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REVIEW OF LOW-MASS ATOM PRODUCTION
IN FAST REACTORS

by

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ABSTRACT

This report presents a literature review of low-mass charged-particle yields in fission. A semiempirical method for calculating the yields of hydrogen and helium isotopes from fast-neutron fission of uranium and plutonium is presented. Total and specific production rates of ^1H , ^2H , ^3H , ^3He , ^4He , and ^6He are estimated for the reactor fuels ^{233}U , ^{235}U , and ^{238}Pu . The tritium production in reactor fuels is estimated at 327 Ci per metric ton of ^{235}U fuel and 440 Ci per metric ton of a 50% mixed ^{235}U - ^{239}Pu thermal reactor fuel having a burnup rating of 20,000 MW(t)-D. Tritium production in fast reactors is estimated to be 10% lower for ^{235}U fuel and 10% higher for ^{239}Pu fuel than for thermal reactors.

An examination of all sources of hydrogen and helium in EBR-II is presented. Neutron reactions of stainless steel are shown to produce 90% of the hydrogen and 10% of all helium produced in EBR-II. Tritium production in fast reactors is shown to result almost entirely from fission. A plutonium-fueled fast reactor is estimated to produce nearly twice as much tritium as a uranium-fuel thermal reactor.

I. REVIEW OF LOW-MASS CHARGED-PARTICLE YIELDS IN FISSION

A. Introduction

It is well known that a nucleus undergoing fission divides generally into two massive fragments of more or less similar masses. Dynamically it is possible for a fissioning nucleus to separate into three fragments. In this case the three fragments may be of