mass Number 127 for
t = 0, 1, 2, 4, 8, 16 days
after Shutdown

The first column, G, is mass in grams. The second column, C, is the sum of beta and gamma activity in curies. The third column, R, is radiation level from gamma emission only, in Roentgens per hour at one meter from a point source of the particular curie level and energy. The fourth column, W, is the rate of formation of heat from beta particle absorption only, in BTU's per hour. All quantities are expressed per 10 kilograms of fissioned material.

Table III

G	C	R	W
		Sb	
1.04	2.76×10^{5}	1.85×10^{4}	1.50×10^{3}
0.89	2.31×10^{5}	1.55×10^{4}	1.25×10^{3}
0.74	1.93×10^{5}	1.30×10^{4}	1.05×10^{3}
0.51	1.35×10^{5}	9.07×10^{3}	7.33×10^{2}
0.25	6.60×10^{4}	4.44×10^{3}	3.58×10^{2}
0.06	1.57×10^{4}	1.05×10^{3}	8.52×10
		Te	
1.55	2.48×10^{5}	5.26×10^{2}	1.52×10^{3}
1.20	2.26×10^{5}	5.56×10^{2}	1.38×10^{3}
1.19	1.93×10^{5}	5.56×10^{2}	1.16×10^{3}
1.22	1.43×10^{5}	5.76×10^{2}	8.18×10^{2}
1.21	8.09×10^{4}	5.93×10^{2}	1.80×10^{2}
1.16	3.52×10^{4}	5.77×10^{2}	4.30
		I	
5.80	-	-	-
6.30	-	-	-
6.46	-	-	-
6.66	-	-	-
6.93	-	-	-
7.17	-	-	-

Table IV

Calculated Mass, Activity, Radiation Level and Rate
of Heat Formation of Fission Product
Nuclides for
t = 0, 1, 2, 4, 8, 16 days
after Shutdown

The columns G, C, R, W have the same significance as corresponding columns in Table III, with the exception of G, which is in kilograms. All quantities are expressed per 10 kilograms of fissioned material.

Table IV

G	C	R	W
	GRAND TOTAL		
9.42	2.99×10^{8}	9.46×10^{7}	3.79×10^{6}
9.42	1.46×10^{8}	3.52×10^{7}	1.58×10^{6}
9.43	1.20×10^{8}	2.48×10^{7}	9.26×10^{5}
9.42	8.78×10^{7}	1.67×10^{7}	6.19×10^{5}
9.25	6.28×10^{7}	1.31×10^{7}	4.32×10^{5}
9.44	4.37×10^{7}	9.21×10^{7}	2.99×10^{5}

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