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COLLEGE OF ENGINEERING
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NUCLEAR ENGINEERING

ENGINEERING APPLICATIONS OF NUCLEAR ENERGY

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PART I - NUCLEAR PHYSICS

1. THE ATOMIC NUCLEUS

A. Introduction

The atom is often described as the smallest amount of material that has a definite chemical identity. An atom consists of a positively charged central core, the nucleus, and a surrounding cloud of orbital electrons. The number of orbital electrons is equal to the number of positive charges on the nucleus so that an atom is electrically neutral. The electrons may occupy a series of different orbits of gradually increasing average radius but once an orbit contains two electrons, no more can be accepted. The chemical properties of an element are determined by the electrons in the outermost orbits. Most all atoms are about the same size with diameters slightly greater than about 10^{-8} cm.

The atomic nucleus is small and very dense. Nuclear radii are all roughly 10^{-13} cm, the heavy nuclei being somewhat larger. About 0.02% of the mass of an atom is due to the mass of the orbital electrons, the remainder being concentrated in the nucleus. The nucleus is believed to consist of Z protons each with unit positive charge, and N neutrons which are neutral particles. The number Z is the atomic number. The chemical properties of an element are determined by Z and so the chemical periodic table is built up with Z as an index number. The number of neutrons N does not influence chemical behavior, but does influence nuclear properties considerably. Different nuclei or nuclides* having the same Z but different N are

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* A nuclide is a particular nuclear species with given values of Z and mass number A = N + Z. Nuclei refer to more than one nucleus and may include a number of nuclides. Where no confusion is likely, nuclei and nuclides are used synonomously in the literature.

called isotopes. The mass number of a nucleus A, is the total number of neutrons and protons. Nuclides having the same A but different Z are called isobars.

Neutrons and protons have very nearly equal masses of about 1.67×10^{-24} grams, the neutron being heavier than the proton by about 0.1 percent. The mass of a nucleus is slightly less than Z x (mass of a proton) + N x (mass of a neutron). The difference or mass defect, being about 0.8 percent of the total mass for most nuclides. A more detailed description of the mass defect is presented in Part C of this section.

B. Chart of the Nuclides

The chart of the nuclides is a plot of Z vs N (see Figure 1). This chart shows both stable and radioactive nuclides and gives information about the modes of decay. Excellent wall size copies of the chart may be obtained from the Knolls Atomic Power Laboratory, Schenectady, New York.

The stable nuclides are closely clustered about a curved line running across the chart. The radioactive nuclides generally fall at some distance on either side of this line, and the schemes by means of which they decay into stable nuclides may be traced on the chart.

A shorthand notation has been developed to specify particular nuclides. The chemical symbol for the element, the atomic number Z, and the mass number A are written in the following combination:

$$_{Z}(\text{chem. symbol})^{A}$$

For example, $_{82}Pb^{206}$ means the lead nucleus with a mass number of 206. All lead isotopes have 82 protons. The number of neutrons in $_{82}Pb^{206}$ is A-Z = 124. Often the value of Z is omitted in this notation as the chemical symbol fixes Z, and we see Pb^{206}, Pb-206, and lead-206 in the literature.

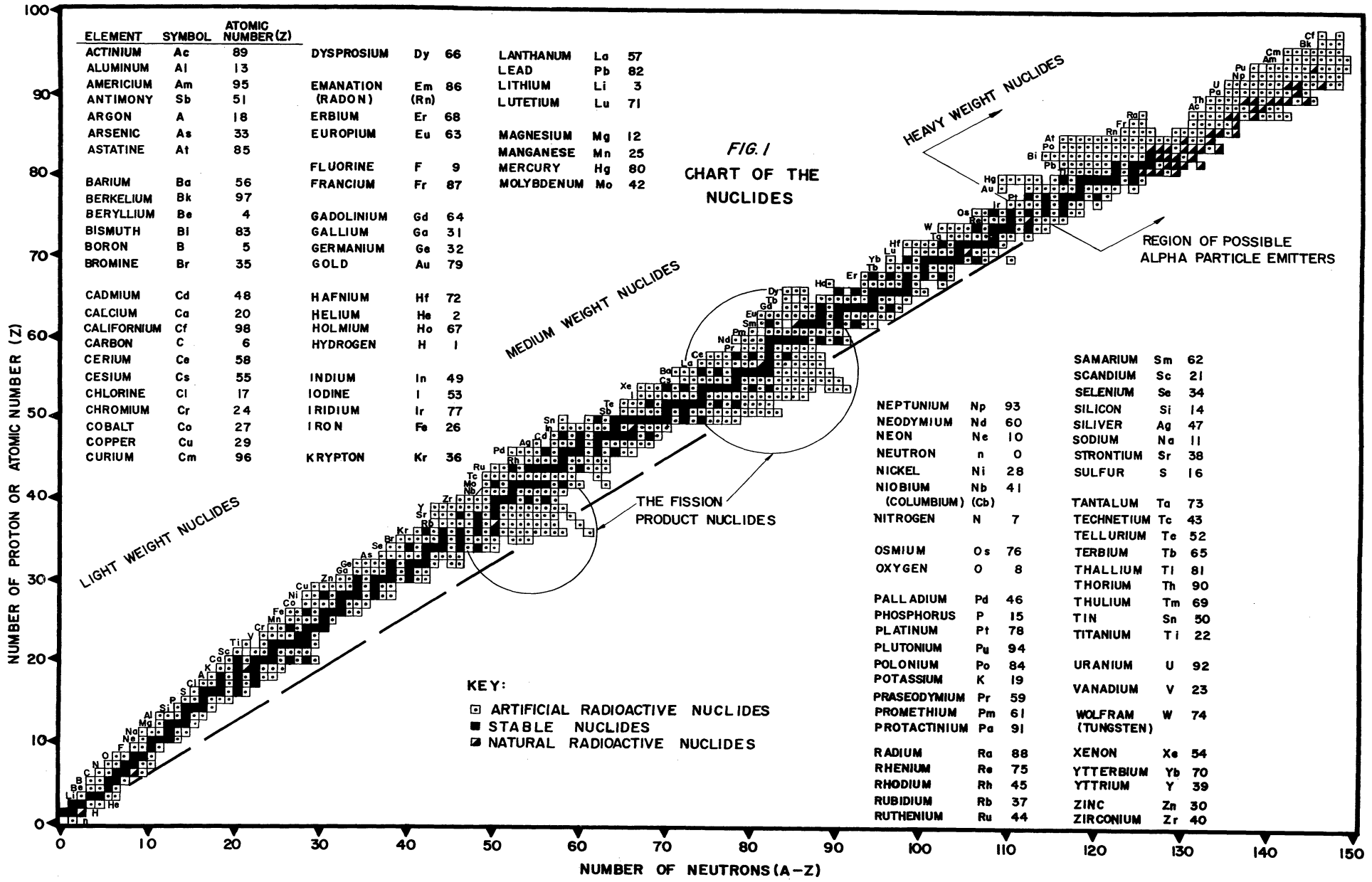

FIG. 1

CHART OF THE NUCLIDES

ELEMENT	SYMBOL	ATOMIC NUMBER (Z)
ACTINIUM	Ac	89
ALUMINUM	Al	13
AMERICIUM	Am	95
ANTIMONY	Sb	51
ARGON	A	18
ARSENIC	As	33
ASTATINE	At	85
BARIUM	Ba	56
BERKELIUM	Bk	97
BERYLLIUM	Be	4
BISMUTH	Bi	83
BORON	B	5
BROMINE	Br	35
CADMIUM	Cd	48
CALCIUM	Ca	20
CALIFORNIUM	Cf	98
CARBON	C	6
CERIUM	Ce	58
CESIUM	Cs	55
CHLORINE	Cl	17
CHROMIUM	Cr	24
COBALT	Co	27
COPPER	Cu	29
CURIUM	Cm	96
DYSPROSIUM	Dy	66
EMANATION (RADON)	Em (Rn)	86
ERBIUM	Er	68
EUROPIUM	Eu	63
FLUORINE	F	9
FRANCIUM	Fr	87
GADOLINIUM	Gd	64
GALLIUM	Ga	31
GERMANIUM	Ge	32
GOLD	Au	79
HAFNIUM	Hf	72
HELIUM	He	2
HOLMIUM	Ho	67
HYDROGEN	H	1
INDIUM	In	49
IODINE	I	53
IRIDIUM	Ir	77
IRON	Fe	26
KRYPTON	Kr	36
LANTHANUM	La	57
LEAD	Pb	82
LITHIUM	Li	3
LUTETIUM	Lu	71
MAGNESIUM	Mg	12
MANGANESE	Mn	25
MERCURY	Hg	80
MOLYBDENUM	Mo	42
NEPTUNIUM	Np	93
NEODYMIUM	Nd	60
NEON	Ne	10
NEUTRON	n	0
NICKEL	Ni	28
NIOBIUM (COLUMBIUM)	Nb (Cb)	41
NITROGEN	N	7
OSMIUM	Os	76
OXYGEN	O	8
PALLADIUM	Pd	46
PHOSPHORUS	P	15
PLATINUM	Pt	78
PLUTONIUM	Pu	94
POLONIUM	Po	84
POTASSIUM	K	19
PRASEODYMIUM	Pr	59
PROMETHIUM	Pm	61
PROTACTINIUM	Pa	91
RADIUM	Ra	88
RHENIUM	Re	75
RHODIUM	Rh	45
RUBIDIUM	Rb	37
RUTHENIUM	Ru	44
SAMARIUM	Sm	62
SCANDIUM	Sc	21
SELENIUM	Se	34
SILICON	Si	14
SILIVER	Ag	47
SODIUM	Na	11
STRONTIUM	Sr	38
SULFUR	S	16
TANTALUM	Ta	73
TECHNETIUM	Tc	43
TELLURIUM	Te	52
TERBIUM	Tb	65
THALLIUM	Tl	81
THORIUM	Th	90
THULIUM	Tm	69
TIN	Sn	50
TITANIUM	Ti	22
URANIUM	U	92
VANADIUM	V	23
WOLFRAM (TUNGSTEN)	W	74
XENON	Xe	54
YTTERBIUM	Yb	70
YTTRIUM	Y	39
ZINC	Zn	30
ZIRCONIUM	Zr	40

C. The Binding Energy Curve

When two or more nucleons (nucleon is a generic term which includes both neutrons and protons) are in close proximity, there is a not too well understood short range force between them which will cause them to form a cluster. It is an energetically favorable situation to have nucleons so clustered as a nucleus. The amount of energy needed to break a nucleus into neutrons and protons infinitely separated from one another is called the binding energy for that nucleus. The binding energy ΔE can be measured by noting that by the Einstein relation between mass and energy, we expect a nucleus to weigh less than its constituent nucleons by an amount $\Delta M = \Delta E/C^2$. Measurement of nuclear masses by means of a mass spectograph* provides a measurement of the mass defect and consequently, the binding energy. Binding energies are usually stated in Mev, (million electron volts) although other units of energy may be used.** The conversion tables on page 113 permit conversion to B.T.U., kilowatt-hours, etc.

The binding energy ΔE is very closely proportional to the mass number A, thus we find there is a nearly constant value of the binding energy per nucleon ($\Delta E/A$). The deviations from proportionality show up best in a plot of ($\Delta E/A$) vs. A (see Figure 2).

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*The mass spectrograph is an instrument which measures the mass of a charged particle, in this case an ion of the element whose mass is to be determined. The ion is given a definite velocity by accelerating it with a known electric potential and then allowed to pass through a region where an intense static magnetic field has been set up. The ion follows a circular path in the magnetic field. The radius of the path is different for ions of different masses and so measurement of the path radius provides a measurement of the mass of the ion.

** The energy unit Mev is the kinetic energy which an electron has after falling through an electrical potential of one million volts. One Mev may be seen to be equivalent to 1.6×10^{-12} ergs. The Mev unit of energy is not restricted in use to describe electrons.

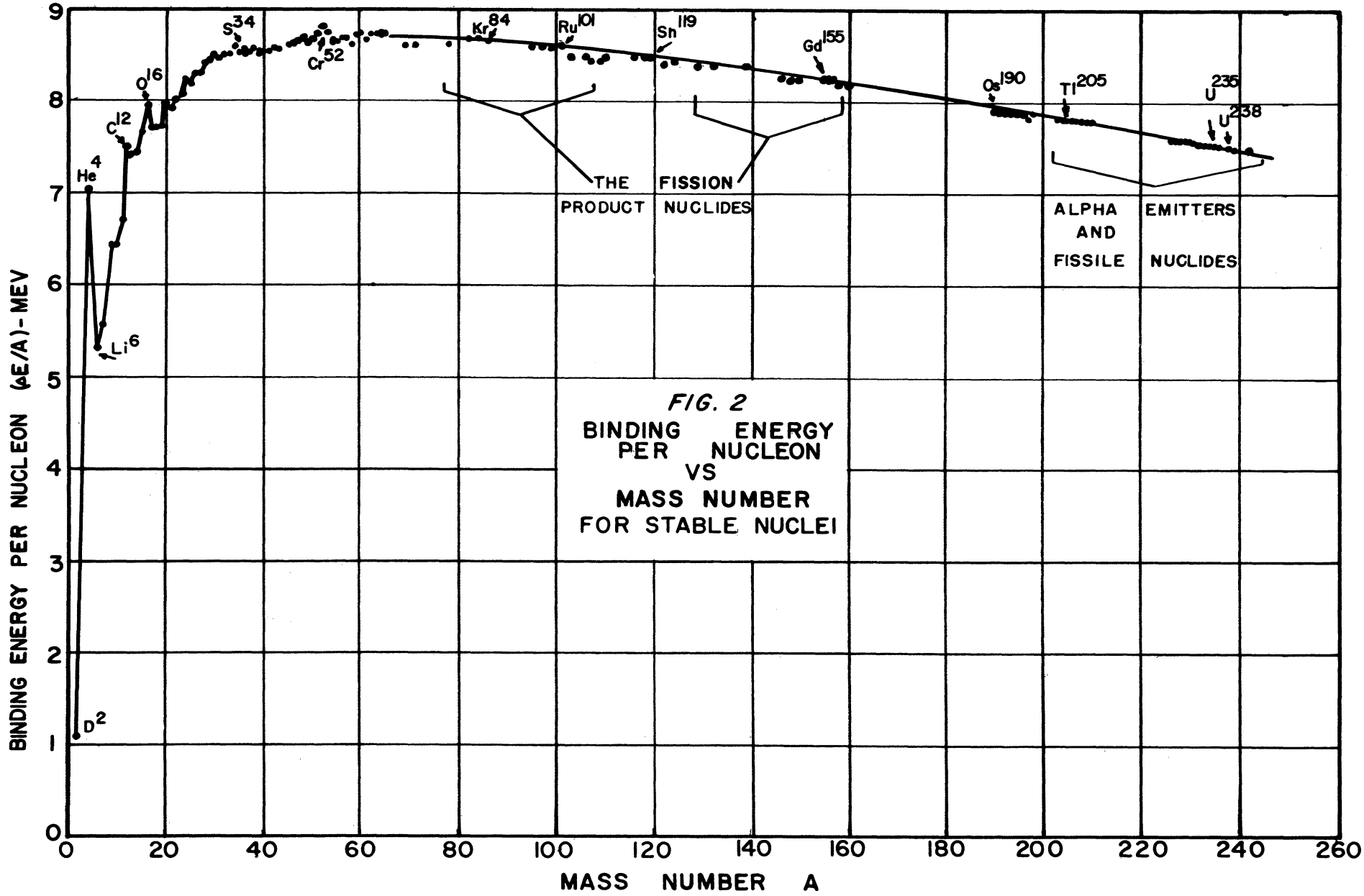

FIG. 2

BINDING ENERGY PER NUCLEON VS MASS NUMBER FOR STABLE NUCLEI

This curve is important to nuclear power engineering in that it shows how much energy may be obtained from a heavy nucleus like uranium by its being broken into two or more fragments by the fission process. The energy available from each fission is given by: (sum of binding energies of fission fragments) - (binding energy of the initial uranium nucleus). For uranium, this amounts to about 200 Mev for each fission.

A little consideration shows that the most stable nuclei are those with the maximum value of $(\Delta E/A)$ and hence in the middle of the curve of Figure 2. It is energetically favorable for some light weight nuclei to combine to form a medium weight nucleus by the fusion process and it is also energetically favorable for the very heavy nuclei to break up into medium weight fragments. The alpha particle or ${}_2He^4$ nucleus is a light weight nucleus of high stability as may be seen from Figure 2. This is important in the theory of alpha particle emission by heavy nuclei and will be discussed in the next Section. The high binding energy of the alpha particle is also the basis of the explanation of the energy output of the sun where it is believed that the initial reactants are hydrogen and light nuclei and the products are helium and other light nuclei. The overall process being the conversion of hydrogen to helium with the release of about 28 Mev for each helium nucleus produced or 0.75 megawatt-hours for each mole of helium produced.

Published values of measured nuclear masses are expressed in atomic mass units, (amu). This unit has been chosen such that oxygen - 16 has a mass on this scale of 16.000. The atomic mass unit may be shown to be 1.660×10^{-24} grams. The chemical mass scale is very nearly the same as this but uses naturally occurring oxygen as a basis instead of oxygen 16.

The conversion tables give the conversion factor between these mass scales.

2. RADIOACTIVITY

A radioactive nucleus is one that is energetically unstable and if given sufficient time will decay to a more stable state by the emission of either a particle, a gamma ray, or both. The probability of the decay of a particular radioactive nucleus is independent of the histories of all other radioactive nuclei in a sample of material. This leads to the result that the rate of decay of a sample of radioactive nuclei is given by

$$\frac{dX}{dt} = -\lambda X; \text{ or } X = X_o e^{-\lambda t}. \qquad (1)$$

Where X is the number of radioactive nuclei which have not decayed at time t, X_o is the value of X at t = 0, and λ is a number characteristic of the radioactive nucleus in question and is related to the half life.

The half life $T_{1/2}$ is the time required for half of the nuclei to decay. From equation 1, we see that $T_{1/2} = \frac{0.693}{\lambda}$. The mean life τ of a radioactive nucleus is the average time a nucleus can be expected to spend before decaying. This can be seen to be given by $\tau = \frac{1}{\lambda}$. In terms of τ, we have $T_{1/2} = (0.693)\tau$. The times for half lives and mean lives may be measured in any convenient unit.

A. Alpha Particle Decay

Many heavy nuclei are unstable against decay by alpha particle emission (see Figure 1). The half lives of naturally occurring alpha particle emitters are generally in the neighborhood of thousands to millions of years thus accounting for their natural existence in the earth's crust.

It is possible by means of the binding energy curve of Figure 2 to determine if a particular nucleus is stable against alpha decay, the condition being that the sum of the binding energies for the daughter nucleus

and an alpha particle be greater than the binding energy of the parent nucleus. Any excess of energy then manifests itself as the kinetic energy of the alpha particle and perhaps also some energy will go into the production of one or more gamma rays. The alpha particles have well defined energies which generally fall in the range 3 to 10 Mev. Alpha particles have well defined short ranges and by themselves present no hazard to workers unless the source is in contact with the skin or the radioactive material is taken internally. The gamma rays associated with the alpha particle decay of some nuclides may be dangerous at a distance however.

B. Beta Decay

The beta rays discovered and investigated by Becquerel, the Curies, and others near the year 1900, have been identified as being high energy electrons. Anderson in 1932, discovered the positron or positively charged electron in cosmic radiation. Shortly later, positrons were observed to be emitted in the decay of certain artificially produced radioactive nuclei. It has been observed that the details of the radioactive electron and positron decay processes are very similar. Further, it is felt that the K capture and L capture processes in which an atomic electron in a K or L orbital is absorbed by a radioactive nucleus are closely related to the electron and positron emission processes mentioned above. Therefore, we often see these three different kinds of radioactive decay referred to as beta processes. In beta processes, the nucleus changes its atomic number Z by one unit, but the mass number A does not change, or we may say a neutron has changed into a proton or vice versa. In the case of electron emission Z increases by unity, and in positron emission and K and L electron capture, Z decreases by unity. We often see electron and positron decays represented

in the following ways:

electron emission $\begin{cases} {}_1H^3 \xrightarrow[12y]{\beta^-} {}_2He^3, \text{ or } {}_1H^3 \xrightarrow[12y]{e^-} {}_2He^3, \\ {}_1H^3 \rightarrow {}_2He^3 + e^- \ (12y), \text{ etc.} \end{cases}$

positron emission $\begin{cases} {}_7N^{13} \xrightarrow[10\ min]{\beta^+} {}_6C^{13}, \text{ or } {}_7N^{13} \xrightarrow[10\ min]{e^+} {}_6C^{13}, \\ {}_7N^{13} \rightarrow {}_6C^{13} + e^+ \ (10\ min), \text{ etc.} \end{cases}$

Simple criteria may be readily developed to determine if a nucleus is stable against beta decay. These, as in the case of alpha particle decay, depend on the binding energies of the parent and daughter nuclei, and on the amount of energy necessary to <u>create</u> the beta particle. (0.511 Mev) The energy change E_o in the process has a definite value, yet a <u>spectrum</u> of electron or positron energies is observed from zero up to E_o. Figure 3 shows such a spectrum. Fermi and Pauli postulate the existence of a neutral particle, the <u>neutrino</u>, which is emitted in coincidence with the electron or positron and carries away an amount of energy such that the sum of the beta particle energy and the neutrino energy is equal to E_o. The tabulated beta decay energies are the values of E_o. Neutrinos interact so slightly with matter that they usually escape the region in which they were produced. Considerable energy is lost from nuclear reactors in this way and amounts to about 5 percent of the fission energy of uranium or plutonium.

The half lives of beta radioactive nuclides vary from fractions of a second to many years. Beta particles may be stopped by thin metal sheets. Their ranges in material are not as well defined as in the case of alpha particles. Figure 18 in Part II, Section 3-F shows the thickness of various materials necessary to stop beta particles of various energies.

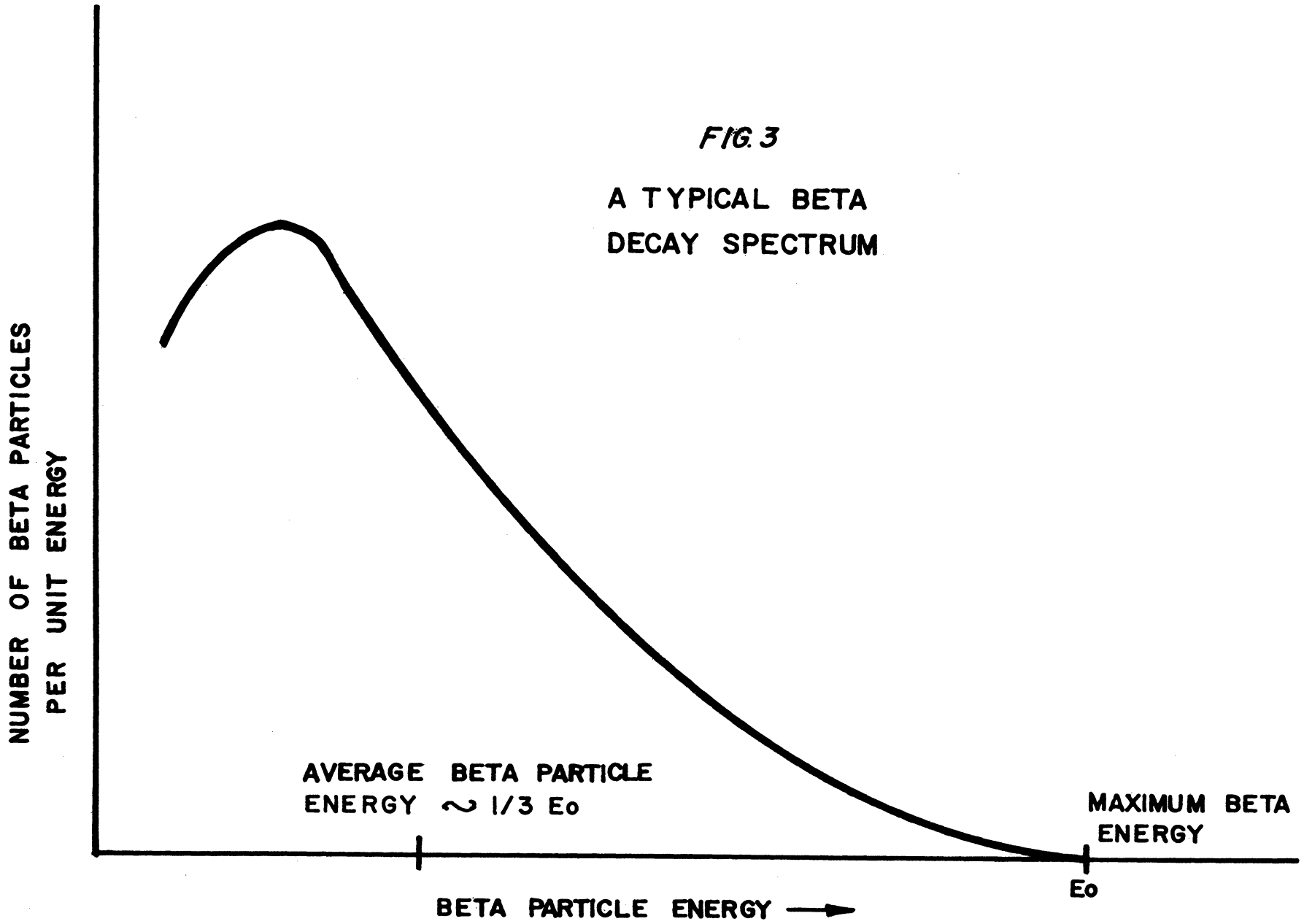

FIG. 3

A TYPICAL BETA
DECAY SPECTRUM

C. Gamma Ray Emission

Gamma-rays are electromagnetic radiation like X-rays but generally have much shorter wave lengths. Gamma rays produced in nuclei are monochromatic and have quantum energies between several kilovolts up to nearly 18 Mev. A nucleus radiates a gamma ray in transitions between excited states just as occurs in the emission of visible light by atoms. It is evident that neither Z nor A changes in the radiating nucleus. Gamma rays do not have well defined ranges but are absorbed exponentially (see the discussion under shielding). The rate of absorption is greatest for heavy metals.

Alpha and beta particle emitters usually emit gamma rays as well. After the emission of the alpha or beta particle, the daughter nucleus is left in an excited state which decays almost instantaneously (within about 10^{-12} seconds) with the radiation of a gamma ray. The products of nuclear reactions are often left in excited states and also radiate gamma rays.

Some gamma radioactive nuclei have lifetimes much longer than the usual value of about 10^{-12} seconds. Nuclides in these long lived excited states are called nuclear isomers and decay by isomeric transitions (often abbreviated I.T.) to the ground state.

D. Natural and Artificial Radioactive Nuclei

The vast majority of naturally occurring radioactive elements are heavy metals at the far end of the periodic table (see Figure 1). There are three distinct naturally occurring decay chains of heavy metal radioactive nuclei. These are the Uranium Series which starts with $_{92}U^{238}$ and after many alpha and beta decays, results in the formation of stable $_{82}Pb^{206}$, the Thorium Series which has $_{90}Th^{232}$ as a precursor and eventually yields stable $_{82}Pb^{208}$, and finally the Actinium Series with $_{92}U^{235}$ as a precursor and stable $_{82}Pb^{207}$

as an end product, the element actnium being an intermediate product. The details of the Uranium Series decay chain are given below in Table 2 as an example.* There is another series of heavy metal radioactive elements similar to the three above series which has been observed in the decay of artificially produced radio-nuclides but has not yet been found to occur in nature. This is the Neptunium Series which starts with $_{94}Pu^{241}$ and ends with stable $_{83}Bi^{209}$.

A selected list of both naturally occurring and artificially produced radioactive nuclides is given below in Table 2.** More detailed compilations of alpha and beta decays are given in: "Table of Total Beta-Disintegration Energies" by R. W. King, and "Table of the Alpha-Disintegration Energies" by F. Asaro and I. Perlman in Reviews of Modern Physics, October 1954. Reprints of these articles may be obtained from the Publications Office, National Research Council, 2101 Constitution Avenue, Washington 25, D. C.

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* This decay chain as well as the others are described in detail in Source-book on Atomic Energy, by Samuel Glasstone, D. Van Nostrand Company, Inc., New York, 1950, p. 125.

** Taken from Introduction to Nuclear Engineering, by R. Stephenson, McGraw Hill Book Company, Inc., New York, 1954, pp. 372-274.

TABLE I

THE URANIUM SERIES

Nuclide	Half Life	Radiation
$_{92}U^{238}$	4.49×10^9 y	α,γ
$_{90}Th^{234}$	24.1 d	β,γ
$_{91}Pa^{234}$	1.2 m	β,γ
$_{92}U^{234}$	2.48×10^5 y	α,γ
$_{90}Th^{230}$	8.0×10^4 y	α,γ
$_{88}Ra^{226}$	1620 y	α,γ
$_{86}Rn^{222}$	3.8 d	α
$_{84}Po^{218}$	3.05 m	α,β
$_{82}Pb^{214}$ (0.04) / $_{85}At^{218}$	26.8 m / 1.5 s	β,γ / α
$_{83}Bi^{214}$	19.7 m	α,β,γ
$_{84}Po^{214}$ (0.04) / $_{81}Tl^{210}$	1.6×10^{-4} s / 1.32 m	α / β,γ
$_{82}Pb^{210}$	22 y	β,γ
$_{83}Bi^{210}$	4.99 d	β
$_{84}Po^{210}$ (10^{-5}) / $_{81}Tl^{206}$	138 d / 4.2 m	α,γ / β
$_{82}Pb^{206}$	STABLE	

TABLE 2
COMMON RADIOSOTOPES*

Radio-nuclide	Half life	Beta particle, Mev	Gamma ray, Mev
H^3	12 yr	0.018	None
Li^8	0.9 sec	12	Weak
Be^7	54.5 days	K capture	0.48 (12%)
Be^{10}	2.5×10^6 yr	0.56	None
B^{12}	0.03 sec	13	Weak
C^{14}	5800 yr	0.155	None
N^{13}	10 min	1.24 (e^+)	None
N^{16}	7.35 sec	10 (18%), 3.8-4.6 (82%)	6.2
O^{15}	2 min	1.68 (e^+)	None
O^{19}	29.4 sec	2.9 (70%), 4.5 (30%)	1.6 (70%)
F^{20}	12 sec	5.1	2.2
Na^{22}	2.8 yr	0.575 (e^+)	1.28
Na^{24}	15 hr	1.39	2.76 and 1.38
Mg^{27}	9.6 min	1.8 (80%), 0.9 (20%)	1.01 (20%), 0.84 (100%)
Al^{28}	2.3 min	3.01	1.8
Si^{31}	2.7 hr	1.6	None
P^{32}	14.3 days	1.71	None
S^{35}	87 days	0.167	None
Cl^{36}	4×10^5 yr	0.7	Weak
Cl^{38}	38 min	4.81 (53%), 2.77 (16%), 1.11 (31%)	1.6 (31%), 2.15 (47%)
A^{37}	34 days	K capture, L capture	None
A^{41}	1.8 hr	1.2	1.3
K^{42}	12.4 hr	3.58 (75%), 2.04 (25%)	1.51 (25%)
Ca^{45}	152 days	0.25	None
Sc^{46}	85 days	1.49 (2%), 0.36 (98%)	1.12 (98%), 0.89 (100%)
V^{52}	3.9 min	2.3	1.45
Cr^{51}	26.5 days	K capture	0.32 (3%), 0.267 (weak)
Mn^{54}	310 days	K capture	0.84
Mn^{56}	2.6 hr	2.86 (60%), 1.05 (25%), 0.73 (15%)	0.845, 1.81 (25%), 2.13 (15%)
Fe^{55}	2.9 yr	K capture	None
Fe^{59}	47 days	0.46 (50%), 0.26 (50%)	1.3 (50%), 1.1 (50%)
Co^{57}	270 days	0.26 (e^+)	0.131
Co^{60}	5.3 yr	0.31	1.17 and 1.33
Ni^{63}	85 yr	0.06	None
Cu^{64}	12.9 hr	0.57 (35%), 0.65 (e^+ 20%), K capture (45%)	1.34 (1%)
Cu^{66}	4.3 min	2.7	1.32
Zn^{65}	250 days	0.32 (3% e^+), K capture (97%)	1.11 (46%)
Zn^{69}	14 hr	IT	0.439
Ga^{72}	14 hr	3.17 max (see charts)	2.5 max (see charts)
As^{76}	27 hr	3.12 max (see charts)	2.1 max (see charts)
As^{77}	40 hr	0.7	None
Se^{75}	115 days	K capture	0.405 max (see charts)

COMMON RADIOISOTOPES (Cont'd.)

Radio-nuclide	Half life	Beta particle, Mev	Gamma ray, Mev
Br^{82}	36 hr	0.465	0.547, 0.787, 1.35
Br^{87}	55.6 sec	2 (55%), 8 (45%), delayed neutrons	3
Rb^{86}	19.5 days	1.82 (80%), 0.72 (20%)	1.1 (20%)
Sr^{89}	53 days	1.5	None
Y^{90}	61 hr	2.2	None
Zr^{95}	65 days	0.887 (2%), 0.4 (98%)	0.708 (98%)
Nb^{95}	35 days	0.146	0.758
Mo^{99}	67 hr	1.2 (75%), 0.5 (25%)	0.141, 0.726
Tc^{99}	3×10^5 yr	0.30	None
Ru^{97}	2.8 days	K capture	0.23
Ru^{103}	42 days	0.35 (50%), 0.665 (50%)	0.5 (50%)
Pd^{109}	13 hr	0.95	None
Ag^{110}	270 days	2.86 max (see charts)	1.5 max (see charts)
Ag^{111}	7.5 days	1.06	None
Cd^{115}	43 days	1.67	0.5
In^{114}	50 days	IT, 2.05 (97%), K capture (3%)	0.192, 0.715 (3%), 0.548 (3%)
Sn^{113}	112 days	K capture	0.09
Sb^{124}	60 days	2.37 max (see charts)	2.3 max (see charts)
I^{131}	8 days	0.60 (85%), 0.32 (15%)	0.638 (15%), 0.364 (85%)
I^{135}	6.7 hr	0.47 (35%), 1.0 (40%), 1.4 (25%)	1.3, 1.7
I^{137}	22 sec	Delayed neutrons	
Xe^{135}	9.2 hr	0.93	0.247
Cs^{134}	2.3 yr	0.658 (74%), 0.09 (26%)	0.794, 0.602, 0.568 (26%)
Cs^{137}	37 yr	1.2 (5%), 0.51 (95%)	0.669 (from 2.6-min Ba^{137})
Ba^{131}	12 days	K capture	0.26, 0.5 (strong)
Ba^{140}	12.8 days	1.022 (60%), 0.48 (40%)	0.54 (40%)
La^{140}	40 hr	2.26 (10%), 1.67 (20%), 1.32 (70%)	2.5 (6%), 1.6 (77%) other low-energy gammas
Ce^{141}	28 days	0.56 (30%), 0.41 (70%)	0.141 (70%)
Ce^{144}	275 days	0.32	0.13 (strong)
Pr^{142}	19 hr	2.15 (96%), 0.64 (4%)	1.57 (4%)
Pr^{143}	13.8 days	0.92	None
Nd^{147}	11 days	0.78 (67%), 0.17 (33%)	0.035 (strong), 0.58 (weak)
Pm^{147}	2.7 yr	0.23	None
Sm^{153}	47 hr	0.8 (33%), 0.68 (67%)	0.10, 0.07
Hf^{181}	46 days	0.42	0.34 (22%), 0.48 (78%)
Ta^{182}	122 days	0.50	1.2 max, many others
W^{185}	77 days	0.43	0.134
W^{187}	25 hr	1.32 (30%), 0.63 (70%)	0.68 max, others
Re^{186}	90 hr	1.09 (67%), 0.95 (30%)	0.132 (37%), 0.275 (23%)
Os^{191}	15 days	0.14	0.13, 0.04
Ir^{192}	70 days	0.67	0.65 max, many others
Au^{198}	2.7 days	0.97	0.411

COMMON RADIOISOTOPES (Cont'd.)

Radio-nuclide	Half life	Beta particle, Mev	Gamma ray, Mev
Hg^{197}	2.7 days	K capture	0.077
Hg^{203}	44 days	0.205	0.286
Tl^{204}	2.7 yr	0.78	None
Pb^{210}	22 yr	0.028	Soft
Bi^{210}	5 days	1.17	None
Po^{210}	138 days	4.95 (alpha)	None
Rn^{222}	3.82 days	5.49 (alpha)	None
Ra^{226}	1620 yr	4.7 (alpha)	0.188
Th^{232}	1.39×10^{10} yr	4.1 (alpha)	None
Th^{233}	23.5 min	1.2	None
Th^{234}	24.1 days	0.205 (80%), 0.11 (20%)	0.093 (20%)
Pa^{233}	27.4 days	0.58 max (see charts)	0.471 max (see charts)
Pa^{234}	1.2 min	2.32 (98%), also IT	See charts
U^{233}	1.6×10^{5} yr	4.82 (alpha)	0.04
U^{234}	2.5×10^{5} yr	4.76 (alpha)	Weak
U^{235}	8.8×10^{8} yr	4.5 (alpha)	0.17
U^{236}	2.5×10^{7} yr	4.5 (alpha)	None
U^{238}	4.5×10^{9} yr	4.18 (alpha)	None
U^{239}	23.5 min	1.2	0.074
U^{240}	14 hr		
Np^{239}	2.3 days	See charts	See charts
Pu^{239}	2.4×10^{4} yr	5.15 (alpha)	Weak
Pu^{240}	6600 yr	5.1 (alpha)	None

*From Introduction to Nuclear Engineering, R. Stephenson, McGraw-Hill Book Co., Inc., New York, N. Y., 1954, p. 372.

(IT) refers to isomeric transitions.

(Charts) refers to more complete descriptions such as found in: Nuclear Data, National Bureau of Standards Circular 499; also, R. W. King, Reviews of Modern Physics 26, 327 (1954).

3. NUCLEAR REACTIONS

A. General

In nuclear reactions, the total number of neutrons and protons is conserved and also the total charge is conserved. Thus in balancing an equation for a nuclear reaction, it is necessary that the sum of the mass numbers A for the reactants be equal to the sum of the A's for the products, and also that the sum of the charge numbers Z be the same for the reactants and products.

In counting the charge numbers, any electrons involved must be counted also. For example:

$_{94}Pu^{239} \rightarrow {}_{92}U^{235} + {}_{2}He^{4}$, Alpha decay of plutonium.

$_{1}H^{3} \rightarrow {}_{2}He^{3} + e^{-}$, Beta decay of tritium.

$_{4}Be^{9} + {}_{2}He^{4} \rightarrow {}_{6}C^{12} + {}_{0}n^{1}$, A neutron production reaction.

$_{92}U^{235} + {}_{0}n^{1} \rightarrow {}_{57}La^{147} + {}_{35}Br^{87} + 2\,{}_{0}n^{1}$, A fission reaction of uranium.

$_{1}D^{2} + \gamma \rightarrow {}_{1}H^{1} + {}_{0}n^{1}$, Photo disintegration of deuterium.

Another way in which nuclear reactions are represented is by classifying them according to the bombarding particle and the product particle or the reaction process. For example: the third reaction in the above list is written $_{4}Be^{9}\ (\alpha,n)\ {}_{6}C^{12}$ and is called an alpha,n reaction.

The deuteron photodisintegration is written ${}_1D^2(\gamma,n){}_1H^1$ and is called a gamma,n reaction. Neutron induced fission reactions are sometimes written as (n,f). Some other reactions often seen written in the literature in this fashion are the following: (n,2n); (n,p); (d,p); (d,p); (γ,d); (γ,np); etc.

B. The Compound Nucleus Theory of Nuclear Reactions

In the nuclear reactions involving medium to heavy weight nuclei in which free neutrons or protons are produced, the compound nucleus theory of Bohr has been very successful in explaining the experimental results and also provides a convenient physical picture. The compound nucleus is a highly excited nucleus which may have enough energy to emit a neutron, a proton, etc. The amount of excitation energy necessary to emit a single neutron or proton varies from one compound nucleus to the next, but on the average is about 8 Mev. According to Bohr's theory, the nuclear reaction may be thought of as occurring in three steps:

1. Excitation of the Initial Nucleus

This may be accomplished by absorption by the initial nucleus of a neutron, proton, deutron, alpha particle, a gamma ray, etc.

2. Formation of the Compound Nucleus

In this stage, the energy of excitation becomes averaged in the nucleus with the result that one says the compound system has been formed. The process probably requires many nuclear periods to take place or a time of the order of 10^{-15} sec. During this time, the compound nucleus forgets how it became excited.

3. Decay of the Compound Nucleus

If the excitation energy of the compound nucleus is greater than about 8 Mev., a neutron or a proton may be emitted. A proton

is much less likely to be emitted than a neutron because a proton encounters an electrostatic barrier at the edge of the nucleus. Proton emission is likely to be less than neutron emission by a factor of 100 to 1000, and so in a nuclear reactor such protons may be neglected. If sufficient excitation energy is available, we may get two neutrons, a neutron and a proton, or a deuteron. For a given amount of excitation energy, the decay particles will be emitted in a spectrum with a maximum value corresponding to the excitation energy. The remaining energy is radiated as gamma rays.

Some of the classes of reactions that may be described by the compound nucleus model are: (γ,n); (γ,p); (d,n); (n,2n); (α,n), etc.

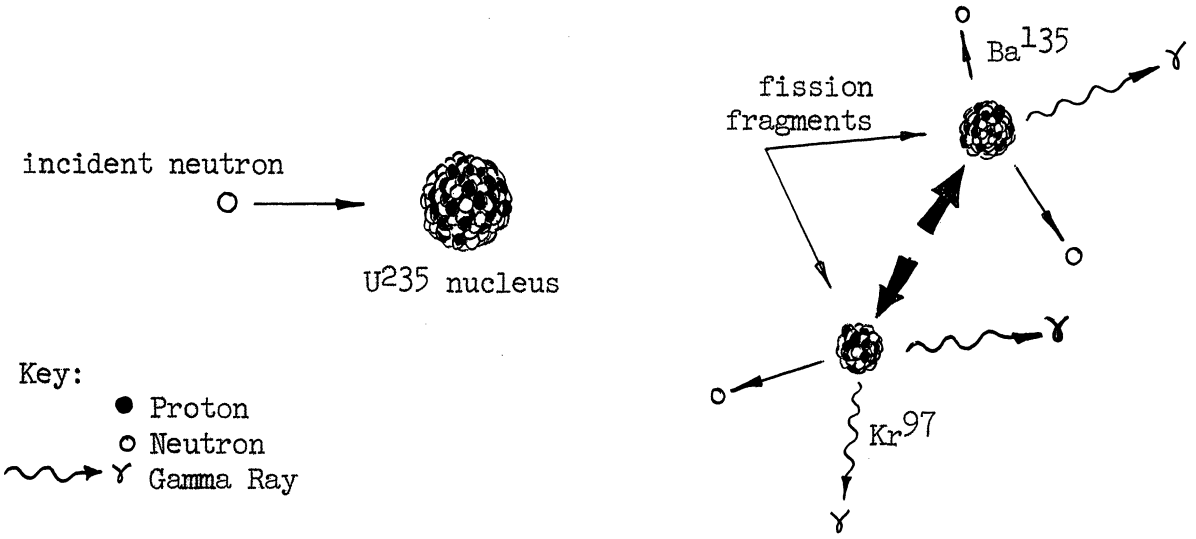
C. Nuclear Fission

Some of the very heavy nuclei are unstable against the breakup into two fragments. In a particular case, this may be determined by examining the binding energies of the fissioning nucleus and the prospective fission fragments. A few nuclei appear to undergo fission spontaneously, but in general the fission reaction requires some energy from an outside source to make the reaction proceed at an appreciable rate. This is similar to the situation with exothermic chemical reactions where heat or light is required to produce the activation energy. In the case of fission, the amount of activation energy required depends on the particular isotope involved. Some nuclei like U-233, U-235, and Pu-239 fission readily with the energy available from the capture of slow neutrons. On the other hand, U-238 requires at least 1.1 Mev. neutrons to cause the fission. Fission may also be induced by irradiation with high energy gamma rays. This latter process is referred to as photo-fission. The fission

process can be represented graphically as shown in Figures 4 and 5. In Figure 5, the ordinate is the potential energy of the system, and the abscissa is the distance of separation of the centers of gravity of the fission fragments. The amount of energy available by fission is E_s, and the activation energy or critical energy is E_c.

FIGURE 4

THE FISSION OF A
TYPICAL URANIUM-235 NUCLEUS

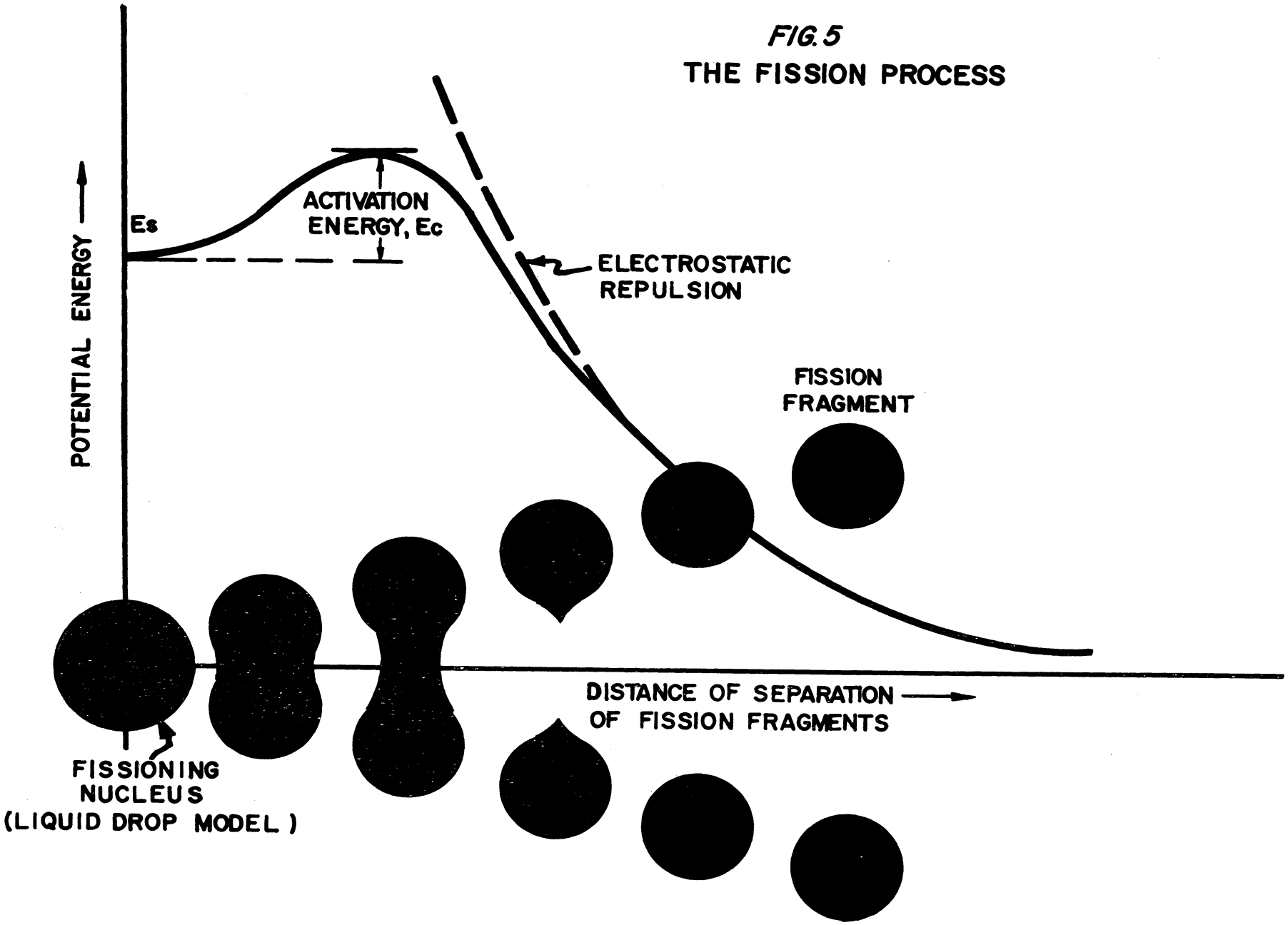


There is no unique mode of fission, i.e., fissioning nuclei may break up in many different ways to produce a variety fission product nuclides. The fission shown in Figure 4 is only one of these many fission modes.

The fission fragments lie near the middle of the dotted line on Figure 1, drawn between the fissioning nucleus and the origin. It can be seen from Figure 1 that the fission fragments have a considerable excess of neutrons over stable nuclei with the same Z. As formed, the fission products are thought to be compound nuclei, as described above, which decay with the emission of one or more neutrons. The emission of only one or

FIG. 5

THE FISSION PROCESS



two neutrons does not mean the fragments are stable, however, as they are still too rich in neutrons. Thus, these fragments undergo a series of beta decay processes in which Z increases and A remains constant until a stable nucleus results. A typical chain of beta decays is:

$$_{54}Xe^{140} \xrightarrow[16\ sec.]{\beta-} {}_{55}Cs^{140} \xrightarrow[66\ sec.]{\beta-} {}_{56}Ba^{140} \xrightarrow[12.8\ days]{\beta-} {}_{57}La^{140} \rightarrow$$

$$\xrightarrow[40\ hrs.]{\beta-} {}_{58}Ce^{140} \quad \text{(Stable)}$$

Gamma rays are often radiated accompanying these beta decays.

Some fission products are delayed neutron emitters, and are important in reactors since they augment the neutron flux available to cause fission. Delayed neutron emission by a radioactive nuclide occurs when there is a beta decay to yield an intermediate daughter nucleus in a highly excited state. If sufficient excitation is available, the intermediate compound nucleus then decays with the emission of a neutron in a time of the order of 10^{-15} seconds. We call the original parent nuclide a delayed neutron emitter since the neutron emission is delayed by the decay rate of the beta process which precedes it.

Below in Table 3 is a list of half lives of delayed neutron emitters observed in the fission products of U^{235}.

TABLE 3

Half Life, sec.	Energy, Mev.	Fraction of Total Fission Neutrons
0.43	0.42	.00084
1.52	0.62	.0024
4.51	0.43	.0021
22.0	0.56	.0017
55.6	0.25	.00026

The 55.6 sec. activity has been identified as being due to Br^{87}, and the 22 sec. activity is associated with I^{137}. The other activities have not been identified.

In Figure 6 is shown the energy spectrum of prompt neutrons obtained from the fission of U^{235} and Pu^{239}. Note that the form of these spectra is very closely given by $Ce^{-E} \sinh(2E)^{1/2}$ where E is the neutron energy in Mev., and C is a constant.

The fission product mass spectrum shows the probability of formation of a nuclide with mass number A when a heavy nucleus fissions. The probabilities are expressed in percent. Fission product mass spectra are shown in Figure 7 for the fission of U^{235}, Pu^{239}, and U^{238} by slow neutrons. Some data are also shown for the fast neutron fission of Pu^{239}.

It is of interest to have a breakdown of the energy evolved in the fission process. Recent estimates show that for the fissioning nucleus of either a U^{235} or Pu^{239} nucleus, the energy available is distributed in the following manner:

		Mev per fission	Kilowatt hrs. per fission
Energy Available Immediately After the Fission Process	Kinetic energy of fission fragments.....	162	
	Kinetic energy of prompt neutrons produced in the fission process.....	6	
	Energy of instantaneous gamma rays.....	6	
	Energy from absorption of excess neutrons produced in the fission process which are captured in non-fission processes by reactor materials.....	8	
	Total	182 Mev	8.08×10^{-18}
Energy Which Appears as the Fission Products Decay	Energy from fission product gamma rays.....	5	
	Energy from fession product beta particles.....	5	
	Total Energy Available	192 Mev	8.52×10^{-18}
Unavailable Energy	Energy carried away by neutrinos accompanying the fission product beta decays.....	11 Mev	0.488×10^{-18}

FIG. 6

SPECTRUM OF NEUTRONS PRODUCED BY SLOW FISSION OF U–235 AND Pu–239

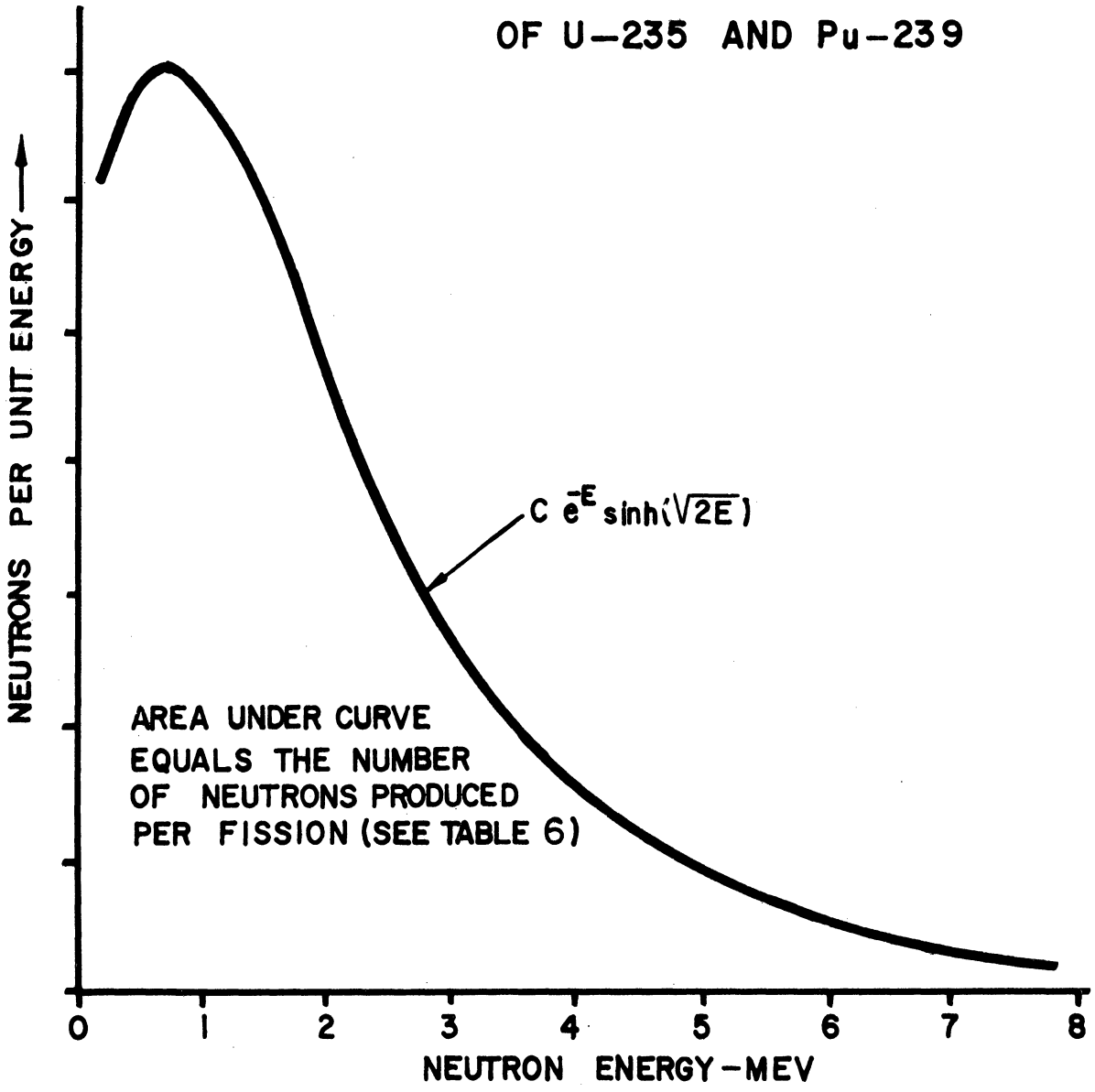
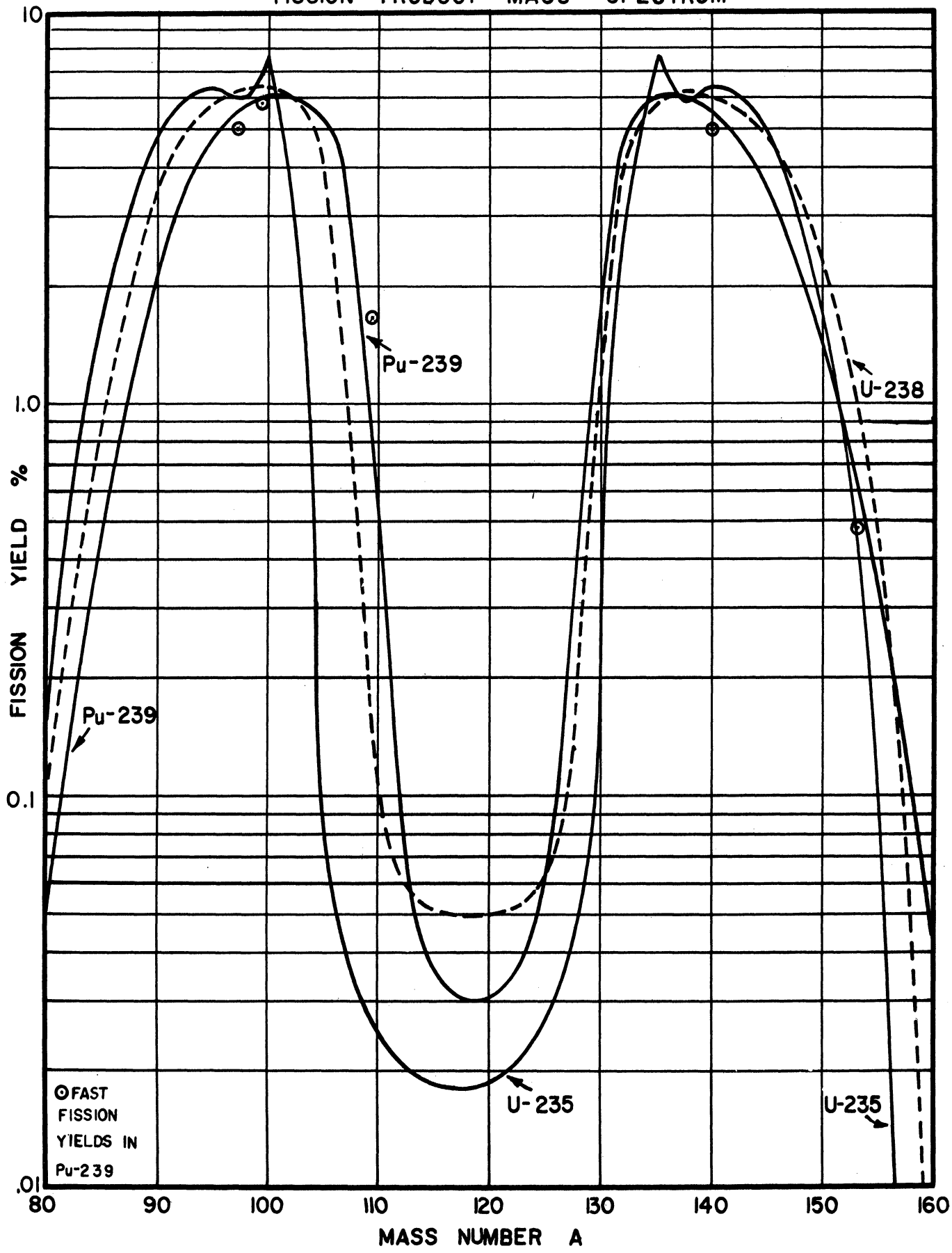


FIG. 7

FISSION PRODUCT MASS SPECTRUM



The energy carried away by the neutrinos cannot be recovered since there is no known shielding material that will stop them.

Although U^{235} is the most widely used nuclear reactor fuel, U^{233} and Pu^{239} can be used in certain reactors as well. Many other heavy nuclides fission besides these three. In order to be of practical use as fuel in nuclear reactors, a fissile nuclide must either be obtainable naturally or readily produced from naturally occurring materials. (See Part II, Section 2-D on fuel breeding).

D. Fusion and Thermonuclear Reactions

Fusion may be thought of as an inverse of the fission reaction. Thus fusion is a reaction in which two light nuclei combine to form a heavier nucleus. There are many fusion reactions which are exoergic, i.e., will give up energy to the surroundings. For example:

$$_1H^1 + {}_1T^3 \rightarrow {}_2He^4 + Q \text{ (energy)}$$

$$_1D^2 + {}_5B^{10} \rightarrow {}_6C^{12} + Q$$

Even though these reactions are exoergic (Q is positive), they will not proceed spontaneously because the electrostatic repulsion between the two nuclei prevents their approach to a distance close enough for a reaction to take place. In order for these reactions to occur, the reactants must come together with a great deal of relative energy. This can be done either by using particle accelerators or very high temperatures. In the latter case, the reactions are called thermonuclear reactions, and it is believed such nuclear reactions are responsible for the large energy output from the sun and other stars.*

*See for example, "The Birth and Death of the Sun," by George Gamow.

E. Nuclear Cross-Sections

The rate at which externally induced nuclear reactions occur brings us to the definition of the cross-section. The reaction rate in nuclear events per cm^3 of sample bombarded per second is proportional to the flux ϕ of incident particles per cm^2 per second inducing the reaction, and to the number N of nuclei per cm^3 of the nuclear species of interest.

$$(\text{Reaction Rate}) = \sigma \phi N$$

The proportionality factor σ is seen to have the dimensions of an area in cm^2. σ is called the cross-section for the nuclear event in question, and may be considered to be the effective area that the nucleus presents to the incident flux. Since many different kinds of nuclear events can take place, there are as many different kinds of cross-sections. In (n,γ) reactions, neutrons are absorbed and gamma rays radiated, so the cross-section for these reactions is called the neutron radiative absorption cross-section σ_c. In a similar manner, the fission cross-section σ_f is defined, as well as others like the elastic scattering cross-section σ_{sc} which describes the rate at which an incident beam of particles is deflected due to elastic collisions with nuclei. Cross-sections are usually additive so that the rate, at which neutrons, for example, are removed from an incident beam is given by

$$\phi N (\sigma_c + \sigma_f + \sigma_{sc} + \cdots) = N\sigma_t,$$

where σ_t is called the total neutron cross-section. If the sample of material bombarded contains many different nuclear species, the rate of removal of neutrons from the incident beam is given by a sum of such terms. viz:

$$\phi (N_1\sigma_1 + N_2\sigma_2 + \text{---} + N_n\sigma_n)$$

where N_1 is the number of nuclei of species 1 per cm^3 and σ_1 is the sum of the cross-sections for absorption and scattering of the incident particles, N_2 and σ_2 are the same for nuclear species 2, etc.

For example, the rate of absorption of neutrons by natural uranium is given by:

$$\phi\left[N_{235}(\sigma_c^{235} + \sigma_f^{235}) + N_{238}\sigma_c^{238}\right]$$

Cross-sections are usually expressed in units of 10^{-24} cm^2. This unit is called the barn (b) and is of the same order of magnitude as the geometrical area that a nucleus presents to the beam of incident particles. Some very small cross-sections are given in units of 10^{-27} cm^2 or millibarns (mb). σ is often called the microscopic cross-section as it describes the reaction rate due to a single nucleus. The macroscopic cross-section Σ on the other hand is defined as the product $N\sigma$ where N is the number of nuclei of the type under consideration per cubic centimeter of material. The units for Σ are cm^{-1}.

Neutron cross-sections are dependent on the energy of the incident neutron. For very low energy neutrons or thermal neutrons,* most neutron absorption cross-sections are proportional to 1/v where v is the velocity of the incident neutron. This is often called the 1/v law. Extensive tables of thermal neutron cross-sections are given in an unclassified document.** Thermal neutron absorption cross-sections range from a fraction of a millibarn to over a million barns, while nuclear areas are all of the order of

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* Thermal neutrons have energies in the range between zero and 0.1 electron volts. They are called thermal neutrons because a neutron gas in thermal equilibrium with its surroundings at room temperature has a distribution of energies in this range.

** Neutron Cross-Sections, AECU-2040, obtainable from the Office of Technical Services, Department of Commerce, Washington 25, D. C.

one barn, and so there is little correlation between the neutron absorption cross-sections and nuclear size at thermal neutron energies. At high neutron energies, (greater than about 15 Mev) the total neutron cross-sections approach the value $2\pi R^2$ where R is the nuclear radius.

Thermal neutron cross-sections are usually stated at an incident neutron energy of 0.026 electron volts which means the neutrons have a velocity of about 2200 meters per second.* (See Table 4)

Of particular interest in reactor design are the thermal neutron capture and fission cross-sections for the fissionable fuel metals. A table of the values of some of these cross-sections is given in Part II, section II-A.

Plots of total neutron cross-sections versus neutron energy for some elements important in reactor design are shown in Figure 8.

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*This velocity of 2200 meters per second = 7,219 feet per second = 4,922 mph is chosen as it corresponds to the most probable neutron velocity found in a neutron gas in thermal equilibrium at 27° C. (81°F).

TABLE 4

THERMAL NEUTRON ABSORPTION CROSS SECTIONS*

Element	Isotope	isotopic Abundance, per cent	Cross Section, barns
H	--	--	0.33
	H^1	100	0.33
	H^2	0.015	0.46 mb
He	--	--	Variable
	He^3	0.00013	np 5200
	He^4	100	0
Li	--	--	67
	Li^6	7.5	nα 910
	Li^7	92.5	33 mb
Be	Be^9	100	9.0 mb
B	--	--	750
	B^{10}	18.8	nα 3990
	B^{11}	81.2	50 mb
C	--	--	4.5 mb
	C^{12}	98.9	
	C^{13}	1.1	1.0 mb
N	--	--	1.78
	N^{14}	99.6	np 1.70, nγ 0.10
	N^{15}	0.37	0.024 mb
O	--	--	0.2 mb
	O^{16}	99.76	Very small
	O^{17}	0.037	nα 0.5
	O^{18}	0.20	0.21 mb
F	F^{19}	100	10 mb
Ne	--	--	2.8
Na	Na^{23}	100	0.49
Mg	--	--	59 mb
Al	Al^{27}	100	0.22
Si	--	--	0.13
P	P^{31}	100	0.19
S	--	--	0.49
Cl	--	--	31.6
A	--	--	0.62
K	--	--	1.97
Ca	--	--	0.43
Ti	--	--	5.6
V	--	--	4.7
Cr	--	--	2.9
Mn	Mn^{55}	100	12.6
Fe	--	--	2.43
Co	Co^{59}	100	34
Ni	--	--	4.5
Cu	--	--	3.59
Zn	--	--	1.06
Zr	--	--	0.18
Mo	--	--	2.4

THERMAL NEUTRON ABSORPTION CROSS SECTIONS (Cont'd.)

Element	Isotope	isotopic Abundance, per cent	Cross Section, barns
Cd	--	--	2400
In	--	--	190
Sn	--	--	0.65
Xe	--	--	35
	Xe^{135}	0	3.5×10^6
Sm	--	--	6500
	Sm^{149}	13.8	50,000
Eu	--	--	4500
Gd	--	--	44,000
Hf	--	--	115
Ta	--	--	21.3
Au	Au^{197}	100	94
Hg	--	--	380
Pb	--	--	0.17
Bi	Bi^{209}	100	32 mb
Th	Th^{232}	100	7.0
	Th^{233}	0	1400
Pa	Pa^{233}	0	37
U	--	--	$n\gamma$ 3.50, nf 3.92
	U^{235}	0.714	$n\gamma$ 101, nf 549
	U^{238}	99.3	2.80
	U^{239}	0	22
Pu	Pu^{239}	0	$n\gamma$ 361, nf 664

*From "Introduction to Nuclear Engineering," R. Stephenson, McGraw-Hill Book Co., Inc., New York, N. Y., 1954, p. 375.

mb means "millibarns" or 10^{-3} barns.
One mb = 10^{-27} cm^2.

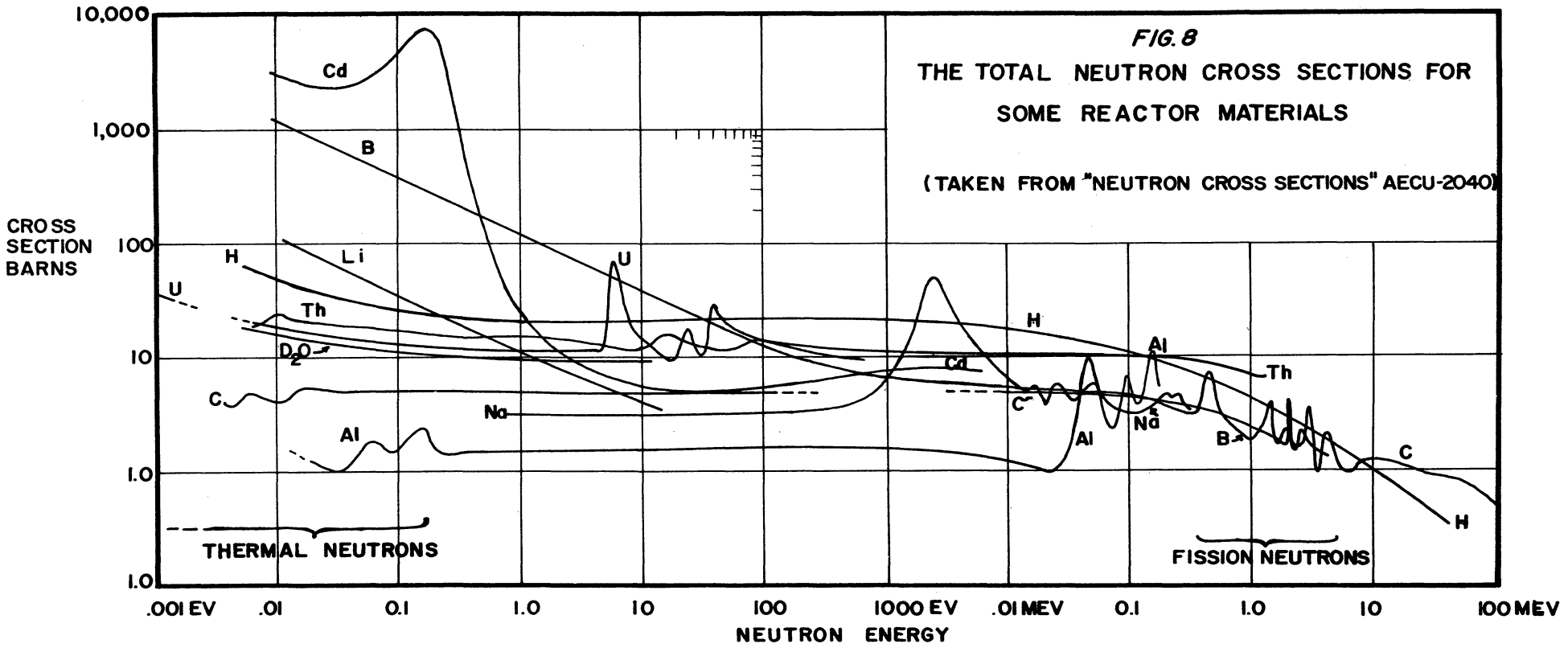
FIG. 8
THE TOTAL NEUTRON CROSS SECTIONS FOR SOME REACTOR MATERIALS
(TAKEN FROM "NEUTRON CROSS SECTIONS" AECU-2040)
CROSS SECTION BARNS
10,000
1,000
100
10
1.0
1.0
Cd
B
H
Li
U
Th
D2O
C
Na
Al
U
THERMAL NEUTRONS
H
Cd
C
Al
Na
Al
B
Th
C
H
FISSION NEUTRONS
.001 EV
.01
0.1
1.0
10
100
1000 EV
.01 MEV
0.1
1.0
10
100 MEV
NEUTRON ENERGY

PART II - NUCLEAR CHAIN REACTION - REACTOR MATERIALS

1. CHAIN REACTIONS AND NUCLEAR REACTORS - GENERAL DISCUSSION

A. Nuclear Chain Reaction; the Critical Fuel Mass

On the average, 2.5 neutrons are produced in the fission of one U^{235} nucleus. These neutrons can cause fission of other U^{235} nuclei and so in principle, it is possible that a chain reaction can occur in which new fissions are being caused by neutrons produced in previous fissions. These considerations also apply to other fissionable materials such as U^{233} and Pu^{239}.

A nuclear reactor is an assemblage of fissionable fuel, structural and other materials in which a nuclear chain reaction can take place. (A specific type of nuclear reactor of historical importance is the pile, which consists of a matrix of metallic uranium slugs and blocks of graphite.) The atomic bomb is a nuclear reactor which is specifically designed to momentarily contain an uncontrolled chain reaction until the fuel is almost completely consumed, before destroying itself. Nuclear reactors in which the chain reaction is controlled are used to produce intense neutron beams for physics research, to produce radioactive nuclei of species not obtainable naturally for medical and radioactive tracer studies, to manufacture plutonium from uranium for use in the weapons program, and to produce heat for power generation, ship propulsion, etc.

Of the neutrons produced in a chain reaction, many either escape the region in which the fuel is concentrated, or are captured in (n,γ) reactions with U^{238} and other nuclides present. In order for a chain reaction to be self-sustaining, the number of neutrons producing new fissions must be equal to or greater than the number of fissions producing them. Another

way of saying this is that the number of neutrons in one generation should be equal to or greater than the number in the previous generation, or that the ratio of these numbers be equal to or greater than unity. This ratio is called the multiplication factor k.

$$k = \frac{\text{(No. of neutrons in one generation)}}{\text{(No. of neutrons in the previous generation)}}$$

To be more precise, it is the product $k\,P = k_{eff}$, the effective multiplication factor, which must be greater than or equal to unity to insure a chain reaction. Here P is the probability that neutrons do not escape from the edge of the reactor, or the non-leakage probability. A reactor having $k_{eff} < 1$ is said to be sub-critical and a chain reaction cannot be maintained. When $k_{eff} = 1$, the reactor is critical and the reaction is self-sustaining. If $k_{eff} > 1$, the reactor is super-critical, and the reaction tends to build up.

The multiplication factor k, defined above, is dependent on microscopic quantities such as the fission and capture cross sections. On the other hand, the non-leakage probability P is dependent mainly on the size and shape of the whole reactor. If the reactor is small, there is a good chance that neutrons produced in the reactor will escape without causing fission and therefore P is small. For a given k and geometrical form for the reactor, there is a minimum reactor size for which $kP = 1$. This is the critical size of the reactor. The mass of fuel in the reactor of critical size is the critical mass. It should be noted that for a given reactor volume, neutrons more readily escape from a reactor with a large surface area, and so the critical mass may be kept at a minimum if the area is a minimum or the reactor is spherical. Another way of keeping the critical

mass small is to use a moderator, and take advantage of the large thermal neutron fission cross sections of fuels. These large cross sections reduce the average distance a neutron travels before being captured by a fuel nucleus, and consequently reduce the leakage.

It is necessary to keep in mind the possibility of inadvertently assembling a mass of fuel as metal or in solution that will be of critical mass. This caution is important mainly in dealing with uranium enriched in U^{235} or with Pu^{239}. For example, about 800 grams of U^{235} in aqueous solution contained in a spherical vessel of one foot in diameter can be critical if a neutron reflector is brought near.

A technique of experimentally determining the critical size and critical mass of an assembly of fuel and moderator is the construction of the critical assembly. Here an external neutron source such as the radium-beryllium source* is placed in the center of a small amount of the fuel-moderator assembly. The neutron flux is measured by two or more neutron detectors placed in various locations, and the fuel-moderator combination is added in small amounts. A plot of the reciprocal of the measured counting rates from the neutron counters vs. the total fuel mass is kept concurrently which will be of the form shown in Figure 9. When the critical mass is reached, the multiplication of the neutron flux from the Ra-Be source by the assembly will be infinite or the reciprocal counting rates will be zero. However, it is not necessary to add enough fuel to make the assembly critical, since an extrapolation of the reciprocal counting rates to zero will intersect the horizontal axis at the critical mass.

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*The alpha particles from the decay of radium react with beryllium to give neutrons through the reaction: $Be^{9}(\alpha,n)C^{12}$.

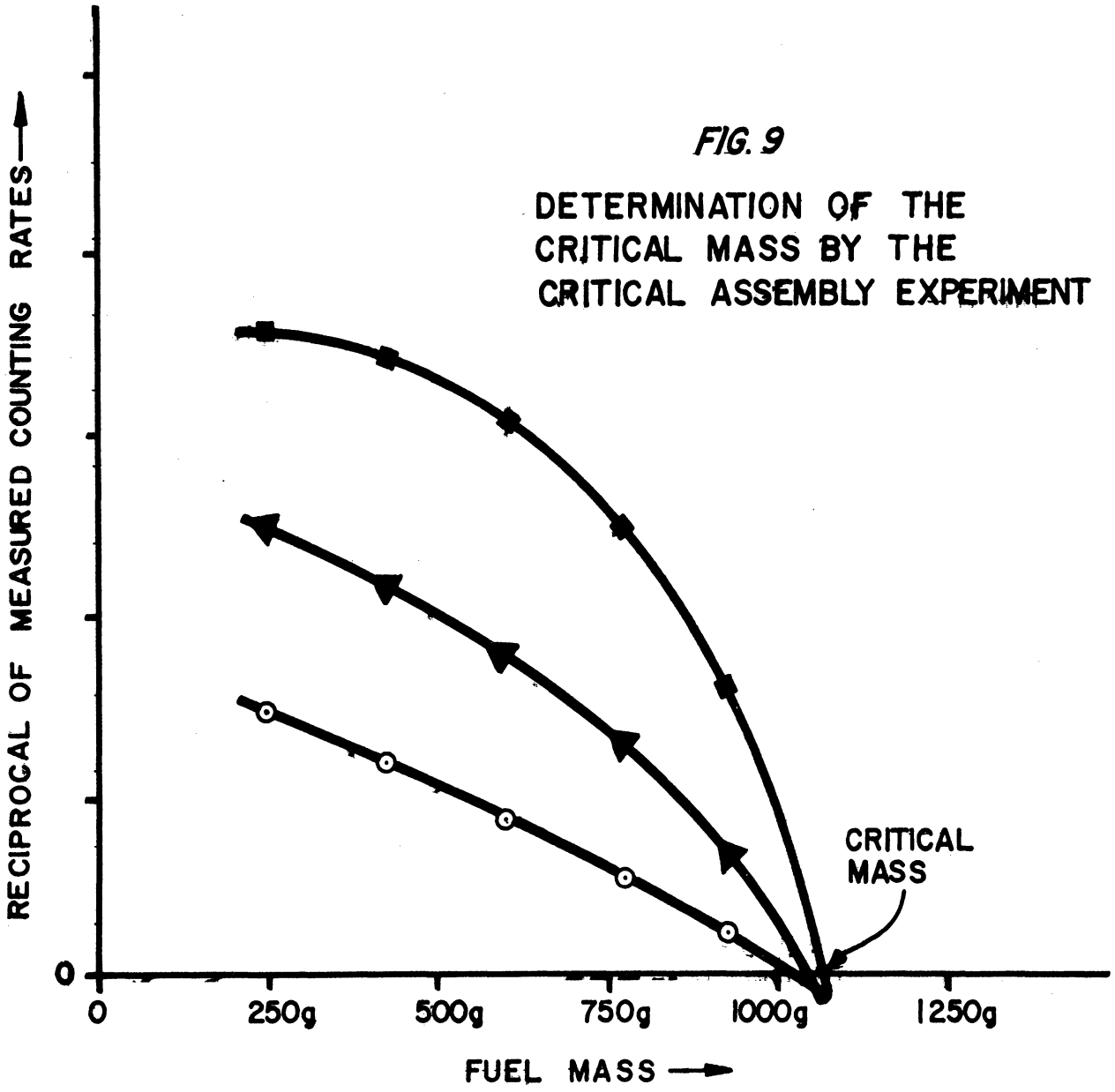


FIG. 9

DETERMINATION OF THE CRITICAL MASS BY THE CRITICAL ASSEMBLY EXPERIMENT

Anything that captures neutrons, other than material which will convert to fissionable materials, may be termed a reactor poison. This includes the fission products that are formed as a result of fissioning, the control rods that are purposely introduced to absorb neutrons, and structural materials which may be inherent in fuel element design or structural components of the reactor itself.

B. Reactor Classification Based on Neutron Energies

The fission cross-section for natural uranium is about one barn for neutrons with energies in the neighborhood of 1 Mev. For thermal neutrons, however, the fission cross-section for U^{235} is 549 barns (See Table 6), and so it may be advantageous to have thermal neutrons available to cause new fissions. Since the neutrons produced by the fission process have average energies of about 1 Mev (See Figure 6), they must be slowed down by substances called moderators to thermal energies in order to take advantage of the high thermal neutron fission cross-sections of U^{235} and Pu^{239}. Moderator nuclei slow neutrons by absorbing their energies in recoils from elastic collisions. A good moderator has a small neutron capture cross-section and a small mass number A. Moderators are discussed in more detail in section III-B.

Reactors using thermal neutrons to cause fission are called thermal reactors, and those using fast neutrons (no moderator used) are called fast reactors.

C. Reactor Classification Based on Fuel Distribution

Reactors may be further classified according to the distribution of fuel. If the fuel is distributed uniformly as in the case of an aqueous solution of a uranium salt or a molten solution of uranium metal in another

metal, the reactor is called a homogeneous reactor. If on the other hand, the fuel is lumped into fuel elements separated from one another by the moderator or some other substance, we have a heterogeneous reactor.

D. Reflectors and Blankets

In order to reduce the escape of neutrons from a reactor, a reflector is used. The reflector action is diffuse reflection and is due to the neutrons being elastically scattered by the nuclei of the reflector material back into the reactor core. Good reflector materials have large neutron scattering cross-sections and small capture cross-sections. Very pure graphite is often used. A beam of neutrons incident on a 15-inch thick slab of graphite is 90% diffusely reflected, i.e., the slab has an albedo of 0.90.

Sometimes a natural uranium blanket surrounds the reactor core to capture neutrons which would otherwise be lost. Capture by U^{238} yields U^{239} which decays by two beta emissions to Pu^{239}. In this way, fissionable Pu^{239} may be made from non-fissionable U^{238}. Similarly, a thorium blanket can be used to produce U^{233} which is fissionable. This is referred to as fuel breeding, and is discussed in more detail in section II-E. (See Fig. 10.)

2. NUCLEAR FUELS

A. Natural Uranium

The main mineral sources of uranium in the world are shown in Table 5. The pitchblende are the richest uranium ores and consist of U_3O_8 and ores of other metals such as lead, copper, and silver.

Carnotite is a uranium-vanadium mineral, $K_2O \cdot 2UO_3 \cdot V_2O_5 \cdot nH_2O$, and is found in Colorado and Utah as a cement in certain sandstones. Carnotite is often found in combination with other uranium-vanadium minerals.

FIG. 10

BASIC REACTOR SCHEMATIC

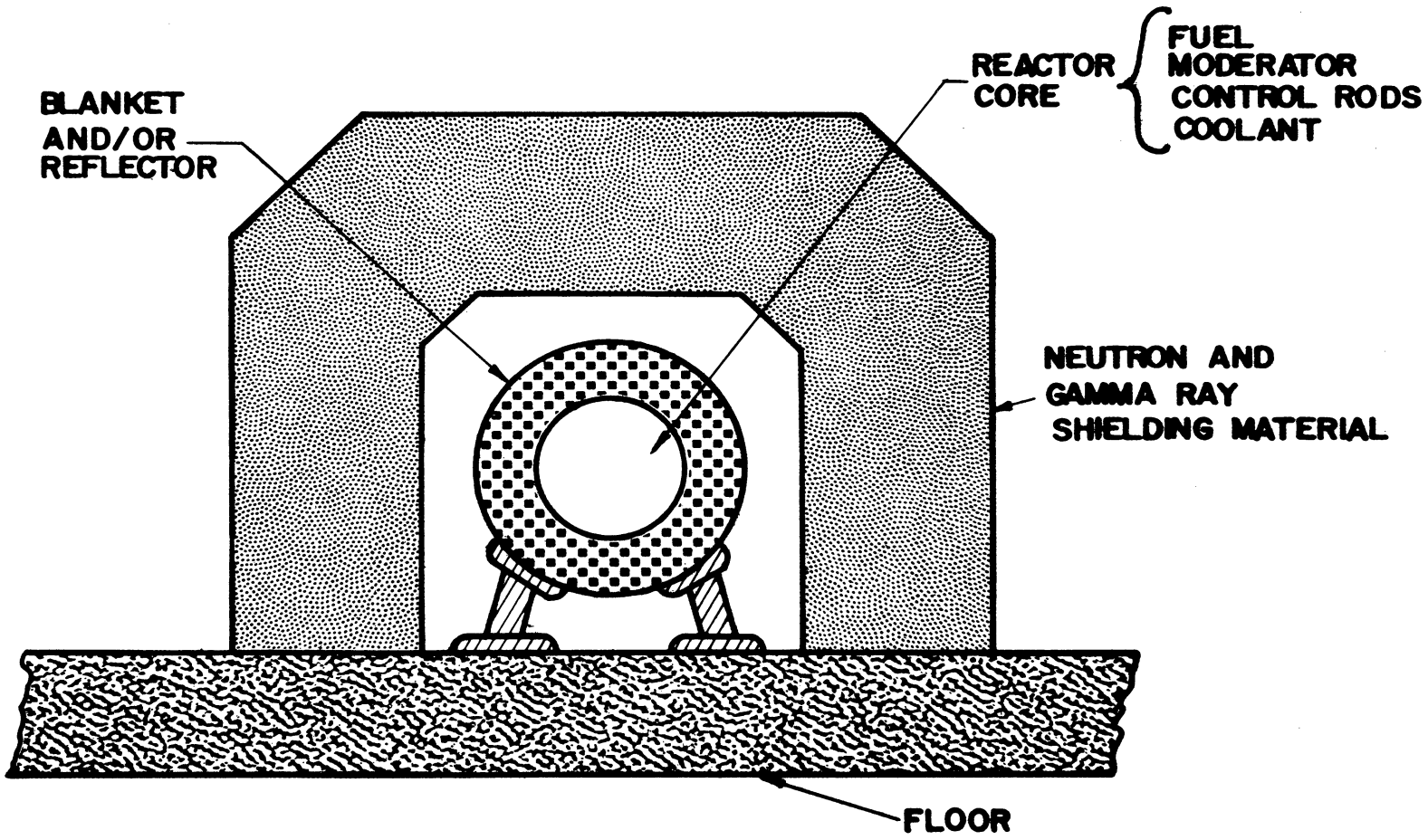


TABLE 5

SOURCES OF URANIUM

MATERIAL	SOURCE	% U	APPROX. RESERVES, TONS URANIUM*
Pitchblende	Belgian Congo	5-50	20,000
	Canada	~ 1	
	Czechoslovakia	0.2-2	
Carnotite	Siberia, Colorado	0.2-1	35,000
Gold Ore	South Africa	> 0.01	
Phosphate Rock	Florida	~ 0.01	
Bituminous Shale	Sweden	0.025	2,000,000
	Russia	0.01-0.025	
	Eastern U. S. A.	0.003-0.01	

*George W. Bain, Geology of the Fissionable Materials, Econ. Geol. 45, 274 (1950).

The process of refining pitchblende from the Belgian Congo and Canada yields $Na_2U_2O_7$ or $(NH_4)_2U_2O_7$, the latter of which may be ignited at 1000°C to give U_3O_8. Carnotite ores are more difficult to treat* than pitchblende since the low uranium concentration necessitates handling large quantities of ore to get a given yield of $Na_2U_2O_7$ or U_3O_8.

Metallic uranium may be produced from uranium compounds in one of the following ways:

a) Reduction of uranium oxides with carbon in an electric arc furnace.

b) Reduction of uranium oxides with aluminum, magnesium, calcium, or calcium hydride.

c) Reduction of uranium halides with alkali or alkaline-earth metals.

d) Electrolytic reduction of uranium halides.

e) Thermal dissociation of uranium iodide. (hot wire method.)

1. Physical Properties of Uranium Metal

Density	19.050 g/cc
Melting point	1133 ± 2°C 2051 ± 4°F
Estimated boiling point	4470°C 8100°F
Estimated heat of fusion	10 to 13 cal/gram

*For a detailed account, see "Studies of Recovery Processes for Western Uranium-Bearing Ores," V. L. Saline and K. B. Brown, AECD-3241, October, 1949.

The breaks in the specific heat and heat content curves of Figure 11 are evidence of solid state phase changes. The low temperature phase is alpha uranium, the next is the beta phase, and the high temperature phase is the gamma phase which exists up to the melting point. The temperatures at which the phase changes occur are measured by many different methods, and have some dependence on the history of the metal. Typical resistivity data yield:

Uranium Metal Phase Change Temperatures

	Heating		Cooling	
	$\alpha \rightarrow \beta$	$\beta \rightarrow \gamma$	$\gamma \rightarrow \beta$	$\beta \rightarrow \alpha$
$T^{\circ}C$	667	772	764	645
$T^{\circ}F$	1233	1422	1407	1193

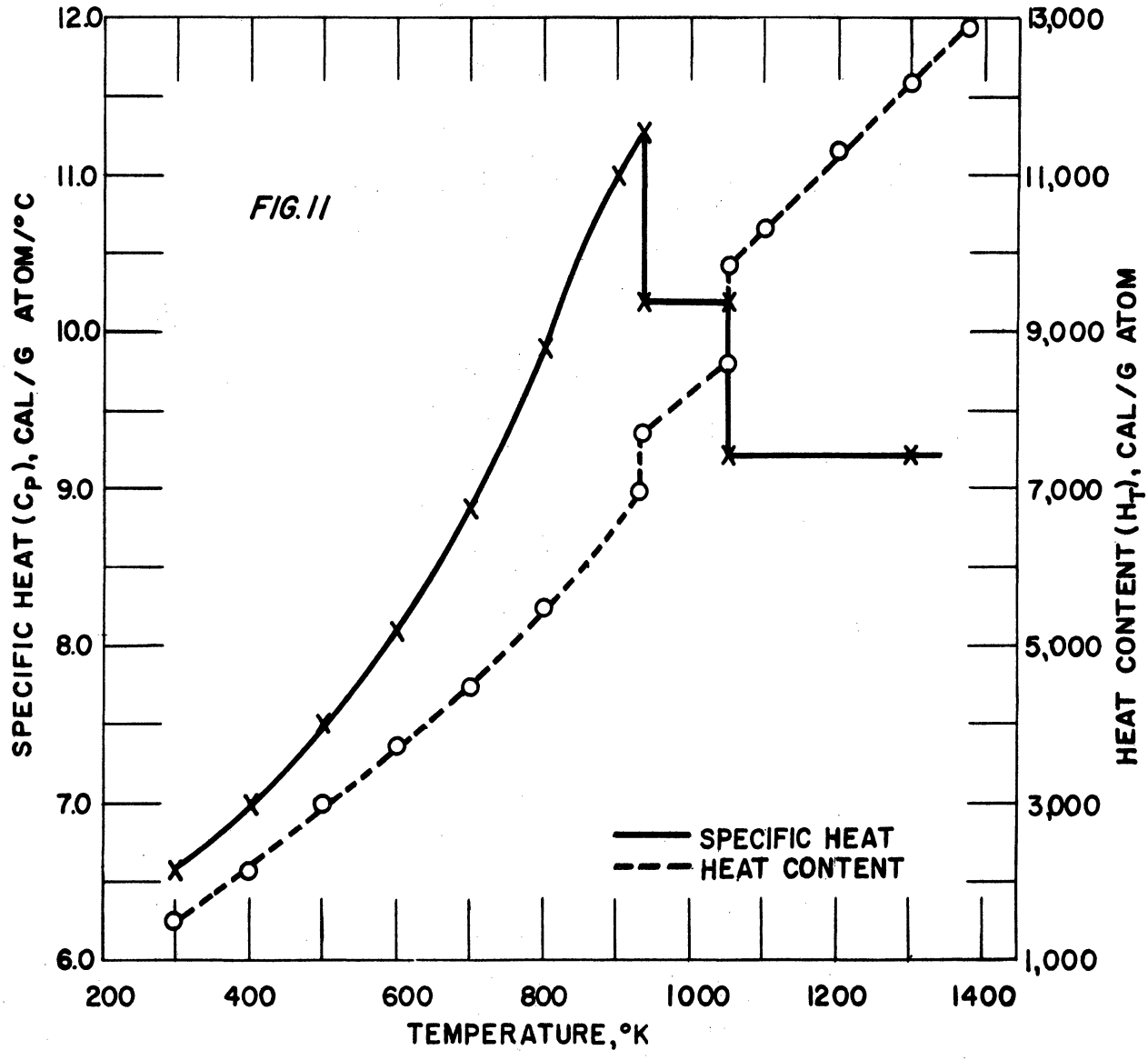
The electrical conductivity of uranium metal at room temperature is about 2×10^4 to 4×10^4 $(ohm - cm)^{-1}$, or about half the conductivity of iron. The thermal conductivity at room temperature is about 0.06 $cal/cm^2\text{-}sec\text{-}^{\circ}C$. At $300^{\circ}C$ ($572^{\circ}F$), it is 0.075 $cal/cm^2\text{-}sec\text{-}^{\circ}C$.

The tensile strength varies between 50,000 and 200,000 psi. The variation depends on the history of the metal. Uranium may be cold worked to give high tensile strengths or annealed to give lower values. The tensile strength decreases to 12,000 psi at $600^{\circ}C$ ($1112^{\circ}F$).

2. Chemical Properties

Uranium metal is highly reactive chemically. It forms hydrides with hydrogen gas at $250^{\circ}C$, and burns brightly in oxygen at about $700^{\circ}C$ ($1292^{\circ}F$). Carbides, nitrides halides, and other compounds are also readily formed. Metallic uranium also reacts with steam and the mineral

SPECIFIC HEAT OF URANIUM AT ELEVATED TEMPERATURES



From "The Chemistry of Uranium," Katz & Rabinowitch, Mc Graw - Hill Book Co. 1951.

acids. The oxidation potential of uranium is believed to be close to that of beryllium. Uranium is a strong reducing agent in aqueous solutions.

3. Nuclear Properties

The radius of the uranium nucleus is about 8.7×10^{-13} cm. For this radius, the cross-sectional area of a uranium nucleus is 2.4×10^{-24} cm^2, or 2.4 barns.

It is interesting to note that the neutron fission and capture cross sections given in Table 6 are much larger than 2.4 barns. Explanation of the anomalous result is based on wave mechanics or quantum mechanics in which particles are represented as waves. A situation analogous to neutron capture by a uranium nucleus is a sound wave incident on a small region that strongly absorbs sound waves. In this analogous problem, the absorbent region appears larger by sound absorption measurements than its physical dimensions.

A list of the uranium isotopes, their half lives, and natural abundances are shown in Table 7. The cross sections for neutron capture and fission of U^{235} are shown in Table 6.

TABLE 6

THERMAL NEUTRON FUEL METALS CROSS SECTIONS

Nuclide	U^{235}	U^{238}	Pu^{239}	Th^{232}
Cross Section, barns				
fission	549	--	664	--
neutron capture	101	2.8	361	7.0 ± 0.4
Total absorption	650	2.8	1025	7.0 ± 0.4
Average number of neutrons produced per fission	2.5 ± 0.1	--	3.0 ± 0.1	--

B. Thorium

Although natural thorium, which is very nearly 100% Th^{232}, does not fission with thermal neutrons, it captures neutrons to form Th^{233}, which is beta radioactive with a half-life of 23.3 min. decaying to protactinium-233 (Pa^{233}). Next the Pa^{233} beta decays to U^{233} with a half-life of 27.4 days. Uranium-233 fissions with slow neutrons and so natural thorium is a potential fuel material.

It is estimated that there is about three times as much thorium on the earth's surface as there is natural uranium. The principal source of thorium is presently the monazite sands found in India and Brazil. Monazite is a rare-earth phosphate mixture containing a few percent of thoria. (thorium dioxide, ThO_2.) The monazite is treated mechanically to increase the thoria concentration. It is then dissolved in sulfuric acid and after a long series of operations necessary to remove the rare-earths, thoria is obtained.

Thorium metal may then be obtained by: a) reduction of $ThCl_4$ by sodium, b) reduction of thoria by calcium metal, and c) electrolysis of $KThF_5$ in a bath of molten NaCl-KCl.

1. Physical Properties of Thorium Metal

Specific gravity		11.7
Melting point	appx.	1500°C 2700°F
Boiling point	appx.	4000°C 7200°F
Thermal conductivity at 100°C		0.076 cal/sec-cm^2-°C
Electrical conductivity at 20°C		Appx. 4×10^4 $(ohm\text{-}cm)^{-1}$
Specific heat		$Cp = 0.03437 + 0.198156 \times 10^{-5}\ T + 0.43152 \times 10^{-8}\ T^2 + 0.452056 \times 10^{-11}\ T^3$ Cal/°C

2. Nuclear Properties

The thorium nucleus is about two percent smaller in size than the uranium nucleus. The thermal neutron capture cross section is 7.0 ± 0.4 barns. The thorium isotopes are listed in Table 8.

C. The Transuranium Elements - Plutonium

The transuranium elements are those which occur as a result of the neutron irradiation of uranium. A schematic showing some of the reactions leading to the formation of some of these nuclides is shown in Figure 12. Not shown are elements 97 (Berklium, B_k) and 98 (Californium, C_f). Of these elements, plutonium has received the most attention in that it fissions under irradiation with thermal neutrons and is readily formed by irradiation of the non-fissile U^{238}.

Large quantities of Pu^{239} are produced in water-cooled graphite moderated reactors located at Hanford, Washington.* Aluminum clad uranium slugs are inserted in channels through the graphite and after a given exposure to the reactor neutron flux, are pushed out the back of the reactor by the insertion of new slugs. The exposed slugs are then treated chemically to obtain a separation of the plutonium produced from the remaining uranium and the fission product elements. This chemical separation must be performed by remote control because of the intense radioactivity of the fission products.

The nuclear reactions leading to Pu^{239} from U^{238} may be traced on Figure 12, and are specifically written down in section II-E.

The isotopes of plutonium and their modes of radioactive decay are listed in Table 9. The thermal neutron fission and capture cross for Pu^{239} are given in Table 6.

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*This plutonium plant was built during World War II as an important part of the weapons program.

TABLE VII

URANIUM FAMILY
ATOMIC NUMBER 92
ATOMIC WEIGHT 238.07

MASS NO. OF ISOTOPE	KIND OF RADIATION EMITTED	HALF-LIFE	REMARKS
227	α	1.3m.	
228	α	9.3m.	
229	α	58m.	
230	α	21d.	
231	α,γ	4.3d.	
232	α,γ	70y.	
233	α,γ	1.6×10^{5}y.	Fissions
234	α,γ	2.5×10^{5}y.	0.06% of nat. U
235	α,γ	7.1×10^{8}y.	0.71% of nat. U Fissions
236	α,γ	2.4×10^{7}y.	
237	β,γ	6.7d.	
238	α,γ	4.5×10^{9}y.	99.28% of nat. U
239	β,γ	23.5m.	
240	β	14h.	

TABLE VIII

THORIUM FAMILY
ATOMIC NUMBER 90
ATOMIC WEIGHT 232.12

MASS NO. OF ISOTOPE	KIND OF RADIATION EMITTED	HALF-LIFE	REMARKS
223	α	$<$ 1m.	
224	α	$<$ 9m.	
225	α	8.0m.	
226	α	31m.	
227	α,γ	18.6h.	
228	α,γ	1.9y.	
229	α	7300y.	
230	α,γ	80,000y.	
231	β,γ	25.6h.	
232	α	1.39×10^{10}y.	~100% of nat. Th
233	β	23.3m.	
234	β,γ	24.1d.	
235	β	$<$ 5m.	

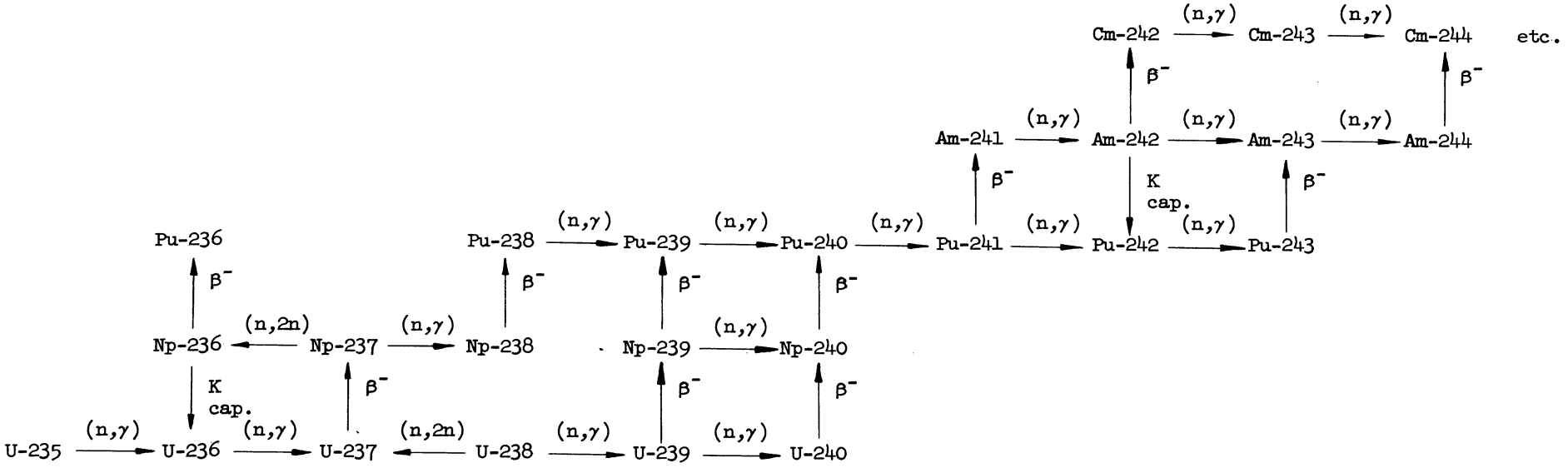
TABLE IX

PLUTONIUM FAMILY
ATOMIC NUMBER 94

MASS NO. OF ISOTOPE	KIND OF RADIATION EMITTED	HALF-LIFE	REMARKS
232	α	36m.	
234	α	9h.	
235	α	26m.	
236	α,γ	2.7y.	
237	Electron Capture	40d.	
238	α,γ	90y.	
239	α,γ	24,300y.	Fissions
240	α	6600y.	
241	β,γ	14y.	
242	α	5×10^{5}y.	
243	β,γ	5.0h.	

FIG. 12

NUCLEAR REACTIONS LEADING TO THE BUILD-UP OF THE TRANSURANIUM ELEMENTS



More detailed information on the physical, chemical, and nuclear properties of the nuclear fuels may be found elsewhere.*

D. Fuel Enrichment

Reactors for ship propulsion, as well as for other mobile nuclear power plants, must be as physically compact as possible. This necessitates the use of fuels rich in U^{235} or Pu^{239} (and perhaps in the future, U^{233}) since for enriched fuels, the critical fuel mass may be made quite small. Enriched fuel reactors in operation at present mainly use U^{235}, said to be worth about $20.00 per gram, rather than Pu^{239}, which is reported to be several times more expensive.

Since 1944, a large scale plant involved in the separation of U^{235} from natural uranium has been in operation at Oak Ridge, Tennessee. The plant output is mainly used in the weapons program. The process used is the gaseous diffusion process which makes use of the slightly different permeability of thin membranes to UF_6 (uranium hexafluoride) gas, depending on whether a U^{235} or U^{238} atom is in the UF_6 molecule involved. UF_6 gas is introduced into a diffusion cell with a membrane stretched across it. On the other side of the membrane, the pressure is somewhat lower and the gas diffuses through the membrane; a slight enrichment in U^{235} taking place in the process. A large number of diffusion cells are necessary to effect an appreciable enrichment. The cells are cascaded in the form shown in Figure 13. At the level of high enrichment, fewer cells are necessary as much less material is being handled. The entire plant at Oak Ridge is extremely large and many new techniques and materials had to

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*G. T. Seaborg and J. J. Katz, The Actinide Elements, National Nuclear Energy Series, McGraw-Hill Book Co., Inc., New York, N. Y., 1954; also, J. J. Katz and E. Rabinowitch, The Chemistry of Uranium, N. N. E. S., 1951.

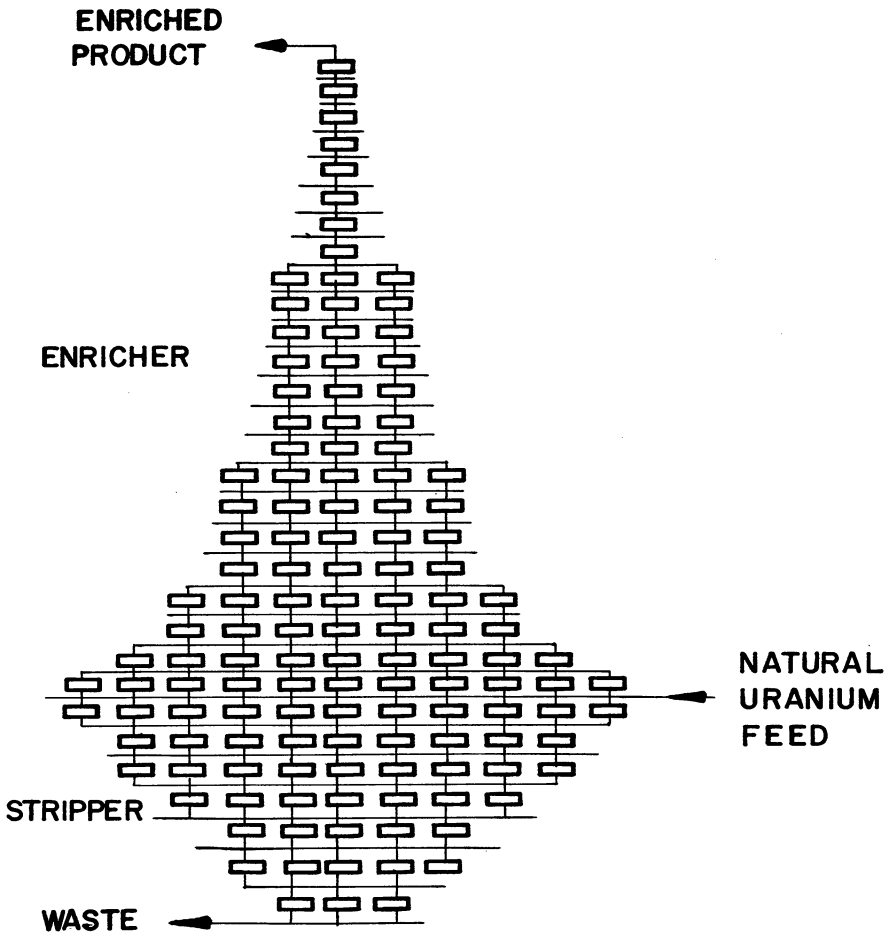


FIG. 13

GASEOUS DIFFUSION CASCADE

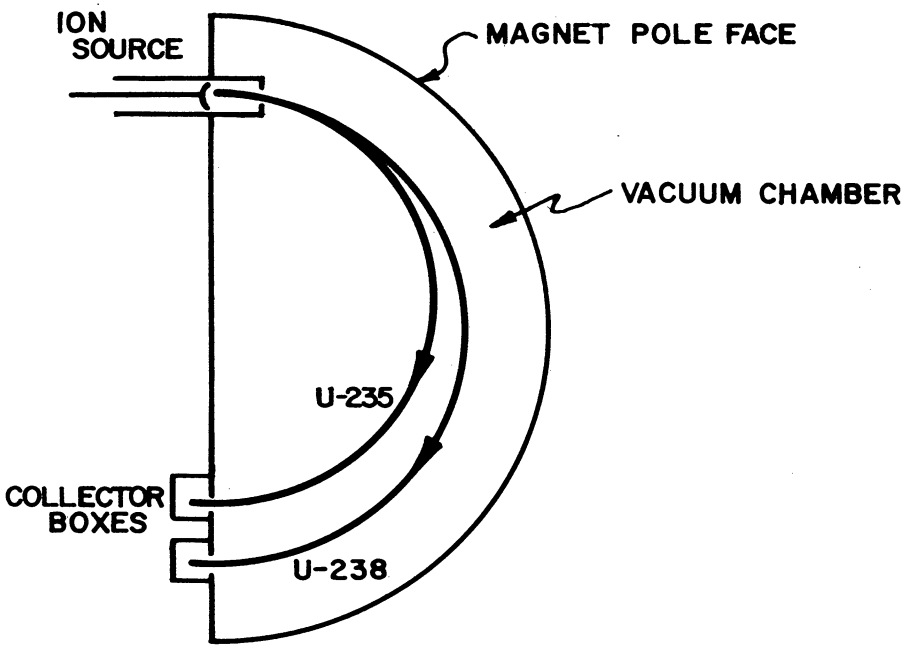


FIG. 14

ELECTROMAGNETIC ISOTOPE
SEPARATION

be developed in order to make the project an operating reality. Other gaseous diffusion plants are located at Paducah, Kentucky and Portsmouth, Ohio.

An attempt was made to develop a practical means of separating U^{235} and U^{238} on the basis of mass spectrography. This involves electrically accelerating uranium ions inside a vacuum chamber and allowing them to drift through a region in which an intense magnetic field has been produced as in Figure 14. The magnetic field causes the ions to follow circular paths where the radius of the circle depends on the mass of the ion. In this way, an excellent separation may be obtained, but the amount of material that can be handled is very small and so this technique is used only in thr production of small quantities of certain isotopes for research. This separation process is called the electromagnetic separation.

E. Fuel Breeding

Two fissile nuclides which do not occur naturally to an appreciable extent are Pu^{239} and U^{233}. These may be obtained by neutron irradiation of U^{238} and Th^{232} as follows:

$$U^{238}(n,\gamma)U^{239} \xrightarrow[23.5\ \text{min.}]{\beta^-} Np^{239} \xrightarrow[2.33\ \text{days}]{\beta^-} Pu^{239}$$

$$Th^{232}(n,\gamma)Th^{233} \xrightarrow[23.3\ \text{min.}]{\beta^-} Pa^{233} \xrightarrow[27.4\ \text{days}]{\beta^-} U^{233}$$

It is possible in principle to produce more fissile fuel than is consumed in reactors, although in practice this is difficult of attainment because many reactor neutrons are absorbed in (n,γ) reactions with poisons such as the fission product nuclides, structural materials, etc.

The reason that breeding is possible is that more than one neutron is produced in fission for each one absorbed by a fissioning nuclide. For example, if natural uranium is used as a fuel in a thermal breeder reactor, the fissioning of U^{235} yields 2.5 neutrons. Of these, a certain number are absorbed by other U^{235} nuclei in radiative capture. (i.e., in the reaction $U^{235}(n,\gamma)U^{236}$.) The percentage remaining available for other fissions and breeding is seen to be

$$\frac{\sigma_f}{\sigma_c + \sigma_f} = \frac{549 \text{ barns}}{101 \text{ barns} + 549 \text{ barns}} = 84.5\%,$$

or (.845) x (2.5) = 2.11 neutrons remaining. One of these neutrons is necessary to keep the chain reaction going and some of the remaining 1.1 neutrons may be captured by U^{238} nuclei to give U^{239}, which becomes Pu^{239} according to the above equation.

A breeder reactor is usually defined as a reactor producing more fissile fuel than it consumes, and a converter reactor produces somewhat less fuel than it consumes. Another system of nomenclature defines a breeder as a reactor that burns the same nuclide that it produces, and a converter as a reactor that burns one nuclide and produces another. In this latter nomenclature, no reference is made to the amount of fuel produced and it is necessary to state if the fuel gain factor is positive or negative. (For example, see Table 10).

It has been shown with the Experimental Breeder Reactor (EBR) at Arco, Idaho, that system 8 of Table 10 has a positive gain factor, and that the breeding principle is in fact realizable for a fast reactor. Table 10 shows many other possible fuel production schemes with predicted gain factors.

TABLE 10
SOME POSSIBLE TYPES OF POWER REACTORS*

SYSTEM	FISSILE MATERIAL	SOURCE MATERIAL	TYPE	OVER-ALL GAIN FACTOR	MATERIAL	RECYCLING OF FUEL	PRODUCT	REMARKS
1	U^{235}	U^{238}	Thermal	Negative	Natural U	No		Large U requirement.
2	U^{235}	U^{238}	Thermal converter	Negative	Natural or enriched U	Yes	Pu	U^{235} separation plant needed for recycling to save U.
3	U^{235}	Th	Thermal converter	Negative	U^{235} & Th	Yes	U^{233}	Separation plant needed for U^{235} with feed of natural U.
4	U^{233}	Th	Thermal breeder	Negative or zero(?)	Th & U^{233}(?)	Yes		May be self-maintaining. If not, feed of U^{233} is required from system 3, 6, or 8.
5	Pu	U^{238}	Thermal breeder	Negative	Pu & depleted U	Yes		Pu could be supplied by system 2, 7, or 8.
6	U^{233}	Th	Fast breeder	Positive(?) or zero	Th	Yes	U^{233}	May breed U^{233}, which could be used to supply system 3.
7	Pu	U^{238}	Fast breeder	Positive	Depleted U	Yes	Pu	Could produce U^{233} if blanket material was Th.
8	U^{235}	U^{238} or Th	Fast converter	Positive	U^{235} & U^{238} or U^{235} & Th	Yes	Pu or U^{233}	Separation plant needed for U^{235} with feed of natural U.

*C. A Rennie, Chem. Eng. Progress Symposium Series, Vol. 50, No. 12, 1954, Amer. Inst. of Chem. Eng., p. 222.

Production of Pu^{239} or U^{233} can take place in fuel elements within a reactor or in a blanket of natural uranium or thorium. The build-up of Pu^{239} or U^{233} in a sample of potential fuel material follows a curve of the form shown in Figure 15. The equilibrium value of the Pu^{239} or U^{233} concentration occurs when that rate of production is equal to the fission rate. Also shown on Figure 15 is the build-up of fission product poisons. After sometime, it will no longer be economical to continue exposure of the sample to the neutron flux because of the loss in neutron economy due to the presence of the fission product poisons. There comes the problem then of performing a chemical separation in which fission products are removed. It may also be advantageous to separate the new U^{233} or Pu^{239} fuel that has been produced from the thorium or natural uranium. Plutonium is more difficult to separate chemically from uranium than is thorium, and so it may be economically advantageous to use the thorium-uranium breeding cycle than the uranium-plutonium cycle. Plutonium is also very posionous. These chemical separations must be carried out by remote control as the fission products are intensely radioactive. It is necessary to provide a thick concrete shield around fission product separation plants. Operation may be viewed with periscopes, through thick lead glass windows (specific gravity $\simeq$6.2), or through glass walled cells containing aqueous solutions of zinc bromide (specific gravity $\simeq$2.5). A variety of remote controlled manipulators are available which permit handling of chemical equipment, metal ingots, etc. from behind shielding walls.

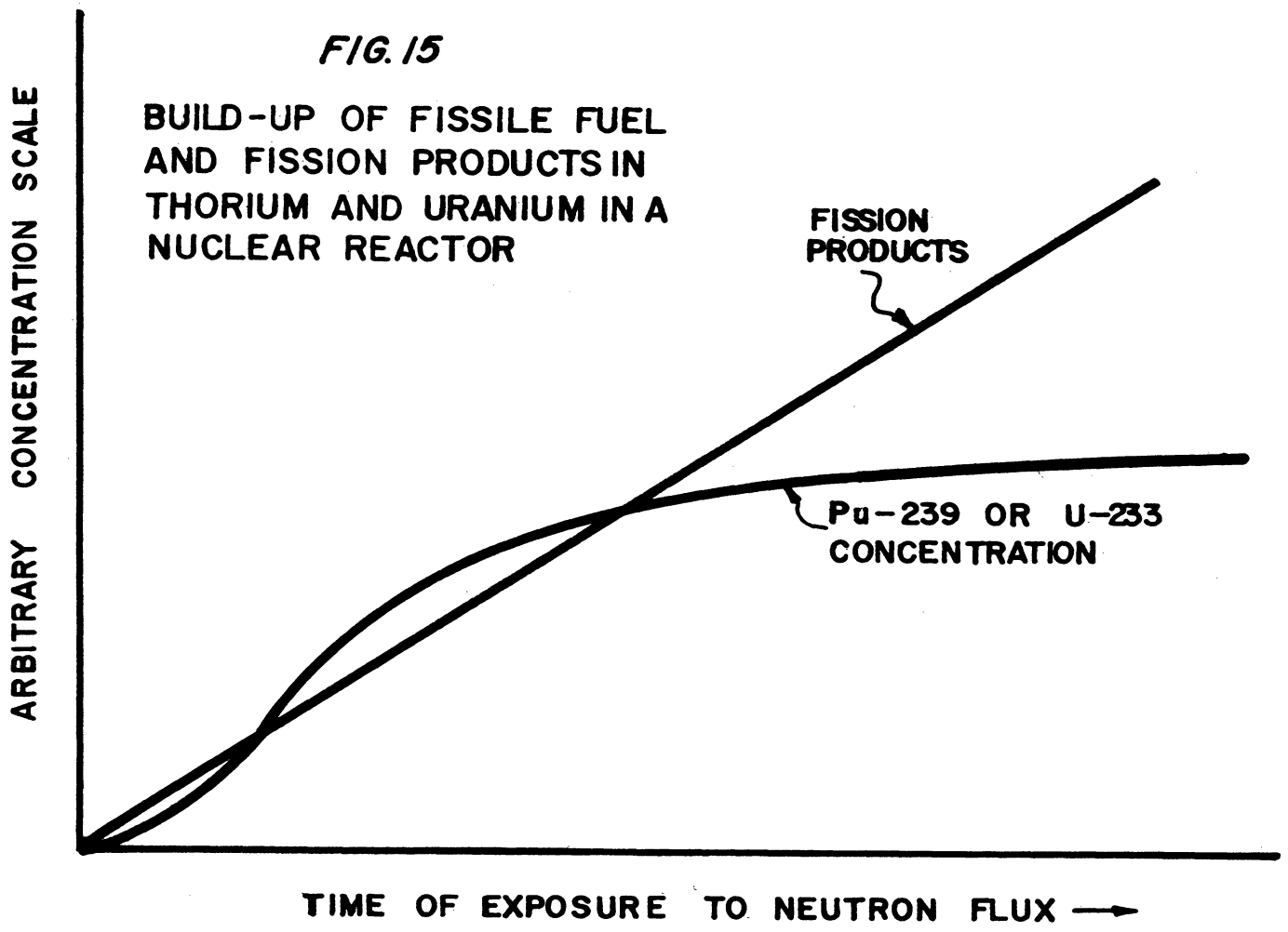
3. REACTOR CONTROL AND REACTOR MATERIALS

A. Reactor Control

In a reactor operating in a steady state condition, the effective multiplication factor k_{eff} is exactly unity, regardless of the

FIG. 15

BUILD-UP OF FISSILE FUEL AND FISSION PRODUCTS IN THORIUM AND URANIUM IN A NUCLEAR REACTOR



power level at which the reactor is operating. Reactor controls are needed to maintain the power level at a given value, and to make it possible to start the reactor and shut it down.

In discussing reactor dynamics and control, it is helpful to introduce the term reactivity. The reactivity ρ is defined as:

$$\rho = (k_{eff} - 1)/k_{eff}$$

The numerator is seen to be the neutron multiplication in excess of what is needed to maintain the chain reaction and so, if the reactivity is positive, the reaction builds up, if negative, the reaction dies out, and if $\rho = 0$, the reactor is at a steady state. Factors tending to reduce the reactivity are: fuel depletion due to burn-up, the presence of poisons, such as structural materials, coolant, and moderator, the build-up of fission product poisons,* the intentional introduction of control rod poisons, and the removal of either fuel or the neutron reflector. Reactivity has a temperature dependence because the thermal expansion of the reactor materials allows the escape of neutrons and also because at higher neutron temperatures, the absorption and fission cross sections are lower. In general, a reactor tends to shut itself down after continued operation mainly because of the accumulation of fission products.

---

*Samarium^{149} and xenon^{135} are the most serious fission product poisons encountered. Sm^{149} is a stable nuclide which results from the beta decay of Nd^{149} through:

$$Nd^{149} \xrightarrow[1.7h]{\beta^-} Pm^{149} \xrightarrow[47h]{\beta^-} Sm^{149}. \quad \text{(Stable)}$$

The fission yield of Nd^{149} is 1.4%. The thermal neutron absorption cross section of Sm^{149} is 5×10^4 barns. Xe^{135} is an intermediate in the decay chain:

$$Te^{135} \xrightarrow[2m]{\beta^-} I^{135} \xrightarrow[6.7h]{\beta^-} Xe^{135} \xrightarrow[9.2h]{\beta^-} Cs^{135} \xrightarrow[2\times10^6\,y]{\beta^-} Ba^{135} \quad \text{(Stable)}$$

The fission yield of Te^{135} is 5.6% in the thermal fission of U^{235}. The thermal neutron absorption cross section of Xe^{135} is 3.5×10^6 barns and so it is the most prominent poison produced.

Thermal reactors may be controlled with rods or plates of neutron absorbing cadmium or boron steel. These may be moved in and out of the reactor by the use of servo operated motors which can be controlled by a neutron flux meter or a temperature indicator and in this way, the flux or power level may be kept constant automatically. Manually operated control of the rods can provide for reactor start-up and shut-down operations. Boron^{10} and cadmium^{113} are used in control rods because these nuclides have high thermal neutron capture cross sections. For fast neutrons, however, these nuclides have small capture cross sections and cannot provide control of a fast reactor.

Fast reactors may be controlled by fuel motion or reflector motion. Reflector motion is more difficult because of the large mass of materials that must be moved, but can be used in emergency scram control.

It might be expected that the reactor control mechanism must be capable of extremely rapid response, as the time interval between two successive generations of neutrons is of the order of a millisecond in thermal reactors and even less in fast reactors. Fortunately, about 0.75% of the neutrons resulting from the fission of a fuel nucleus are not emitted immediately as are the rest, but are delayed by an average time of about 0.1 sec. (See Part I, Sec. 3-C for more information on delayed neutrons.) This time delay means that reactor control can be effected with slower control mechanisms if positive reactivity changes are kept less than 0.75%. If a reactor becomes critical on prompt neutrons alone (k_{eff} = 1.0075), it is said to be prompt critical, and slowly operating control mechanisms are of little use in stopping the build-up of the reaction.

B. Moderators

Thermal reactors use very low energy neutrons or thermal neutrons (average speed 7200 ft./sec.) to cause fission of the fuel. Neutrons resulting from fission, however, have much higher energies with an average value of one Mev. (average speed 45 million ft./sec., see Figure 6 for the fission neutron energy spectrum), and must be slowed down to thermal energies by substances called moderators. The neutrons are slowed down by a series of elastic collisions with moderator nuclei. Figure 16 shows a typical neutron-moderator collision. The average percentage energy loss per collision is given by $2A/(1 + A)^2$, where A is the mass number of the moderator nucleus. This function has a maximum at A = 1, and so hydrogen (A = 1) in some form is a good moderator. Other light weight nuclei given in order of increasing A and therefore decreasing moderating efficiency are: deuterium, helium, lithium, beryllium, boron, carbon, nitrogen, and oxygen. Of these, lithium and boron are unsatisfactory as they have large neutron capture cross sections and act as poisons.

Moderators in the gaseous state are inefficient, as gases have such low density that the mean free path of a neutron between collisions with moderator nuclei is so long that a gas moderated reactor would have to be very large. The ideal moderator is dense, has a large neutron scattering cross section, small neutron capture cross section and small mass number. Some moderators and their properties are listed below in Table 11.

Heavy water (D_2O) is composed of heavy hydrogen (deuterium) and oxygen. Heavy water is a very good moderator and may be used as a reactor coolant as well. At $20 per pound, heavy water might well find use in

FIG. 16

ELASTIC COLLISION
OF A NEUTRON WITH
A MODERATOR NUCLEUS

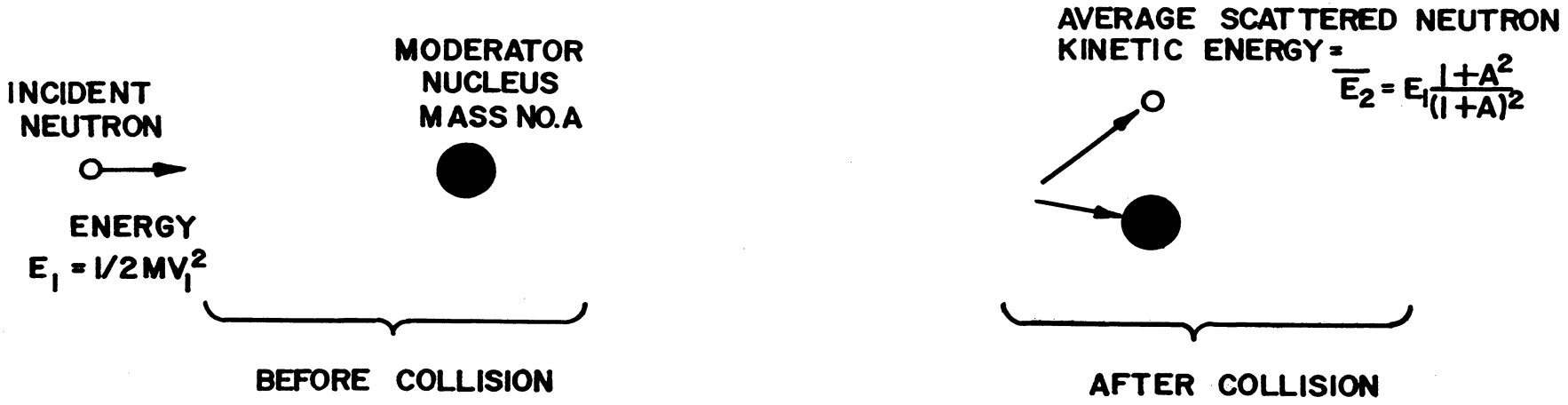


TABLE 11

PROPERTIES OF MODERATORS

Material	H_2O	D_2O	Be	C	BeO
Density	1.00	1.10	1.84	1.60	2.80
Atomic or molecular weight	18	20	9	12	25
Atoms/cm^3 or molecules/cm^3	3.35×10^{22}	3.32×10^{22}	1.23×10^{23}	8.05×10^{22}	6.75×10^{22}
σ_a at 0.025 ev, barns	0.66	0.92 mb	9 mb	4.5 mb	9.2 mb
σ_s at 0.025 ev, barns	110	15	6.9	4.8	11.1
Epithermal σ_s, barns	46	10.5	6	4.8	9.8
Moderating ratio	67	5820	160	169	180
Slowing down length, cm	5.7	11.0	9.9	18.7	12.0
Slowing down time, sec	10^{-5}	4.6×10^{-5}	6.7×10^{-5}	1.5×10^{-4}	7.8×10^{-5}
Albedo (infinite)	0.82	0.97	0.89	0.93	0.93

the competitive nuclear power field. Heavy hydrogen occurs naturally with an isotopic abundance of 0.015%, and may be separated from natural hydrogen by either distillation processes or chemical exchange reactions. Mass spectrography is not a practical method since such large quantities of material must be handled to get an appreciable yield of deuterium.

Of the possible processes listed in Table 12, the fractional distillation of liquid hydrogen at -252.7°C offers great promise since the vapor pressure ratio is high. If there is sufficient external demand for liquid hydrogen, utilization of this process could bring the price of heavy water down to a reasonable value. The two other distillation processes in Table 12 require many separation stages to yield an appreciably enriched product. The steam distillation requires an economical source of heat. Consideration is being made of using the heat from natural hot springs in New Zealand in this process.

The first of the exchange reactions listed in Table 12 is used at Trail, British Columbia and in Norway in the gas phase. The large amounts of hydrogen gas needed in the separation is eventually used in the synthesis of ammonia. The second of the exchange reactions of Table 12 using hydrochloric acid has promise of becoming an economical method as it can be carried out in the liquid phase without catalysis.

Ordinary water is being used as a moderator in many reactors. It is necessary to use enriched fuels in water moderated reactors because many neutrons are captured by the hydrogen in the water. Swimming pool reactors and water boiler reactors use ordinary water as a moderator.

Graphite is employed in many thermal reactors today. It is necessary to obtain very pure graphite for this service. The presence of

TABLE 12

DEUTERIUM SEPARATION PROCESSES

DISTILLATION	N.B.PT., °C.	VAPOR PRESSURE RATIO
H_2O - HDO	100	1.017
NH_3 - NH_2D	- 33.4	1.037
H_2 - HD	-252.7	1.73

EXCHANGE REACTIONS	EQUIB. CONST. AT 25°C
$H_2O + HD \rightleftharpoons HDO + H_2$	3.70
$H_2O + DCl \rightleftharpoons HDO + HCl$	4.69

even small traces of boron is not allowable because of the large neutron capture cross section of boron. This impurity may be removed by passing freon gas through hot graphite.

Beryllium as BeO is a good moderator material, but presesnt costs are high. BeO can be pressed to form bricks with a specific gravity of 2.8. Beryllium oxide is used as a reflector material in the Los Alamos water boiler reactor.

C. Coolants

The problem of removing heat from a reactor has resulted in the development of many new materials, mechanisms and techniques. Most of the reactors in operation today are water cooled and a few are air cooled, but liquid metal coolants are viewed as being much more satisfactory for the removal of heat from large power reactors now being planned.

In reactors built to produce neutron beams for physics research or radioisotope production, the heating of the reactor core is in general a nuisance and operation at a low temperature is usually desired as it simplifies the design and operation considerably. Where the reactor is to be used in electric power production using a heat engine, high operating temperatures are preferable to give high thermodynamic efficiencies.

Extracting reactor heat poses problems different from those encountered in coal or oil burning power plants. The heat produced per unit volume in a reactor can be made very high to keep the fuel inventory and shielding requirements low with the result that the heat transfer surfaces are small and require high heat transfer coefficients. Further, the primary coolant must have a small neutron capture cross section. Liquid sodium is presently receiving a great deal of attention and most proposed

power reactors plan on using sodium as a coolant, as it satisfies many of these requirements. In this plan, sodium is pumped at high velocity through the reactor core to a heat exchanger where the heat is removed by steam or by some intermediate coolant. An electromagnetic pump has been developed which requires no seals or moving parts, and considerably simplifies handling a liquid metal. These pumps operate by passing an electric current across the pipe containing the liquid metal. A force is exerted on the liquid metal by the interaction of a strong megnetic field which is arranged so that it is mutually perpendicular to the direction of the axis of the pipe and the direction of the electric current. Resistive losses in the liquid metal and the magnet coils reduce the efficiency of these pumps considerably. They operate best on direct current. Another type is designed to use alternating current, if a further reduction in efficiency can be tolerated.

An alloy of sodium and potassium which melts at room temperature is another candidate for coolant service. It has the disadvantage of a higher neutron capture cross section than pure sodium, but otherwise is quite satisfactory.

For high temperature homogeneous reaction, a solution of uranium in bismuth in power reactors is possible. The uranium is used as the reactor fuel and the bismuth is a carrier. This metal solution is pumped from the reactor to a heat exchanger and then back to the reactor. Bismuth has a low neutron capture cross section and does not attack graphite which may be used as a moderator in this type of reactor.

Heavy and ordinary water may be used as primary coolants in homogeneous reactors; the fuel being a uranium salt. The water then also serves as a moderator.

The properties of some coolants are given in Table 13 and in Figure 17. More extensive information on the properties of liquid metals may be found in Liquid Metals Handbook, R. N. Lyon, ed. (obtainable from U. S. Government Printing Office).

D. Structural Materials

Consideration of the choice of reactor structural materials depends on the size and type of reactor to be built, the intended service, and the operating temperatures. Thermal reactors allow use of only those materials with low thermal neutron absorption cross sections. This restricts the number of possible structural materials which may be used in thermal reactors considerably, although fast reactors permit choice from a greater variety of materials in this respect. The economic design of reactors which are to operate at high temperatures narrows the field of structural materials since many metals which are satisfactory at low temperature (such as aluminum, for example) have melting points too low for high temperature use.

Among the factors to be considered in choosing reactor materials are: mechanical quantities such as tensile strength, hardness, ductility, etc.; chemical properties such as corrosion resistance and chemical reactivity with the coolant, moderator, fuel, etc.; thermal properties like the melting point, specific heat, heat conductivity, the thermal coefficient of expansion, and solid state phase transitions; nuclear properties such as the neutron absorption and scattering cross sections (the specific gravity weights the effectiveness of these cross sections); radiation damage, which is the effect on the chemical and mechanical properties of the material on exposure to neutrons, beta and gamma rays; and there is, of course, the very important matter of cost. Table 14 shows many of these properties for some reactor materials.

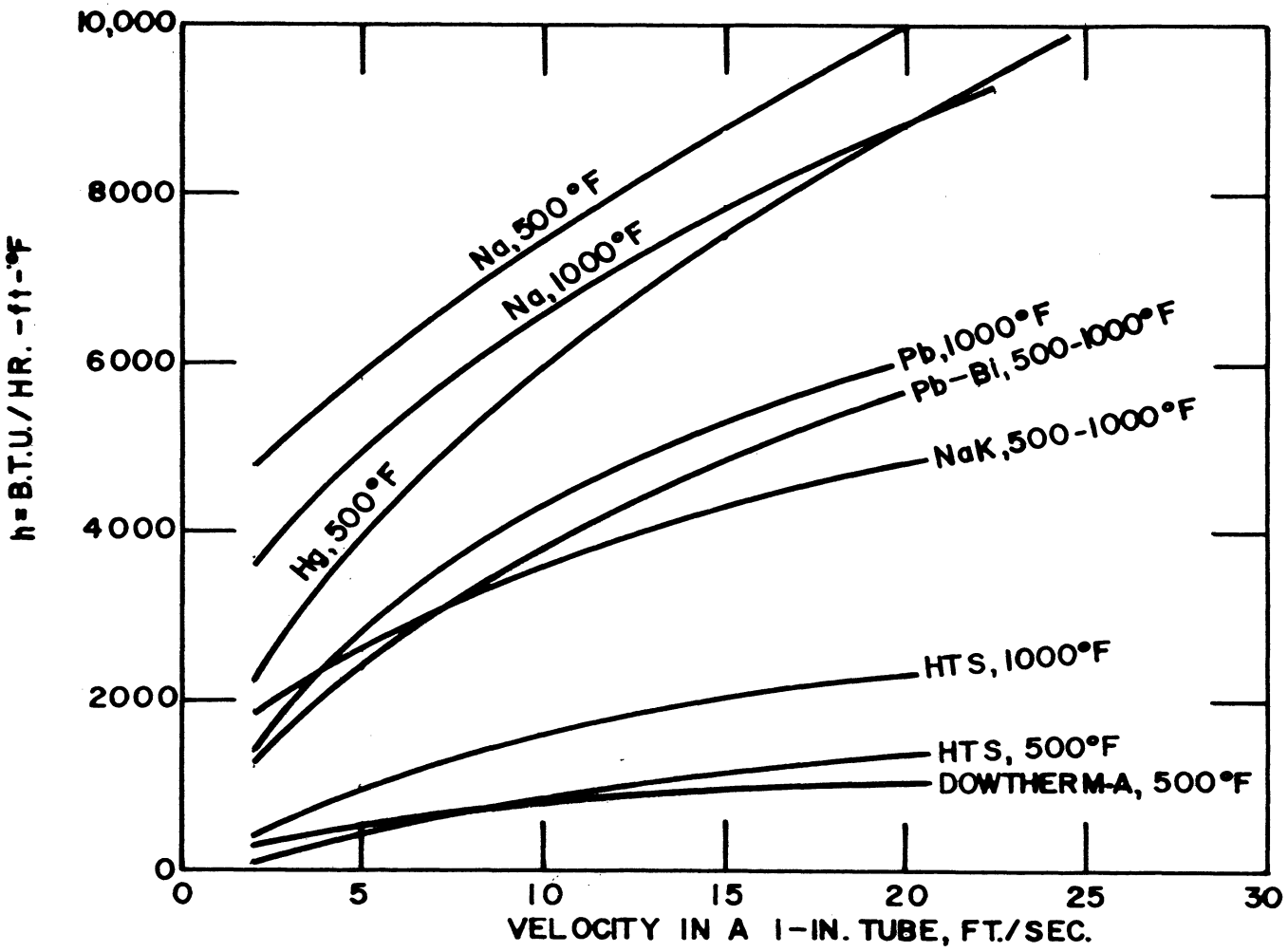
TABLE 13

PROPERTIES OF COOLANTS

Coolant	Thermal Neutron Cross Sections: Microscopic (barns) Absorption	Thermal Neutron Cross Sections: Microscopic (barns) Scattering	Thermal Neutron Cross Sections: Macroscopic Absorption Cross Section at 650°F (cm^{-1})	Melting Point °F	Boiling Point °F	Operating Pressure Psia	Density (lb/ft^3) 100°F	Density (lb/ft^3) 500°F	Density (lb/ft^3) 1,000°F	Specific Heat (Btu/lb-°F) 100°F	Specific Heat (Btu/lb-°F) 500°F	Specific Heat (Btu/lb-°F) 1,000°F
H_2O	0.602	164	0.0079	32	212	14.7	62.0			0.9976		
						1500		48.6			1.165	
D_2O	0.92 mb	15.3	0.000037	38.87	214.7							
Na	0.45	4	0.0074	208	1621	14.7		55.3	51.1		0.3150	0.3005
K	2.5	1.5	0.0166	147	1400	14.7		48.7	44.6		0.19	0.18
Na-K alloy (44%K)	1.1	3.2	0.0113	66.2	1518	14.7	5.61	52.9	48.7	0.2733	0.2583	0.2486
Bi	0.015	9.0	0.00059	520	2691	14.7			608			0.0369
Bi-Pb alloy (44.5% Pb)	0.17	9.9	0.0021	257	3038	14.7		646	624		0.035	0.035

FIG. 17

COMPARISON OF FLUIDS FOR HEAT TRANSFER



From "Liquid Metals Handbook"
R. N. Lyon, editor NAVEXOS p-733 (Rev.)

TABLE 14

PROPERTIES OF STRUCTURAL MATERIALS

ELEMENT	THERMAL NEUTRON ABSORPTION CROSS SECTION (Barns)	DENSITY (Grams/ cm^3)	MELTING POINT (^oC)	SPECIFIC HEAT (Cal./gm $-^oC$)	THERMAL EXPANSION COEFFICIENT (Per $^oC \times 10^6$)	MODULUS OF ELASTICITY (10^6 psi)	HARD-NESS (BHN)	TENSILE STRENGTH (10^3 psi)	
								Annealed	Cold Worked
Aluminum	0.22	2.70	660.2	0.22	24	10	20-25	13	24
Beryllium	0.010	1.85	1,300	0.5	12	42	110	45	--
Magnesium	0.059	1.74	651	0.25	--	6.5	50	32-46	32-50
Molybdenum	2.4	10.2	2,620	0.065	5.5	40-50	147	100	250
Nickel	4.5	8.9	1,455	0.11	13	30	75	47	125
Tantalum	21	16.6	2,996	0.036	6.5	27	75-125	50	125
Titanium	5.8	4.5	1,725	0.129	8.5	16.8	200	80	122
Vanadium	4.8	6.02	1,735	0.12	--	20-22	260	--	--
Tungsten	19	19.3	3,410	0.034	4.0	50-60	260	50	300
Zirconium	0.18	6.5	1,830	--	5.0-5.8	12	--	35	85
18-8 Stainless Steel (Fe, Cr, Ni, Mn, Nb)	2.9	7.92	1,400-1,420	0.12	16.7	29	160	90	--
Inconel "X" (Ni, Cr, Fe, Ti, Nb)	4.1	8.3	1,400-1,420	0.11	13.9	31	200-400	160-180	--

In regions in the reactor where the neutron flux is very high, some of the more expensive reactor materials, such as zirconium, may be used and then more reasonably priced materials employed outside the core. The use of small, high temperature reactors may provide a saving on the expensive materials. At any rate, in the overall reactor design, a compromise must be made on reactor size, operating temperature, fuel inventory, etc., and a very important factor in arriving at a sensible compromise is the properties and cost of structural materials.

E. Radiation Damage

Radiation damage is the term applied when the mechanical, chemical, and nuclear properties of a substance are altered by exposure to neutrons, beta and gamma rays, and fission fragments. Organic compounds suffer greatly in exposure to all these radiations. This means that hoses, hydraulic fluid, organic electric insulators, plastics "Dowtherm", etc., may not be used in regions where neutron flux or radioactivity are high. Water is readily dissociated under irradiation and provisions have to be made to re-combine the hydrogen and oxygen either through the use of a catalytic agent or direct burning. (Dissociated water is highly corrosive to metals at high temperatures.) Deuterium, either as heavy water or deuterated compound, suffers slightly through the photodissociation of the deuteron into a neutron and a proton by reactor gamma rays. Ceramics and glasses are more resistent, but most resistant of all are the metals, mainly because of the lack of covalent bonds. Where fast particles strike a metal, atoms are displaced from their original position. This leads to a change in conductivity, creep, hardness, etc. These changes may be corrected by annealing. Radiation damage has the effect of cold working the metal,

except that the radiation effects are nondirectional and the radiation induced hardenings is not as great as occurs in the case of cold working. A specifically nuclear effect that weakens the metal is neutron capture reactions which may eventually result in the formation of atoms of new elements. Carried to the extreme, this eventually has the effect of weakening the metal due to the presence of impurities.*

F. Shielding

Reactors must be shielded to protect operators and other personnel from the intense neutron and gamma ray fluxes that issue from the reactor core. In addition, shielding performs the important service of protecting organic compounds in gaskets, hoses, etc., from radiation damage. It is also important that the steam driving a turbine be shielded to prevent dissociation of the water into free oxygen and hydrogen peroxide which are highly corrosive at the temperatures of superheated steam.

1. Charged Particles

The charged particles which result from radioactive decay are most easily stopped and require the least consideration in shielding calculations. Alpha particles such as occur in the radioactive decay of heavy nuclides, may be stopped in a few centimeters of air or a few thousandths of an inch in metals. Beta particles are somewhat difficult to stop but do not present a very difficult problem. The range of betas in some materials is plotted vs. energy in Figure 18.

Gamma rays and neutrons are the most difficult to stop of all. The reason for this is that they are uncharged and there is

*For a more detailed account of the effects of radiation damage to metals and non-metals, see Nucleonics, September, 1954.

no electrostatic interaction with the shielding material which in the case of charged particles, provides the slowing-down mechanism.

2. Gamma Rays

When the radiation from a point source is absorbed catastrophically, i.e., the particle or gamma ray is lost entirely from the flux by a single collision or event, the law which describes the decrease in intensity with distance is:

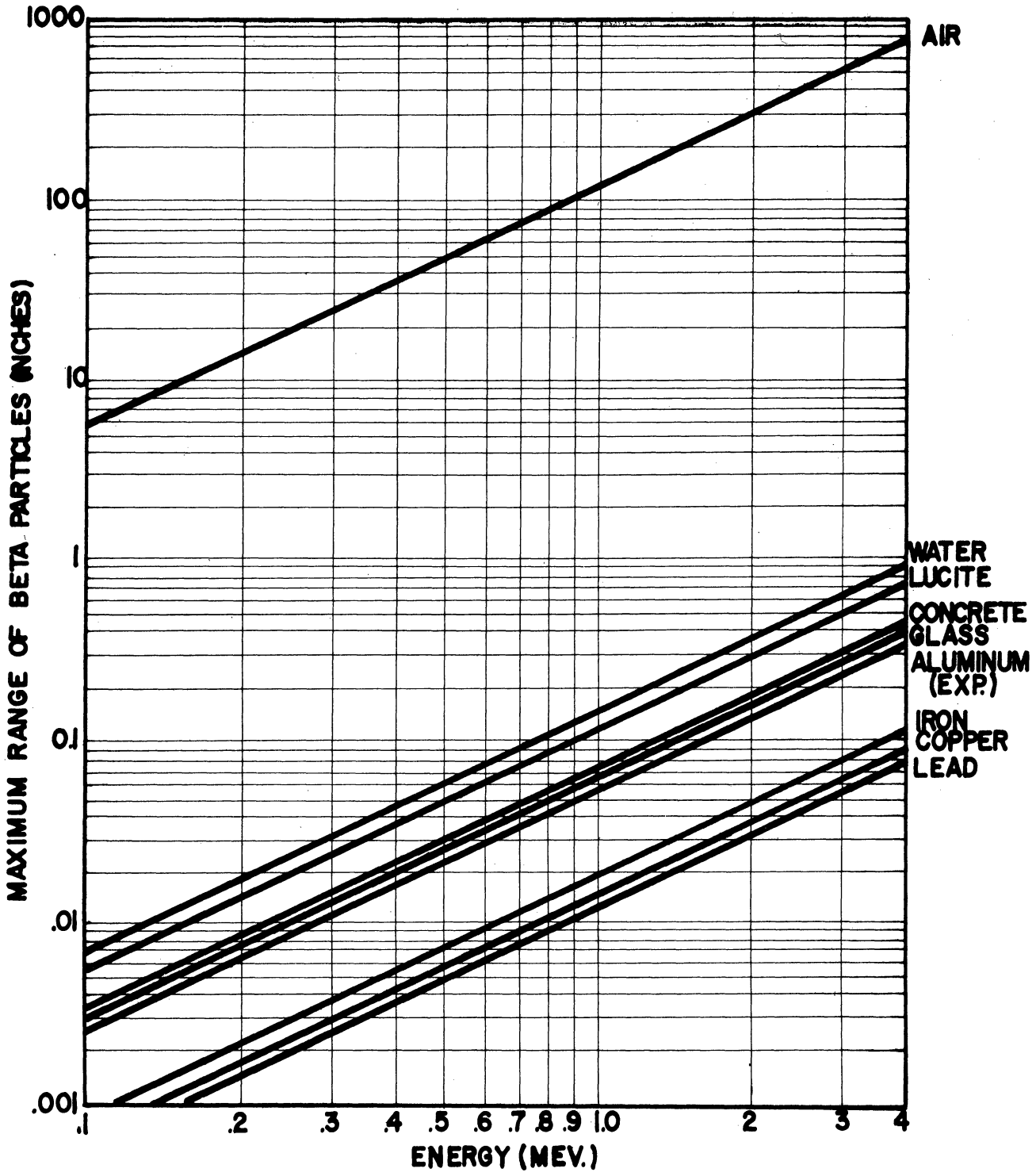
$$I = \frac{S}{4\pi \chi^2} e^{-\Sigma \chi_o} \qquad 2)$$

where S is the point source strength in particles per second, I is the flux in particles per square centimeter per second at a distance χ from the source, χ_o is the thickness of the shielding material measured along the line between the source and the point at which the flux I is to be determined, and Σ is the macroscopic cross section for catastrophic absorption of the radiation or particle. Σ is given by $N\sigma$, where σ is the cross section for the absorption due to a single atom and N is the number of such atoms per unit volume. (Units must be chosen so that the product $\Sigma\chi_o$ is dimensionless) N is given by $(\frac{N_o d}{A})$ where $N_o = 6.02 \times 10^{23}$ (Avogadro's number), d is the density of the absorbing medium and A is its atomic weight. From this shielding law, it is seen to be advantageous to have the product $\Sigma \chi_o$ large for good shielding.

If source of radiation is not a point source but is distributed in space, we must add the contributions from all the radiation source material. This means we must compute:

FIG. 18

THE MAXIMUM RANGE OF BETA PARTICLES AS A FUNCTION OF ENERGY



From Stanford Research Institute
Report No. 361 "The Industrial
Uses of Radioactive Fission Products"

$$I = \frac{1}{4\pi} \iiint \frac{\rho(r)}{\chi^2} e^{-\Sigma \chi_0} \, dv \qquad 3)$$

where $\rho(r)$ is the source density in particles emitted per second per unit volume at the point r, χ is the distance from r to the point at which I is measured, and dv is the differential volume element. Evaluation of the integrals involved in particular source configurations is generally difficult or impossible to do exactly, but may be approximated numerically.

For gamma rays, the cross section σ for absorption depends on the energy of the gamma involved and on the shielding material. Materials with large atomic numbers are more effective as shields since σ is larger. Below in Table 15 is a list of values of Σ in cm^{-1} for three common shielding materials.*

Some useful numbers which point out the relative effectiveness of shielding materials are the thicknesses required to reduce the intensity of 2 Mev gamma rays by a factor of 10. This intensity reduction is produced by 48 cm of water, 25 cm of concrete, 6.7 cm of iron, or 4.3 cm of lead.

The gamma rays from a reactor can be roughly computed using Table 16. The gamma rays considered include the prompt gammas,

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*The Science & Eng. of Nuclear Power, Vol. II, C. Goodman, ed., Addison Wesley Press, Inc., 1949, p.201.
More extensive data may be found in Radiological Health Handbook, obtainable from Public Health Service, Cincinnati, Ohio, p. 124; Experimental Nuclear Physics, Vol. I, E. Segre, ed., John Wiley & Sons, Inc., New York, 1953, p. 309; Introduction to Nuclear Engineering, R. Stephenson, McGraw-Hill, 1954, Chapter 5.

TABLE 15

GAMMA RAY ABSORPTION COEFFICIENTS IN CM^{-1}
FOR IRON, LEAD, AND CONCRETE

(cm^{-1})

Energy (Mev)	Fe	Concrete	Pb
0.04	25	7	
0.06	10	1.05	
0.08	4.8	0.55	
0.10	3.1	0.45	75
0.20	1.3	0.27	17.5
0.40	0.7	0 20	4.5
0.60	0.63	0.17	2.1
0.80	0.52	0.14	1.32
1.00	0.47	0.11	1.0
2.00	0.35	0.09	0.53
3.00			0.45
4.00			0.45
6.00			0.5
8.00			0.56

TABLE 16

REACTOR GAMMA RAYS

Energy Interval (Mev)	No. of Gammas Per Fission	Total Energy in Interval (Mev)
0-2	9.31	9.31
2-4	0.75	2.25
4-6	0.099	0.495
6-8	0.0154	0.1078
8-10	0.0029	0.0241

fission product gammas and also fuel capture gammas.* The number of gamma photons produced per second is proportional to the number of fissions per second or the power level. Figure 19 shows the relation between the fission rate and power level.

3. Neutrons

The neutron flux emerging from a given reactor is proportional to the power level at which the reactor is operating. Reactor construction and geometry define how great this flux will be, and there is no simple relationship that can be stated between reactor size and the neutron leakage flux. The history of typical neutrons emerging from a reactor is of interest. A slow neutron will be captured after traversing a small thickness of shielding material, one or more capture gamma ray photons being emitted in the process. A fast neutron may be captured while it is still fast or it may be slowed down by the moderating action of the shielding material and eventually be captured as a slow neutron. A conservative estimate of the neutron shielding required may be made if the moderating property of shielding material is neglected and only fast neutron capture is considered. This is a conservative estimate since fast neutron cross sections of elements are much smaller than if taken at thermal energies. Since fast neutron capture is a catastrophic process, the same law given above for gamma rays applied except that Σ now refers to neutron capture by the shielding material and is given by $(N \sigma_a)$ where σ_a is the neutron absorption cross section in barns. Some values of Σ are shown in Table 17.

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*From Introduction to Nuclear Engineering by R. Stephenson, McGraw-Hill Book Co., Inc., New York, N. Y., 1954, p. 209.

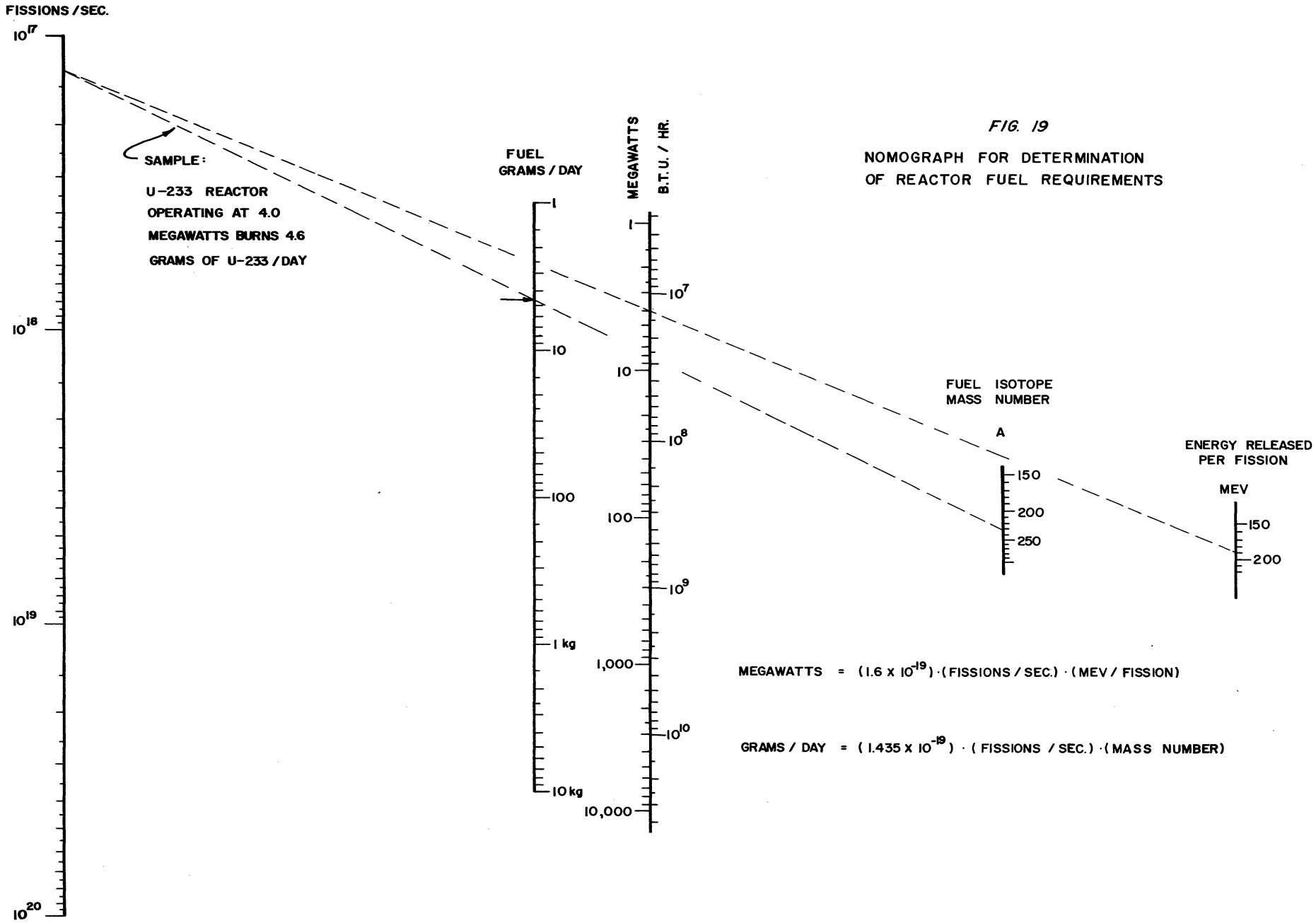

FIG. 19

NOMOGRAPH FOR DETERMINATION
OF REACTOR FUEL REQUIREMENTS

TABLE 17

NEUTRON SHIELDING DATA

Neutron Energy	Shielding Material	Σ cm^{-1}	Thickness to reduce Intensity by 1/10, cm.
1 Mev	Hydrogen (in water)	0.281	8.2
	Oxygen (in water)	0.268	8.6
	Lead	0.178	13
10 Mev	Hydrogen (in water)	0.064	36
	Oxygen (in water)	0.050	46
	Lead	0.165	14

Tables such as this are easily constructed using neutron cross section data such as presented in "Neutron Cross Sections" AECU-2040.

A nucleus which captures a neutron usually emits gamma rays which must be considered in the shield design. Ideally, a material such as concrete composed of light to medium weight nuclei to slow down neutrons should be used in conjunction with a heavy metal for gamma shielding where the heavy metal is dispersed in the concrete. Some shielding concretes made up in this manner are listed in Table 18.

The neutron and gamma ray fluxes allowable outside the shielding must be less than certain maximum allowable values which are described later in this section.

A problem to be considered is the heating of the shield by absorption of the neutrons and gammas. Sometimes a water cooled lead shield, close to the reactor core, is used to absorb a large fraction of the energy so produced. A concrete shield outside then reduces the flux down to a safe level.*

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* A description of the shield heating is given in Introduction to Nuclear Engineering by R. Stephenson, p. 217.

TABLE 18

HEAVY-AGGREGATE CONCRETES AND OTHER SHIELDING MATERIALS

	Cement	Aggregate	Density (gm/cm^3)
CONCRETES	Portland	Sand, gravel	2.3
	Portland	Barytes	3.5
	Portland	Iron punchings	6
	Portland	Barytes-colemanite	3.2
METALS	Cast Iron		7.85
	Lead		11.2
	Uranium		19.0
	Thorium		11.4

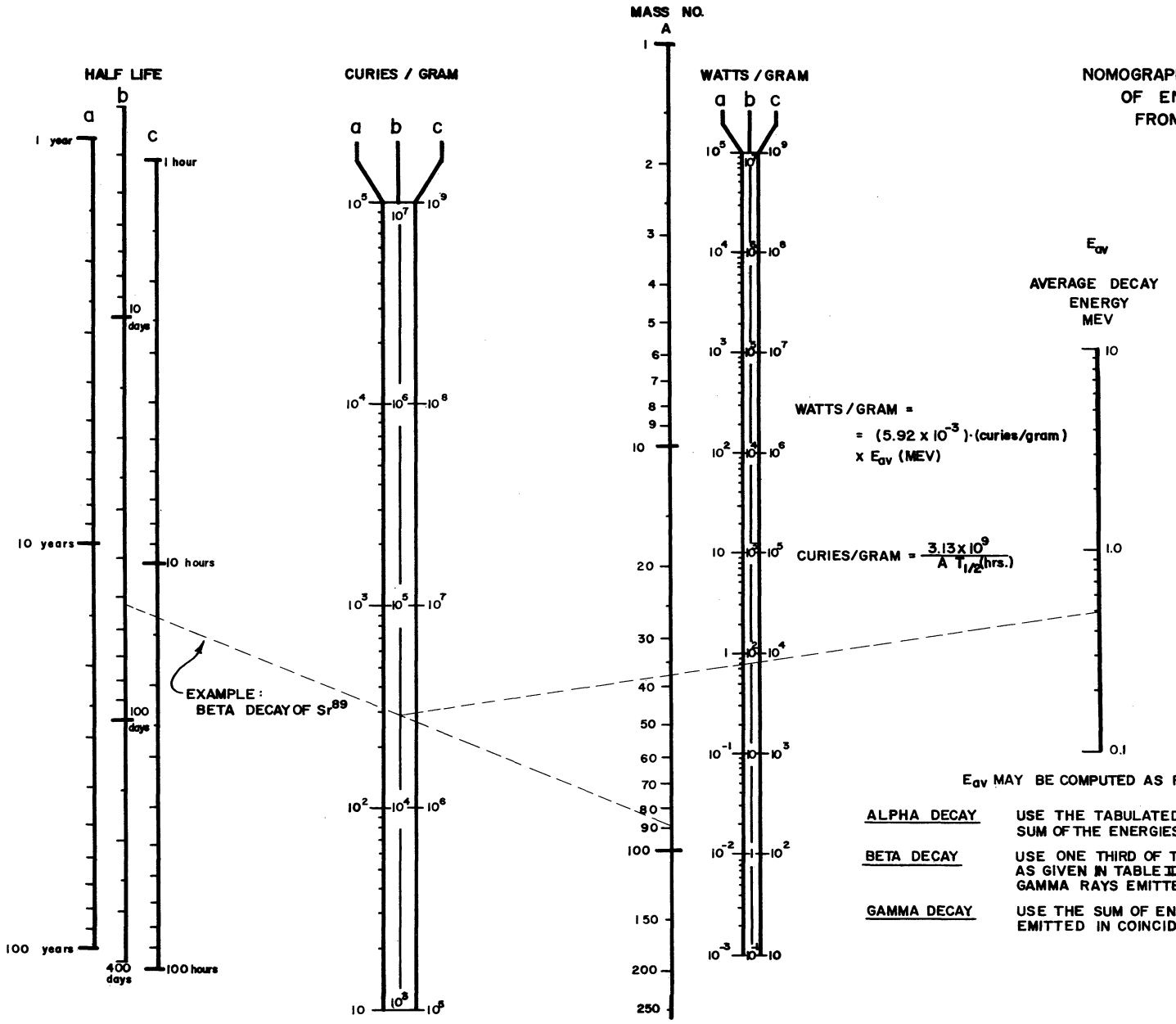
4. Dosimetry

The curie is that amount of radioactive material in which 3.7×10^{10} disintegrations occur per second. Knowledge of the half-life of a radioactive nucleus permits calculation of the number of curies per gram of the radioactive nuclide in question. The nomograph of Figure 20 shows how this is done. It is possible to convert the fission product yields given in Figure 7 to curies using this nomograph and the known half-lives of the fission product nuclides such as given in Table 2. Where gamma rays are emitted in the radioactive decay process, we may obtain the photon flux in $(\text{photons})/(\text{cm}^2\text{-sec.})$ from the number of curies in a sample through use of Equation 2. Next, Figure 21 may be used to compute the dose rate in roentgens per hour at a given distance from the source.

Consideration of the amount of gamma radiation or neutron flux that the human body can tolerate without producing sickness or more permanent damage is the basis on which all reactor shields must be designed. A detailed treatise of the present status of the allowable dose of a penetrating radiation is beyond the scope of this

FIG. 20

NOMOGRAPH FOR CALCULATION OF ENERGY AVAILABLE FROM RADIOACTIVE NUCLIDES



IN USING THIS NOMOGRAPH, CARE MUST BE TAKEN TO BE CONSISTENT IN USING THE SAME SET OF SCALES (a, b, or c) THROUGOUT A GIVEN CALCULATION. FOR EXAMPLE, THE HALF LIFE OF Sr^{89} IS 53 DAYS, ENTER COLUMN b AND USE COLUMN b IN READING CURIES/GRAM AND WATTS/GRAM.

E_{av} MAY BE COMPUTED AS FOLLOWS:

ALPHA DECAY	USE THE TABULATED ALPHA PARTICLE ENERGIES PLUS THE SUM OF THE ENERGIES OF ANY GAMMA RAYS EMITTED IN COINCIDENCE.
BETA DECAY	USE ONE THIRD OF THE TABULATED DATA DECAY ENERGIES SUCH AS GIVEN IN TABLE II. ADD THE SUM OF THE ENERGIES OF ANY GAMMA RAYS EMITTED IN COINCIDENCE.
GAMMA DECAY	USE THE SUM OF ENERGIES OF THE GAMMA RAYS WHICH ARE EMITTED IN COINCIDENCE IN A CASCADE.

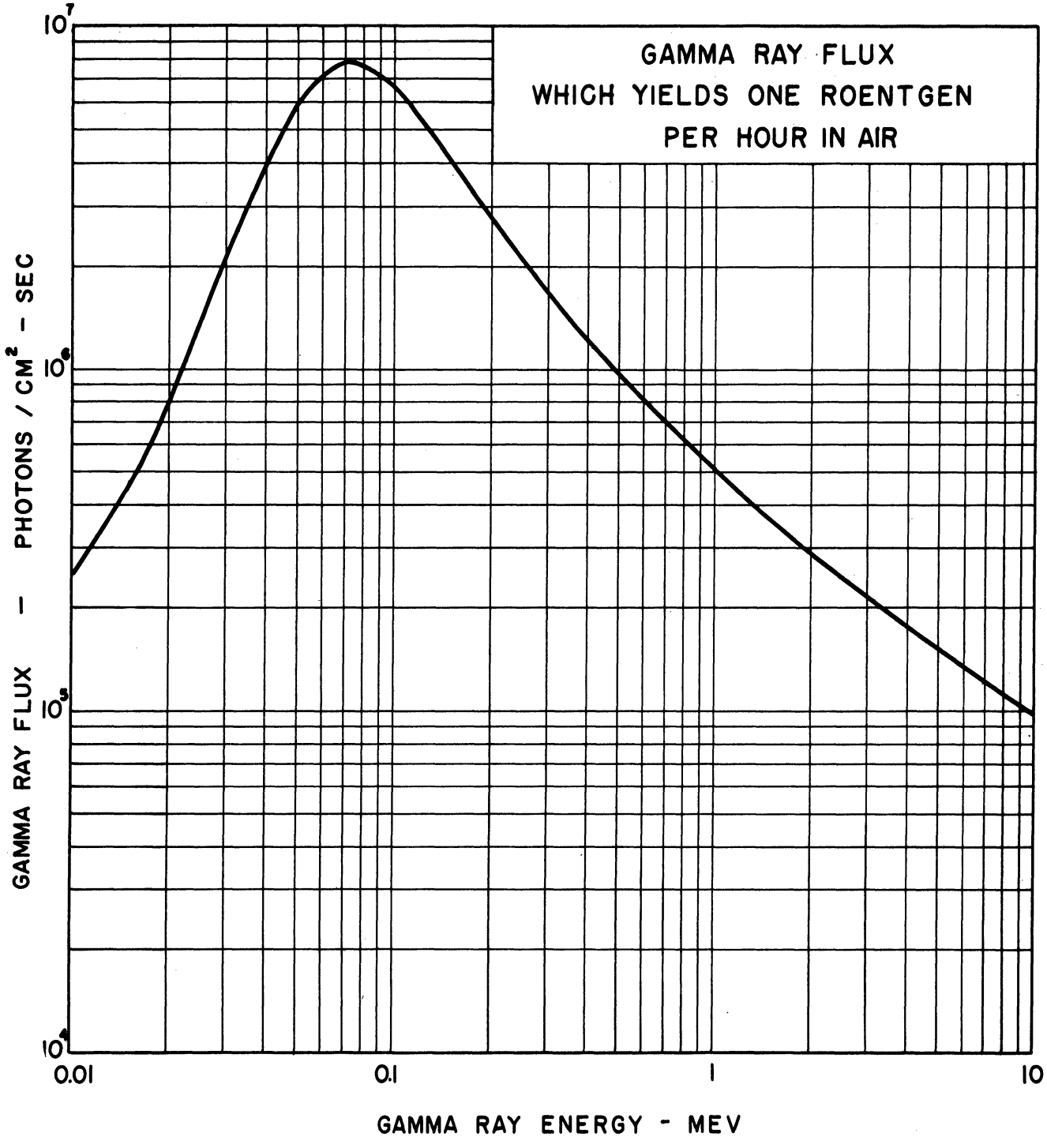

GAMMA RAY FLUX
WHICH YIELDS ONE ROENTGEN
PER HOUR IN AIR
GAMMA RAY FLUX - PHOTONS / CM² - SEC
10⁷
10⁶
10⁵
10⁴
0.01
0.1
1
10
GAMMA RAY ENERGY - MEV

FIG. 21

article;* however, a brief treatment is included here.

The human body can tolerate being exposed to weak radiation over an extended period of time so that the accumulated dose may be quite large although, if the same dose were administered in a short time, it might be lethal. The human body has the ability to repair itself from minor radiation damage if given time to do so. Realization of this has resulted in the development of standards defining the time average exposure tolerable and also the maximum dose taken in a short time regardless of the long time average. (See Table 19). The energy necessary to disable a tissue sample is curiously not enough to cause any appreciable heating. This suggests that the damage mechanism is much more subtle. It is believed that the mechanism by which radiation damages living tissue is the ionization of atoms in complex organic molecules in the tissue cells. There are many ways in which the ionization can take place. Gamma rays produce photoelectrons in tissue which may then cause ionization. Beta particles, protons, and other charged particles ionize material directly as they slow down. Neutrons travelling through tissue collide with protons in the organic tissue molecules to give these protons enough kinetic energy so that they ionize surrounding tissue. When a neutron is captured by nuclei in tissue, capture gamma rays are emitted which may cause further damage.

The unit of exposure is the roentgen (r). This unit was originally devised to measure X-ray dose and is the amount of radiation necessary to produce 1 esu (electro-static unit) of charge in one cubic centimeter of dry air at 0° C and 760 mm pressure. To state this definition in a manner which is independent of the

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* See, for example, National Bureau of Standards Handbook 59

temperature and pressure, this is the radiation necessary to produce 1 esu in 0.001293 grams of dry air. To produce one esu of charge in air, 83 ergs are absorbed from the radiation. The number of gamma ray photons per cm^2-sec. necessary to yield one roentgen per sec. is dependent on the gamma ray energy. (See Figure 21). The photon flux producing one roentgen per second or 83 ergs per gram-sec. will produce 93 ergs per gram-sec. in water or soft animal tissue. The roentgen equivalent physical or rep is a newer unit of radiation dose than the roentgen and is defined as that quantity of radiation which produces 93 ergs per gram in tissue.* The rep for beta rays, protons, and neutrons corresponds to a different flux of such particles than in the case of gamma rays.

It has been recommended that the rad be accepted as the unit of dose where one rad corresponds to an energy absorption of 100 ergs per gram.

Another unit which is defined in terms of biological effect, rather than through energy absorption, is the roentgen equivalent man (mammal) or rem. The rem may be defined as that dose of an ionizing radiation which has the equivalent biological effect on man (or a mammal) as a dose of one rep of gamma rays. The relationship between the rem and the rep or rad is expressed by a multiplicative factor, the relative biological effectiveness (RBE).

$$\text{dose in rems} = (\text{dose in rads or reps}) \times (\text{RBE})$$

---

* Often seen in the literature is the figure 83 ergs per gram in the definition of the rep. This figure is not so convenient to work with as the 93 ergs per gram figure as gamma ray flux meters which measure the ionization in an air chamber are calibrated in roentgens. The conversion to rep's is 1:1 if the 93 erg figure is used in defining the rep.

It makes little difference whether reps or rads are used in this relationship since they correspond to nearly the same energy absorption per gram. The RBE's of several ionizing radiations are given in Table 19 along with some other tolerance data.

TABLE 19

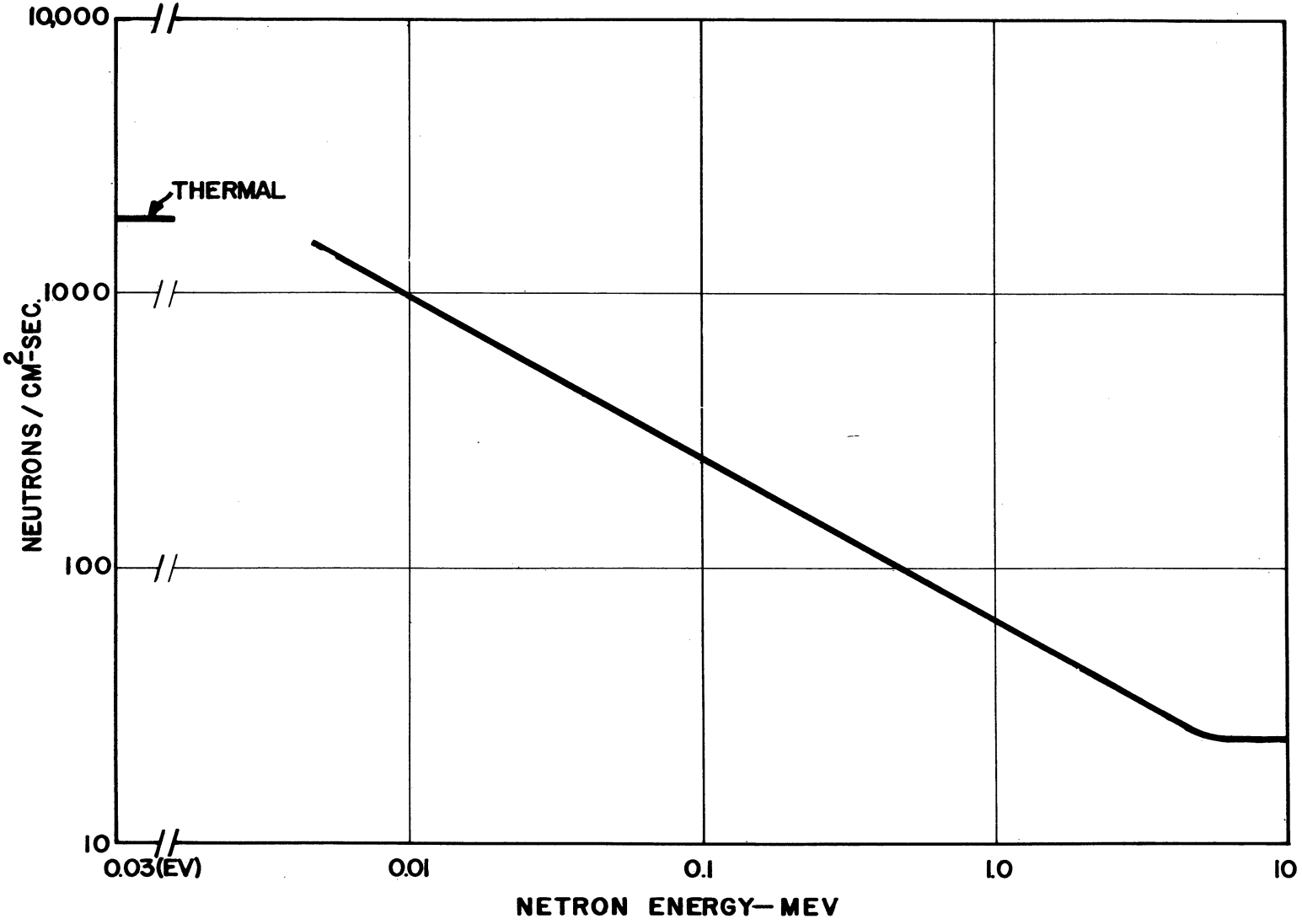
RADIATION TOLERANCE LEVELS

	(Chalk River RBE Conference 1949)
X-rays, gamma rays, beta rays	1
Thermal neutrons	5
Fast neutrons	10
Alpha particles	20
Max. tolerance dose at U. S. Atomic Energy Commission facilities	0.100 rem/day or 0.300 rem/week
Background radiation to which man is exposed. Includes effects of cosmic rays, natural radioactivity of the earth, etc.	0.001 rem/day
Fatal dose applied to the human body in a short time	200 to 800 r

The neutron flux tolerance level is dependent on neutron energy. Figure 22 shows recent estimates.

FIG. 22

APPROXIMATE PERMISSIBLE NEUTRON FLUX BASED ON AN EXPOSURE TIME OF 40 HOURS PER WEEK



1. REACTOR TYPES AND FUELS

A. Thermal Reactors

A uranium-graphite pile is a graphite moderated thermal reactor which is built by stacking graphite blocks containing holes for the insertion of uranium fuel elements and control rods.

The swimming pool reactor consists of an active lattice of fuel elements and control rods which is suspended in a deep water filled concrete tank. The water acts as both a moderator and a coolant. If this reactor is made slightly supercritical, the water locally expands, boils, or decomposes due to radiation damage and the resulting average lower moderator density allows a greater proportion of neutrons to escape the fuel region with the result that this type of reactor is self-regulating.

The water boiler reactor is a homogeneous reactor consisting of a solution of uranyl sulfate or nitrate in light or heavy water. The solution, or soup, is contained in a spherical metal tank with provisions for cooling and control. The water in the soup is the moderator. In principle, the soup could be used as a coolant. Properly designed, a water boiler reactor is self-regulating the same way a swimming pool reactor is.

Studies are being conducted at Brookhaven National Laboratories on the plausibility of building thermal reactors using uranium fuel dissolved in molten bismuth coolant.

It is quite possible that thermal reactors will never be successful as breeders with net fuel gain since the absorption of thermal neutrons by the large quantities of moderator, structural materials, and coolant nuclei tend to destroy the slim margin of breeding possibility.

The reactors shown in Figures 23a and 23b are thermal reactors.

B. Intermediate Reactors

Reactors can be built in which fission is mainly caused by neutrons in an energy range between thermal neutron energies and the higher energies which neutrons produced in the fission process have. (See Figure 6). Such reactors are called intermediate reactors. Since the fission cross sections of the fuel nuclides are lower at intermediate neutron energies than at thermal energies, a larger inventory of fuel is necessary to sustain the chain reaction in these reactors. This disadvantage is off-set by the wider variety of structural materials and coolants which may be used as compared with thermal reactors. The reason for this difference is that at intermediate and higher neutron energies, the neutron absorption cross sections of many readily available materials are low enough so that their use does not seriously poison the reactor.

The slowing-down of the fission neutrons to intermediate energies is effected by the structural materials, coolant, and fuel rather than by use of an auxiliary moderating substance such as graphite or hydrogenous materials.

It is possible that intermediate reactors may succeed as breeders since the neutron absorption by reactor materials is not as great as occurs in thermal reactors. Figure 23c shows an intermediate reactor system.

C. Fast Reactors

Few fast reactors have been built and information on the operation of these is mainly classified. Interest in fast reactors is partly due to the variety of structural materials that may be employed, as in the case of intermediate reactors, but even greater interest resulted when it was shown with the Experimental Breeder Reactor (EBR-I) at

FIG. 23a

HETEROGENEOUS THERMAL
FISSIONING REACTOR

COOLANT – H_2O OR D_2O
MODERATOR – H_2O OR D_2O
PRESSURE – 1000 PSIA OR HIGHER
PRODUCTS – POWER, RADIOCHEMICALS

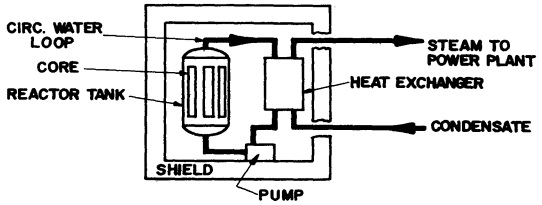


FIG. 23b

HOMOGENEOUS THERMAL
FISSIONING REACTOR

COOLANT – AQUEOUS URANYL SULFATE OR NITRATE SOLUTION
MODERATOR – H_2O
PRESSURE – 1000 PSIA OR HIGHER
PRODUCTS – POWER, RADIOCHEMICALS

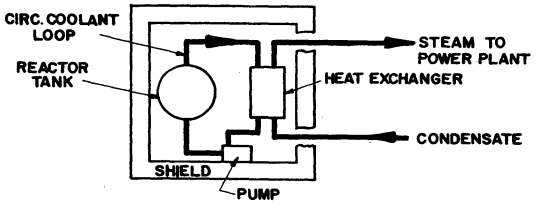


FIG. 23c

HETEROGENEOUS FAST OR INTERMEDIATE
ENERGY FISSIONING REACTOR

COOLANT – LIQUID Na
INTERMEDIATE COOLANT – LIQUID Na-K
PRESSURE – ATMOSPHERIC
PRODUCTS – POWER, RADIOCHEMICALS

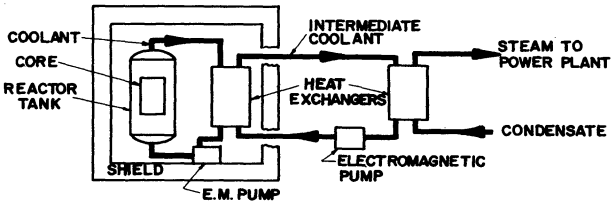
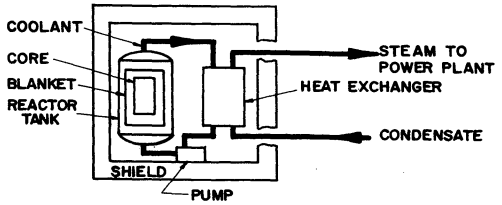


FIG. 23d

HETEROGENEOUS FAST BREEDER OR
CONVERTER REACTOR

COOLANT – LIQUID Na
PRESSURE – ATMOSPHERIC
PRODUCTS – POWER, RADIOCHEMICALS, FISSIONABLE MATERIAL



Arco, Idaho, that more fissionable fuel could be produced than consumed in a fast reactor. This reactor uses U-235 as fuel with a core about the size of a football. Plutonium is produced in a natural uranium blanket surrounding the core. Fast reactors must be small to keep the fuel inventory low, which incidentally is larger than required by either thermal or intermediate reactors. With small reactors, there arises the problem of removing large amounts of heat from small areas. This points to the use of liquid sodium or Na-K as a primary heat transfer medium as these materials have large heat transfer coefficients. It may be possible that the attractive breeding gain of fast reactors will more than justify the inconveniences, in which case we shall likely see many power plants employing fast reactors in the future. (See Figures 23c and 23d).

D. Reactor Fuel Elements and Fuels for Homogeneous Reactors

Reactor fuels may be prepared using any desired mixture of fissile and non-fissile isotopes of the fuel in question,* since the chemical and mechanical properties of chemical elements are independent of the isotopic constitution. Chemically, the fuel may be in the form of metals, oxides, metal solutions, aqueous solutions of fuel salts, etc., as the nuclear properties of the fuel nuclei are, for all practical purposes, uninfluenced by their chemical environment.

A reactor fuel element is a piece of material containing macroscopic quantities of a fuel nuclide for use in a heterogeneous reactor. This term is not to be confused with the meaning of "element" taken in the chemical sense. A number of different designs for fuel elements have been used. In general, it is desirable to enclose the fuel in a jacket

*
The ratio of fissile to non-fissile nuclides is dependent on the reactor design.

which does not allow the radioactive fission products to contaminate the surrounding parts of the reactor. The jacket must have high conductivity to allow escape of the fission heat, must be resistant to corrosion by the coolant, moderator, etc., and have a low neutron absorption cross section. Metallic uranium is often clad with aluminum for this service and is useful so long as the melting point of the aluminum is not reached in operation.

Fuels for homogeneous reactors may be in the form of solutions of uranyl sulfate or uranyl nitrate in H_2O or D_2O, solution of uranium metal in molten bismuth, etc.

A high temperature reactor system under study proposes using UO_2 particles in a bed which is fluidized by an air stream. This fuel has properties in common with both homogeneous and heterogeneous reactor fuels.

2. POWER FROM NUCLEAR REACTORS: THE NUCLEAR MANUFACTURING PLANT

A. Introduction

Since the U. S. Atomic Energy Commission released classified information on nuclear reactors to industrial power study groups, a variety of power plant designs have evolved, although at the time of this writing, none of the proposed plants have been built. Perhaps the most attractive idea which nuclear power plants offer is the possibility of obtaining so much energy from a given mass of fuel. Using the conservative figure of 182 Mev released in the fission of one U-235 nucleus, we readily see that the fission of one gram of U-235 releases 7.0×10^7 B.T.U. or 0.86 megawatt-days of energy. It has been estimated that a cubic block of U-235 metal eight feet on a side would provide all the world's fuel needs for a year.

Figures such as these permit consideration of designing ships which are to operate for long periods without re-fuelling and having the additional advantage of providing greater cargo capacity which in conventional coal and oil burning vessels is involved in the storage of fuel. Long range nuclear powered aircraft may make an appearance in the future as well and studies are presently being made in this direction. There is also the possibility of building electric power plants for remote locations in which nuclear fuels will be used with the advantage that fuel may be transported much more conveniently than is possible with conventional fuels.

There is the further prospect of finding industrial uses for the intense radioactivity of the fission products. A great deal of exploratory work is now in progress with the aim of finding new large scale markets for the fission products, which may become valuable as by-products of power production. This subject is discussed further in Section 4B.

Serious consideration must be given to the present rate of depletion of the world's supply of fossile fuels. A recent survey* of the rate of the world's population growth, the present and estimated rate of increase of fuel use per capita, and the estimated reserves of the fossile fuels, points out that new fuel resources must be developed. Figure 24 presents the results of some of these estimates of future fuel requirements and known fuel reserves. The vast low grade reserves of the fissionable nuclides gives us hope that the high standard of living now enjoyed in the United States and some other countries will not have to be sacrificed in the next few centuries on account of power shortage.

B. Power from Nuclear Reactors

The energy produced in the nuclear reactor must generally be abstracted in the form of heat. This heat may be either used at low

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* Palmer, Putnam, Energy in the Future, D. Van Nostrand and Co., New York, 1953.

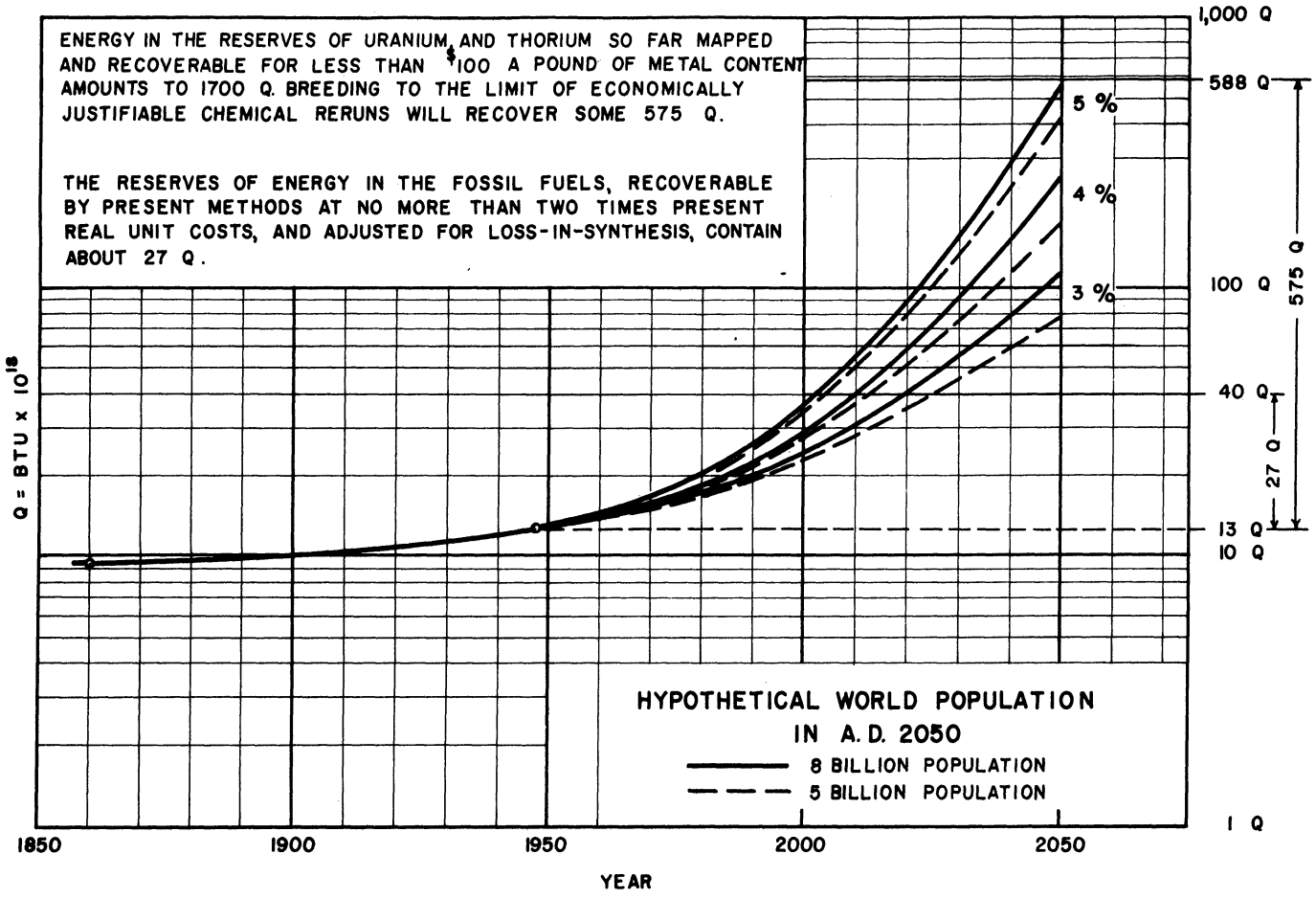


FIG. 24

Estimates of maximum cumulative World demand for energy input, A.D. 1947 to 2050, assuming two different maximum plausible populations, three different maximum plausible rates of growth in the per capita demand for energy output measured at the point of end use, and one minimum plausible trend in the weighted average world efficiency curve. The value of 9 Q at A.D. 1860 represents the approximate cumulative input to the World energy system between A.D. 1 and the earliest date for which there exist reasonably adequate estimates of annual fuel consumption. The demands are compared with the 27 Q recoverable from fossil fuels and the 575 Q recoverable from nuclear fuels, at less than 2 times 1950 costs.

From "Energy in the Future" by Palmer Putnam, D. VanNostrand Company, Inc., New York 1953.

temperatures for building heat, or it may be used at high temperatures to produce steam which may be fed through a turbine to produce electrical power. A third possible means of using energy from a nuclear reactor is as high temperature heat for the initiation of chemical reactions or the cracking of petroleum, or other high temperature requirements such as metallurgical operations. The choice of which of these modes of heat utilization is to be adopted has a profound influence upon the design of the reactor. If heat is merely to be abstracted in order to prevent destructively high temperatures in the reactor, then the heat may be discarded at low temperature in cooling water or air, or may be used for some purpose such as building heating. A reactor operated in this manner has less stringent requirements upon the physical properties of the structural and coolant materials than does one operated at a higher temperature. However, if one is to employ the energy for the generation of power, then a relatively high temperature must be produced in the reactor. Liquid metals may be used as coolants as shown in Figure 23c and 23d, or water at high temperature and pressure may be used as shown in Figure 23a. In either of these two alternatives, the coolant channels within the reactor must be designed to withstand the corresponding chemical and physical conditions. In addition, care must be taken that the fuel within the reactor may transfer heat to the coolant through sufficiently short distances so that destructively high temperatures are not reached within the fuel itself.

Coolant materials passing through the reactor core will in general absorb some neutrons, and may become radioactive. Consequently, it has been proposed that the coolant not be charged directly to the turbine or to other heat utilization devices, but rather should be caused to exchange its heat with a secondary coolant loop. A secondary coolant,

although exposed to the radioactivity of the primary coolant, would not become radioactive, and consequently could be used in machinery without fear of radioactive contamination. Such a secondary heat exchange system is shown in Figure 23c. Liquid metals have been proposed as a primary reactor coolant because they may attain relatively high temperatures without correspondingly high vapor pressures. These liquid metals, such as sodium, may then exchange their heat with a secondary coolant which would be suitable for operation of a turbine. It appears possible that the secondary coolant might also be a chemical or petroleum stock which is to be subjected to high temperature in order to initiate cracking or other chemical change. Inasmuch as radiation often initiates chemical reactions, it may be possible to obtain products with properties resulting from a combination of the thermal and radiation effects. Such properties may not be attainable in present thermally initiated reactions, and therefore offer promise of investigation.

C. The Nuclear Manufacturing Plant

Nuclear Reactors, together with their attendant auxiliaries, can be considered from one point of view as manufacturing plants. There are three chief products of such a plant, namely:

1. Energy which appears initially in the form of radiation and is degraded to heat and may ultimately be converted to electric power.

2. The fissionable materials, uranium^{233} or plutonium^{239}, which are produced by the absorption of neutrons in source material.

3. Gross fission products.

One kind of raw material for these plants are source materials such as uranium^{238} and thorium^{232} which, upon the absorption of neutrons, become converted to fissionable materials. Another kind of raw materials

are the fissionable materials themselves which may be regarded as the fuels for the manufacturing plant.

If one is to consider a nuclear reactor as a part of a manufacturing plant, it is necessary to consider sources of raw materials, the operation of the reactor and its auxiliaries, and markets for the products of the manufacturing plant. The sequence of interdependent operations upon which the nuclear manufacturing would be based are the following:

1. The preparation of feed materials, which are the source and fissionable isotopes required for the operation of a reactor.

2. The enrichment of naturally occurring ruanium by means of gaseous diffusion to U-235 concentrations sufficiently great that it may serve as a fuel for a reactor.

3. The formulation of source and fissionable materials into physical and chemical states suitable for use as nuclear fuels.

4. The operation of a nuclear reactor.

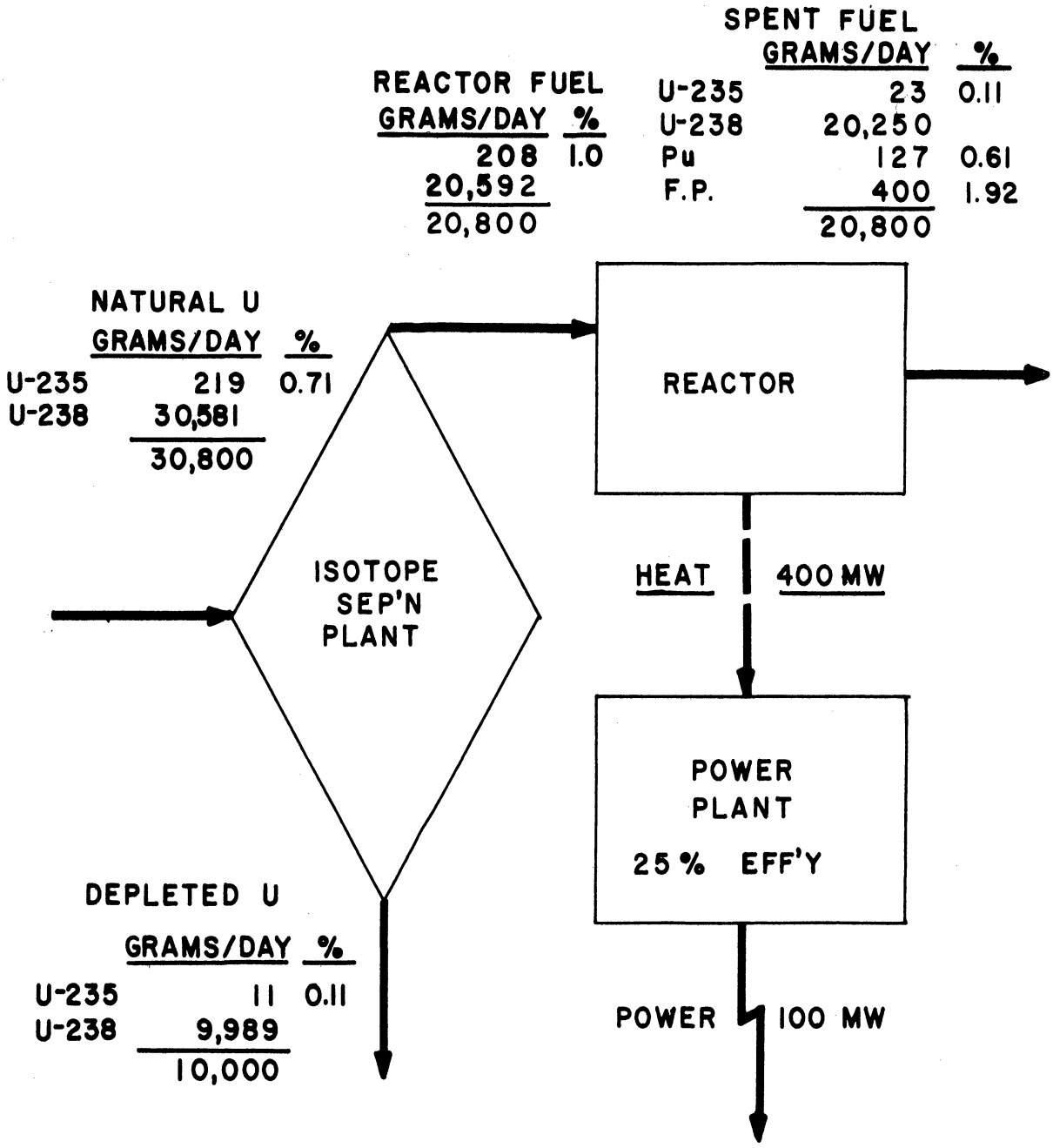
5. The conversion of the energy from the fission reaction to electricity or to heat for other purposes.

6. The chemical or physical separation of irradiated fuel to permit the recycling of unused source and fissionable material for further use as fuels.

7. The processing of separated fission products with the production of packaged gross fission products or the isolation and packaging of selected individual products.

In Figure 25, a hypothetical scheme is shown which would permit conducting the operations just described. According to this scheme, natural uranium containing 0.71% of the fissionable isotope uranium^{235} is the raw material input to an enrichment plant. Partial enrichment of

FIG. 25



FIGURES 25-28 FROM:
M. BENEDICT, IND. & ENG. CHEMISTRY, NOV. 1953.

the fuel would occur, resulting in the production of two streams from the isotope separation plant. The enriched stream contains 1% of uranium^{235} and the depleted streams 0.11%. The enrichment of the fissionable isotope from 0.71% to 1% in concentration imparts to the enriched fuel a sufficient reactivity in excess of that occurring in natural uranium to permit certain additional latitude in reactor design and operation. The products of the reactor described in this system will be plutonium, fission products and power. The production of power from heat is assumed to be straightforward, and it is assumed in this operation that the steam turbine and generating equipment have an overall efficiency of 25% based upon the incoming heat. The spent fuel discharged from the reactor contains a residual 0.11% of uranium^{235} and 0.61% of plutonium. The total of these is 0.72%, which is not as great as the 1% of U^{235} which entered the reactor. Consequently this type of reactor does not produce as much fissionable plutonium as it burns uranium^{235}. The operation of this type of system depends upon the availability of an isotopic separation plant, since the uranium^{235} which is produced is consumed without the production of corresponding quantities of plutonium. The plutonium itself might be used as a fuel for recycling through the reactor if it were produced in quantities sufficient to replace the uranium^{235} originally charged. The depleted uranium which is produced by the isotope separation plant is a by-product of this system.

An alternative scheme, calling for the operation of a reactor with natural uranium raw material, is given in Figure 26. In this scheme, natural uranium is mixed with plutonium from previous reactor operations. Plutonium might have been produced previously in some reactor employing a cycle using no recycle plutonium, such as that described in Figure 25. Once a supply of plutonium is available, it can be used to enrich the

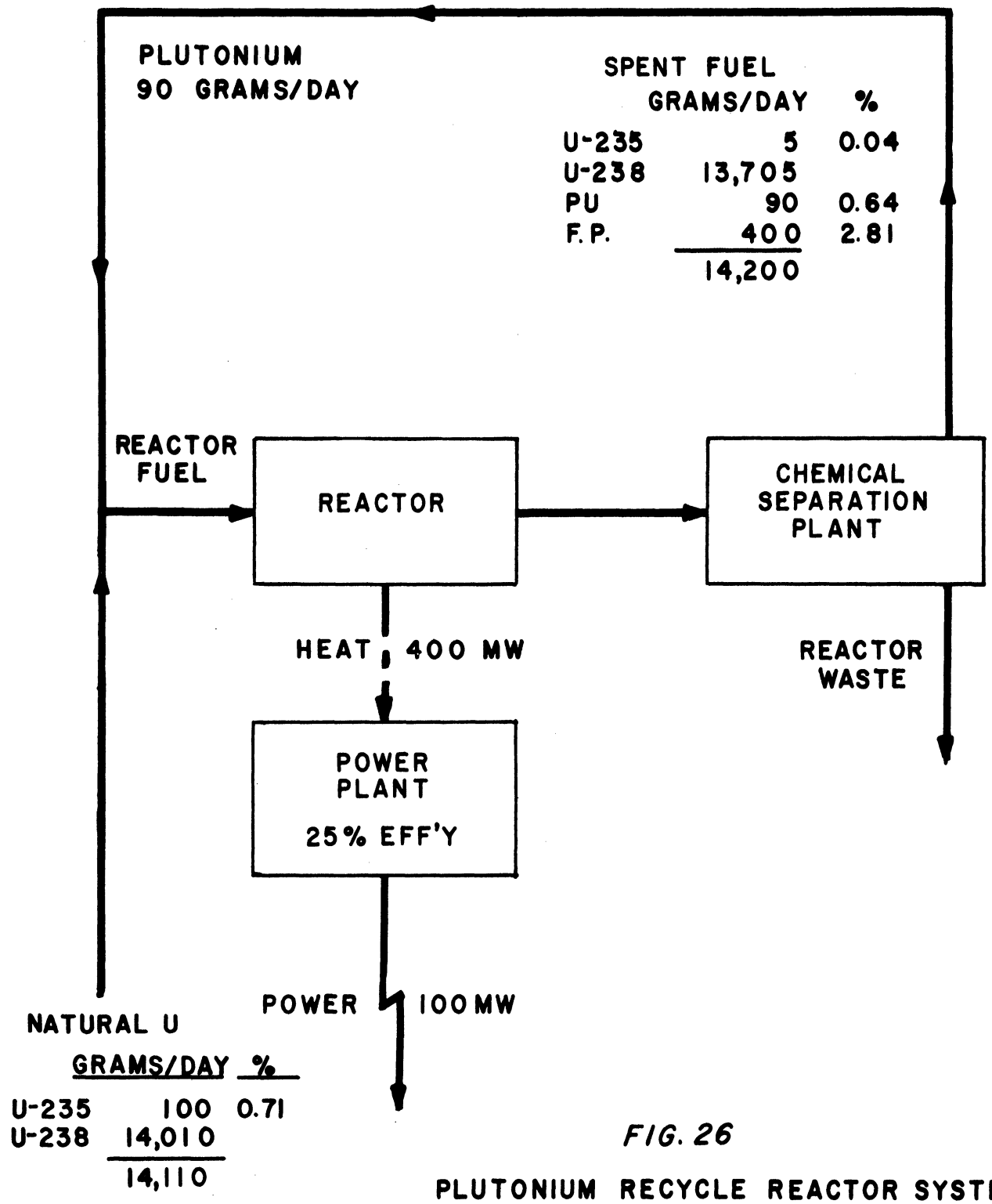


FIG. 26

PLUTONIUM RECYCLE REACTOR SYSTEM
USING NATURAL URANIUM FUEL

PRODUCTS:

POWER
PLUTONIUM
RADIOCHEMICALS

incoming natural uranium with fissionable plutonium instead of adopting the expedient described in Figure 25 of conducting an isotopic separation upon a natural uranium in order to increase the content of fissionable uranium^{235}. During operation of the reactor of Figure 26, the plutonium and uranium^{235} which are charged, undergo fission, releasing neutrons which are absorbed in uranium^{238} and thereby, producing more plutonium. Some of the plutonium thus produced fissions in turn and produces more power and more plutonium. This sequence of events may be continued until the presence of fission product poisons necessitates the removal of the fuel because of declining reactivity. At the conclusion of this reactor cycle, there remain a total of 95 grams of fissionable material compared with the 190 grams which were charged to the reactor. Consequently, a system of this kind requires the continual charging of fresh natural uranium, and hence, causes a gradual depletion of the stock of fissionable 235. However, if the reactor described here were to operate as shown, no net depletion of the original charge of plutonium would occur. The products from such a system would be power, plutonium to sustain reactor operations, and radiochemicals.

Figure 27 describes a scheme for self-sustaining nuclear manufacturing system. This system produces slightly more fissionable material than it consumes, and differs in this respect from the systems described in Figure 25 and 26. Consequently, the only raw material required is uranium^{238}. The uranium^{238} could be taken in the form of depleted uranium from the operations of an isotope separation plant such as that described in Figure 25, or from the discharged fuel from a reactor such as that of Figure 26. The scheme shown in Figure 27 is a U^{238} - Pu fast breeder reactor. This scheme is more complicated than either of the

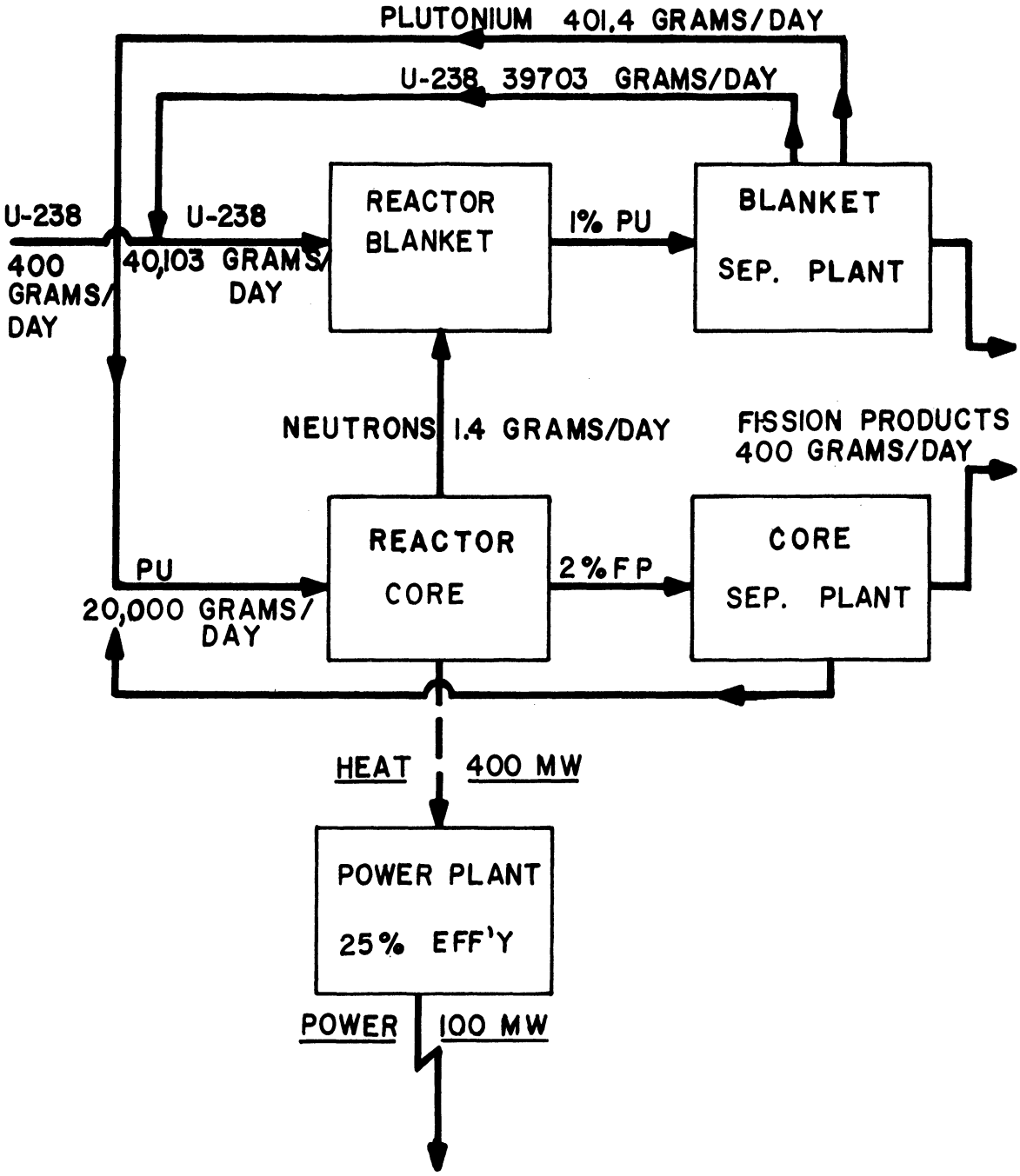


FIG. 27

FAST PLUTONIUM U-238 BREEDER REACTOR SYSTEM

PRODUCTS :

POWER
PLUTONIUM
RADIOCHEMICALS

previous reactor systems described. Most of the fission occurs in the reactor core. Neutrons escaping from the surface of the core are absorbed in the surrounding blanket of uranium^{238} instead of being allowed to escape as in the previously described systems. The capture of these neutrons accounts in large part for the fact that more fissionable material is produced than is consumed in this type of system. The course of operations is that uranium^{238} is brought into the system and mixed with recycled uranium^{238} from the blanket separation plant. This mixture is then fed through the blanket, whereupon absorbing neutrons, some of the U^{238} is converted to plutonium. This plutonium is removed upon subsequent separation. Some of this plutonium may be mixed with plutonium separated from previous core operations and fed to the core as fuel. After undergoing the reaction of fission in the core, the plutonium fuel is again subjected to the separations process to remove the fission products, and is recycled for more burn-up. Again it is considered that the operation of the power plant is of a conventional nature and is not further described here. The raw material upon which this type of plant operates is naturally occurring uranium or depleted uranium^{238}, and the products are power, plutonium and radiochemicals.

Because construction of this type of reactor system is more complex than those of Figures 25 and 26, the capital and operating costs may be greater. This fact may offset to some extent the economic attractiveness of producing more fissionable material than is consumed.

In Figure 28, an alternative reactor scheme is shown which is known as a U^{233} - Th thermal breeder reactor system. The raw material to the system is throium^{232}. This is mixed with a certain amount of thorium 232 containing uranium^{233} produced in previous reactor operations, and is charged to the reactor. During reactor operations the

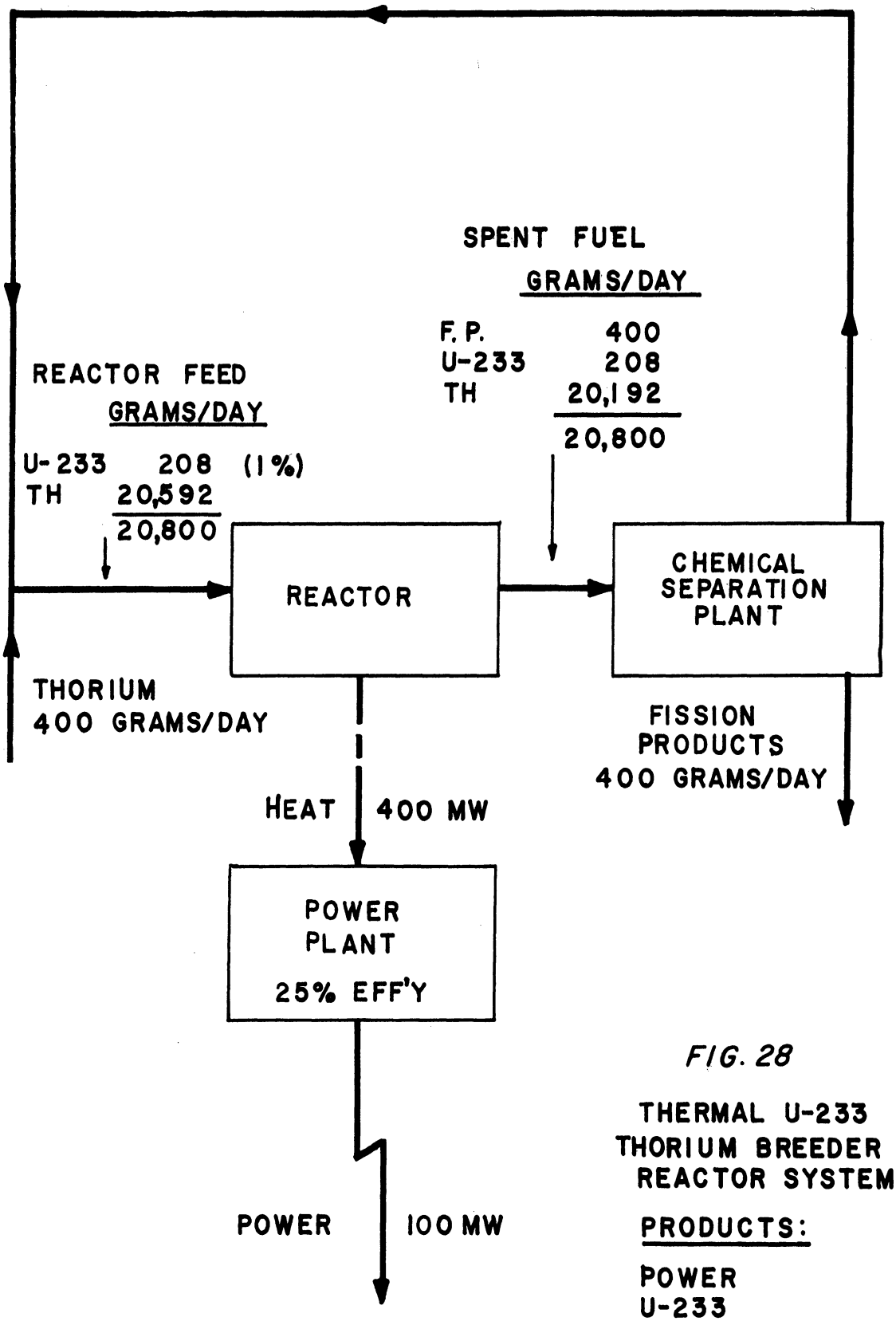


FIG. 28

THERMAL U-233
THORIUM BREEDER
REACTOR SYSTEM

PRODUCTS:

POWER
U-233
RADIOCHEMICALS

reaction of fission occurs and excess neutrons are absorbed in the thorium^{232} rather than in uranium^{238} as in the previously described reactor systems. During startup operations, it will probably be necessary to use uranium^{235} as a fissioning fuel in order to initiate the cycle. After the reaction of fission and absorption of neutrons has occurred, the fuel is sent to the chemical separation plant, where fission products are removed and stored or sold. The decontaminated fuel contains uranium^{233}, as described in Part II, Sec. 2E. The uranium^{233} and thorium^{232} output from this separation plant can then be recycled to the reactor for further conversion and power production. If uranium^{233} were produced in excess of recycle requirements the surplus would be a product of operations. The raw material required for this system is thorium^{232}, which occurs in nature. The products are power, uranium^{233}, and radiochemicals. It is not known for certain however, that the thermal Th-U^{233} breeding cycle is self-maintaining without the occasional addition of small amounts of U^{235}.

The choice of the system to be employed is governed as in the case of other types of manufacturing plants by many considerations not necessarily related to the technical feasibility of the operation. Among these considerations are availability and cost of raw materials, such as natural uranium and natural thorium, uranium isotopically enriched in uranium^{235}, plutonium, structural materials, coolants, moderators; the cost of operating the separation plant and many other factors which can be determined only through a detailed technical and economic analysis.

It is thought that the development of breeder reactors will be a necessary part of a nuclear manufacturing economy. This will be the case if full advantage is to be taken of our natural resources of uranium. Since most of natural uranium is in the form of uranium^{238}, the use of

breeder reactors should produce supplies of fissionable material in excess of those used. Such a circumstance would justify the operation of less efficient fissioning or converter reactors for power production in applications where a less complicated type of reactor installation is desirable.

3. FUEL SEPARATION

A. Introduction

As operation of the reactor continues with a given fuel charge, the concentration of fission product materials increases, and consequently neutron losses due to absorption by the fission products increases. Ultimately, a condition is reached in which the original fuel has produced enough fission products so that the nuclear chain reaction can no longer be sustained and the initial charge of fuel must be removed and replaced by purified fuel. Another type of behavior exhibited by metallic nuclear fuels undergoing irradiation is that of radiation damage. As a consequence of operation at elevated temperatures, and as a consequence of the production of the fission products within the crystal lattices of the nuclear fuels, the fuels themselves undergo structural changes which are deleterious to their mechanical and thermal functioning. Consequently, both radiation damage and fission product build-up constitute reasons why nuclear fuel must eventually be removed from a reactor and replaced by purified and reconstituted fuel.

Fuels which are no longer suitable for reactor operation might be discarded in some suitable manner except for two reasons. The first of these reasons is that only a fraction of the fissionable material originally charged to the reactor will be consumed before radiation damage and fission product build-up necessitate removal from the reactor. Consequently, if the fuel were discarded, much valuable fissionable

material would also be discarded along with the waste products. The second reason why the fuel cannot be discarded is that in reactors which contain any uranium^{238}, the capture of neutrons by the uranium^{238} results in the production of plutonium which is a reactor fuel too valuable to discard. Consequently, it is found that irradiated reactor fuels must be treated in some manner so that the fissionable uranium and plutonium may be recovered from these fuels for re-use as reactor fuel.

B. Aqueous Methods

In the following discussion, various means will be described of accomplishing the treatment of irradiated nuclear fuels in such a manner that fissionable materials are recovered for re-use. The first method of fuel separation which will be described is aqueous chemical processing. Aqueous chemical processes are applied to the separation of uranium, plutonium, and fission products, when the component parts of the fuel must be separated in high degrees of purity.

For heterogeneous reactor fuels in which the fuels are discreet bars of metal surrounded by a moderator and coolant, it is necessary to dissolve the solid fuel elements in acid. The dissolution is conducted in such a manner that the resultant solution has properties which will permit effective chemical separations.

Since ordinary or light water is used in aqueous chemical separations, an ideal application of the aqueous method is the separation of fuels in a light water homogeneous reactor. If heavy water is used in a homogeneous reactor, it is necessary to remove the heavy water from the dissolved salts of fissionable material, since the heavy water is so expensive, and to redissolve the dehydrated fuel and fission product salts in light water for subsequent chemical processing.

The separations of uranium, plutonium, and fission products may be accomplished by one of three aqueous methods or combinations thereof. These methods are:

1. Precipitation
2. Solvent extraction
3. Ion exchange

Earlier methods of aqueous chemical processing employed a precipitation technique. The use of bismuth phosphate permitted the selective separation of plutonium from a mixture of uranium and fission products. This permitted plutonium isolation in pure form, but did not permit the separation of uranium from highly radioactive fission products.

A typical solvent extraction process is illustrated in Figure 29. The uranium and plutonium contained in the dissolved reactor fuels are oxidized to the hexavalent state prior to solvent extraction. In this state of oxidation, these materials may be extracted from an aqueous solution by means of solvents such as: diethyl ether, tributyl phosphate dissolved in kerosene or in carbon tetrachloride, and hexone. The various steps required in the solvent extraction process are:

1. The extraction of uranium and plutonium from an aqueous solution of fission products by means of a solvent.
2. The extraction of the plutonium salt from the uranium salt.
3. The stripping of the uranium salt from the solvent.
4. The extraction of the separated plutonium salt from small quantities of fission products.
5. The conversion of the aqueous solution of uranium salt to the form of metallic uranium and the subsequent refabrication of fuel elements from the uranium.

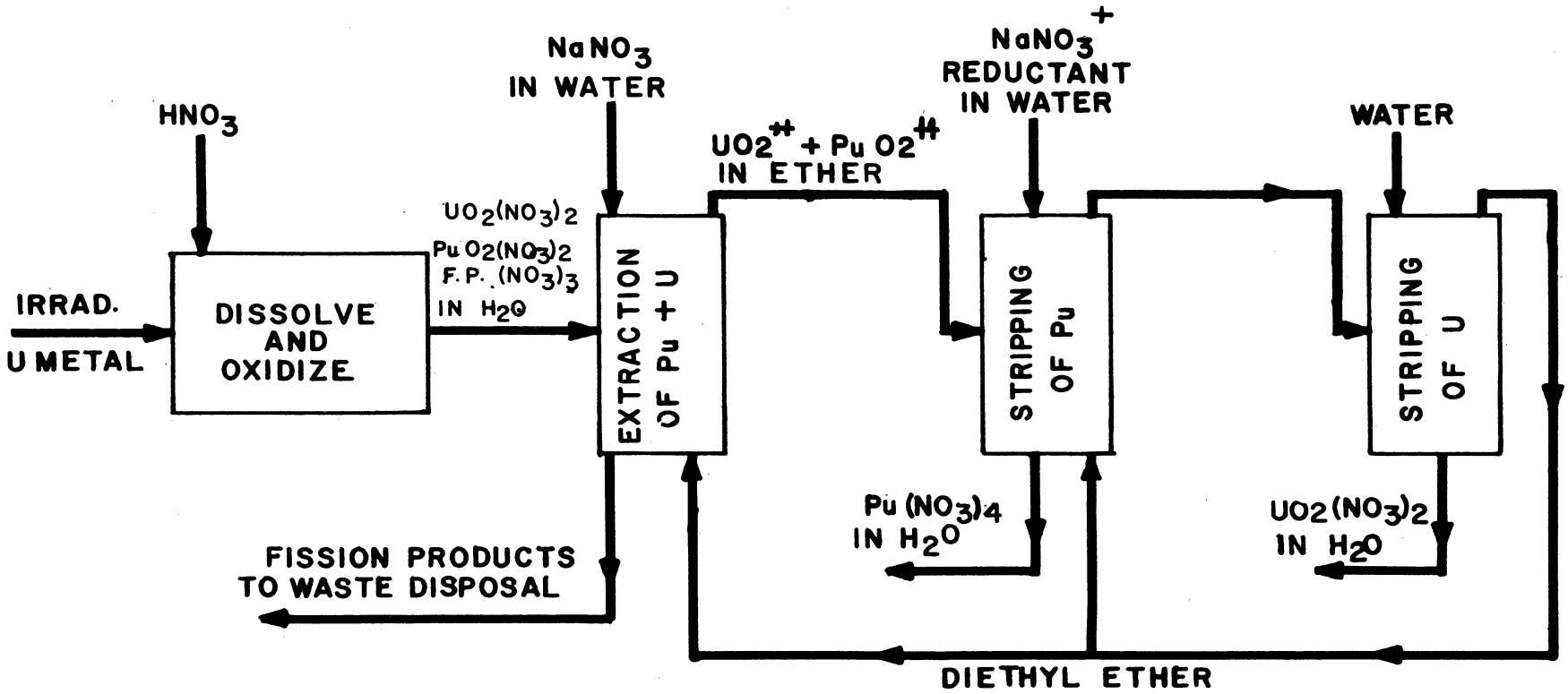


FIG. 29

SOLVENT EXTRACTION PROCESS FOR
IRRADIATED URANIUM

PRODUCTS:
URANIUM NITRATE SOLUTION
PLUTONIUM NITRATE SOLUTION
RADIOCHEMICAL SOLUTION

Methods of aqueous chemical separation employing ion exchange techniques offer promise in certain uranium separations, plutonium purification, and in the isolation of specific radiochemicals from the solution of fission products.

C. Fluoride Volatility Methods

An alternative to the aqueous chemical processing technique for the separation of uranium, plutonium, and radiochemicals, is that of the fluoride volatility technique. The basis of the fluoride volatility technique is that uranium hexafluoride is a volatile material which condenses at conditions not far removed from ordinary temperatures and pressures. There would seem to be the possibility, therefore, of forming uranium hexafluoride from the uranium present in irradiated nuclear fuels and then distilling this uranium away from the plutonium and fission products in the form of the uranium hexafluoride. The residual materials from the distillation may be treated further as required, and the uranium hexafluoride is either reduced to the form of metallic uranium for recycle to a reactor, or the uranium hexafluoride may be charged directly to a gaseous diffusion plant for re-enrichment of the uranium^{235} content.

A block flow diagram of a representative fluoride volatility process is shown in Figure 30.

D. Pyrometallurgical Processing

A third method for the treatment of irradiated nuclear fuels to render these fuels suitable for re-use in a nuclear reactor is that of pyrometallurgical treatment. Pyrometallurgical methods have the common feature of preserving the chemical state of the reactor fuel during processing. Consequently, a minimum of chemical conversions are required in this method of fuel treatment. Alternative pyrometallurgical processes are:

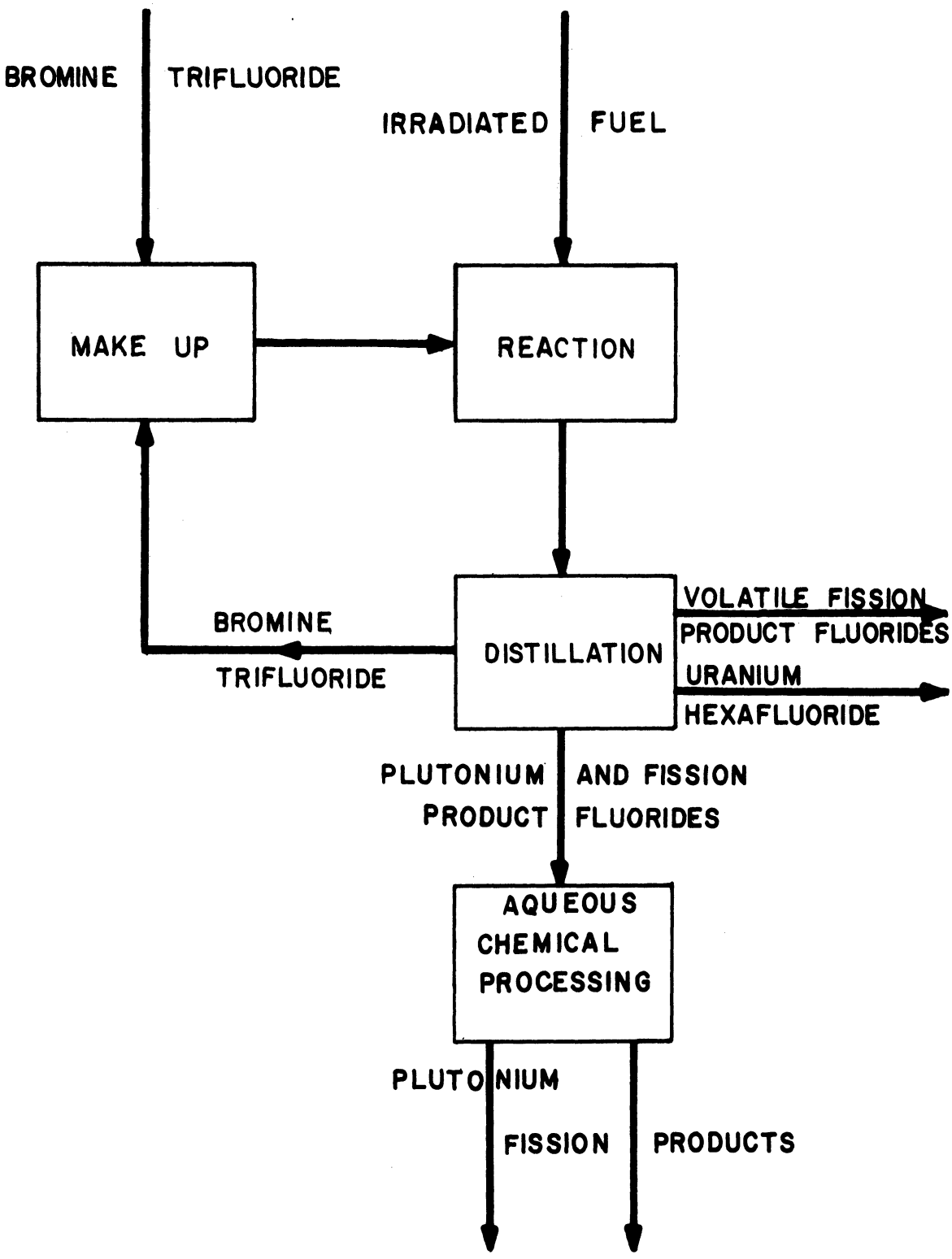


FIG. 30

BLOCK FLOW DIAGRAM OF A

FLUORIDE VOLATILITY PROCESS

1. Melting and re-solidification of the fuel.

2. Extraction of the molten fuel by means of metals or fused salts.

3. The distillation and condensation of fuel metals at high temperatures.

4. Zone melting.

Pyrometallurgical processes are quite straightforward in concept. These processes are dependent, however, upon materials capable of withstanding high temperatures. With present knowledge of high temperature materials, melting and extraction have attractive features. Melting alone has application to fast reactors where rigid specifications of purity in the fuels are not essential.

Extraction at high temperatures is a technique whereby separations between uranium, plutonium in the fission products, as well as structural materials, may be achieved.

A flow diagram of a representative pyrometallurgical process is illustrated in Figure 31.

4. USE AND STORAGE OF THE FISSION PRODUCTS

A. Introduction

Economical nuclear power may be contingent on the successful development of large-scale industrial uses for the extensive quantities of fission products produced in nuclear chain reactors. Thus far, the fission products have constituted a high-cost liability. Their presence in reactor fuel elements requires expensive chemical separations plants to provide purified nuclear fuel suitable for re-use in reactors. Present as wastes after chemical separation, the fission products incur an

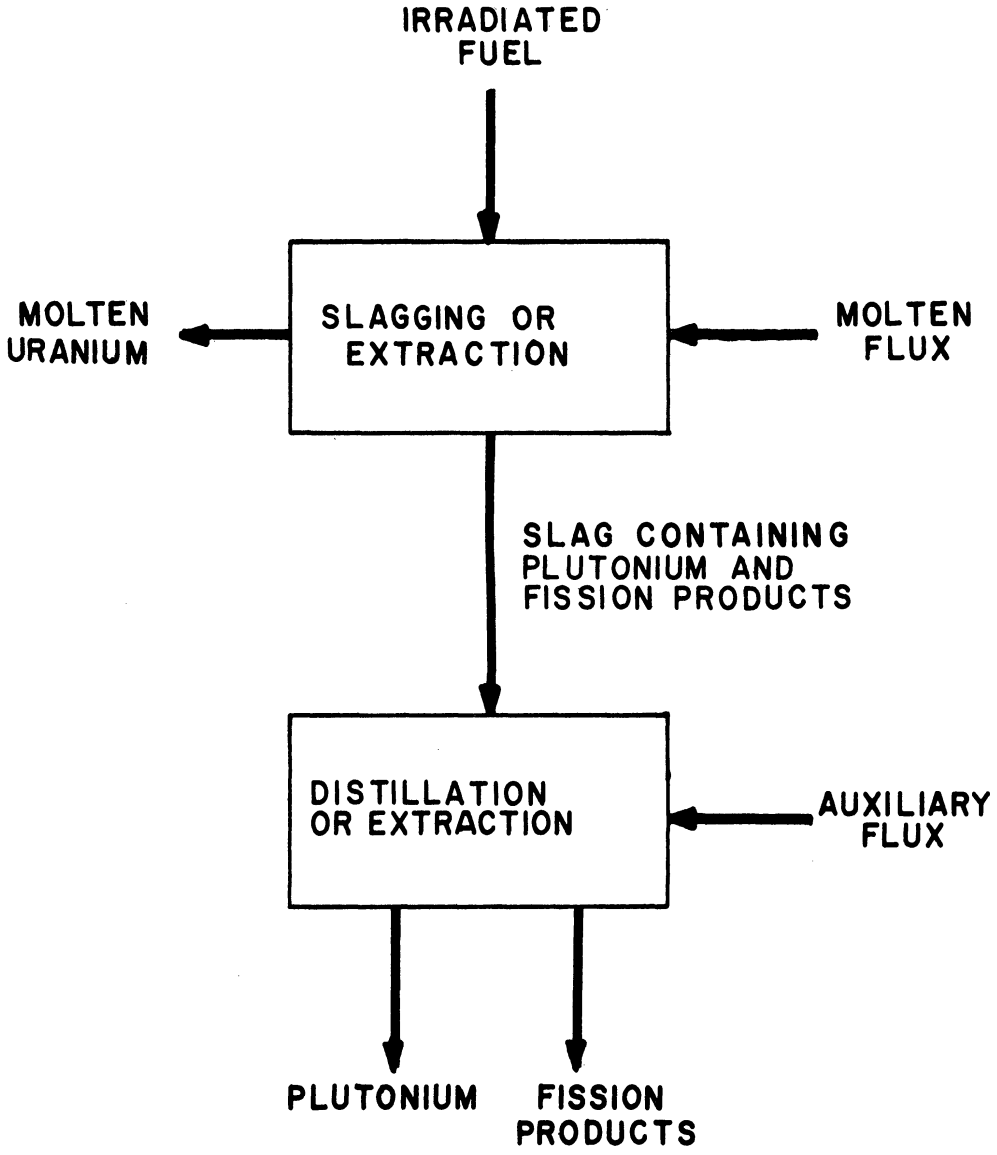


FIG. 31

BLOCK FLOW DIAGRAM OF A PYROMETALLURGICAL PROCESS

even greater expense in handling and storage. Economic studies indicate that handling and storage costs may approach 60 per cent of the total costs of chemical separations. The consumers of reactor power will bear the expense of these operations, unless methods are devised to defray the cost by industrial utilization of the fission products. Research groups from industry and the universities are presently striving striving to resolve the fission-product problem by investigating potential uses for the fission products. Many of their findings have previously been put to profitable use, and some show definite promise for future applications; others are highly speculative. The final solution to this challenging problem will determine the degree to which power from nuclear reactors will compete in a free market.

B. Storage of Fission Products

These materials must be maintained in biologically safe locations. If the fission products are to be regarded as wastes, one may consider dispersing them in the atmosphere or in the ocean, casting them in concrete or clay and burying them in special plots or at sea, storing them in tanks as aqueous solutions, or in the form of solids, etc. Dispersal appears to be unsafe for the very huge quantities produced by a nuclear power industry. Consequently, there is strong incentive to develop economical methods of containment. The most economical storage method would probably be that in which fission products are contained in the smallest volumes in order to minimize the costs of surrounding shielding.

In the case of the long lived fission product nuclides such as Sr^{90} and Cs^{137} continued addition of a stockpile at a constant rate will eventually yield and equilibrium value of the radioactivity (in curies) of the nuclide in question. This equilibrium value may be calculated

through:

$$(\text{curies at equilibrium}) = 8.45 \times 10^{-3} \text{ (power level in watts)(fission yield, \%)}$$

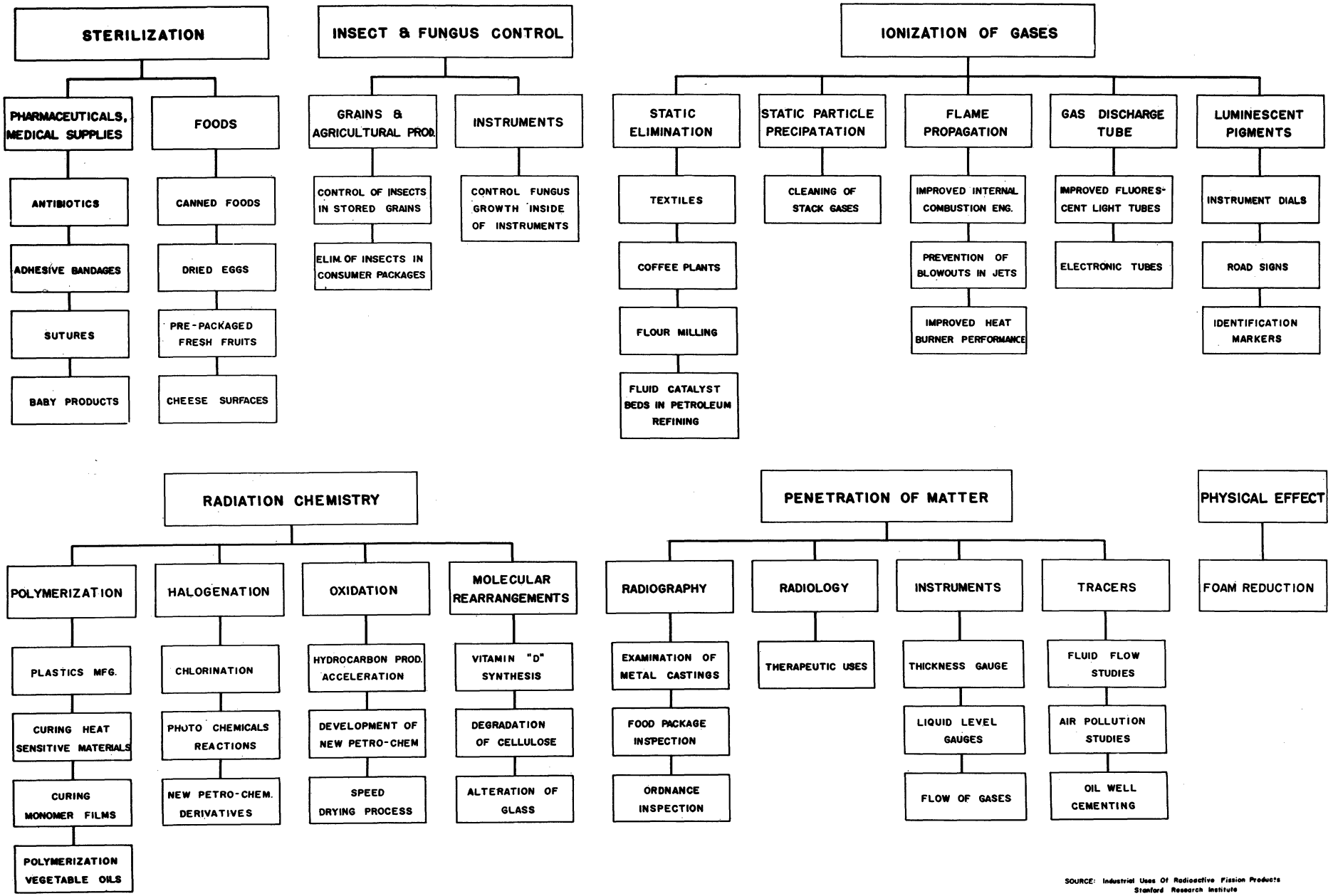
where the power level refers to the sum of the gross power levels of the reactors adding the fission product to the stockpile and the fission yield is given by curves such as Figure 7. Thus, a reactor operating at 100 megawatts eventually builds up a stockpile of 4.5 megacuries of Sr^{90}. The time required to accumulate this much Sr^{90} will be two to three times the half-life of Sr^{90} or 40 to 60 years.

C. Use of Fission Products

Much heat is evolved as fission products decay and so megacurie amounts of fission products are useful as heat sources as may be seen in Figure 20. A great variety of other uses for fission products are being investigated. Figure 32 shows a list of possible uses, many of which have been investigated. Such a listing is of temporary validity, since new uses are rapidly being discovered.

FIG. 32

POSSIBLE USES FOR FISSION PRODUCTS



SOURCE: Industrial Uses Of Radioactive Fission Products
Stanford Research Institute

5. CONVERSION FACTORS

To Convert From	To	Multiply By
Ergs	British Thermal Units	9.481×10^{-11}
Ergs	Gram Calories	2.389×10^{-8}
Ergs	Joules	10^{-7}
Ergs per second	Kilowatts	10^{-10}
Million Electron Volts	Ergs	1.60×10^{-6}
Million Electron Volts	Killowatt-hrs	4.44×10^{-20}
Million Electron Volts	British Thermal Units	1.51×10^{-16}
Mev	Megawatt-days	1.85×10^{-24}
10^5 curies at 0.7 mev. per disintegration	Kilowatts	0.41
10^5 curies at 0.7 mev. per disintegration	Btu per hour	1400
10^5 curies at 0.7 mev. per disintegration	Lbs. steam per hr. at 1,000 Btu per lb.	1.4
Curie	Disintegrations per second	3.7×10^{10}
Rad	Ergs per gram	100
Roentgen	Rad	0.83

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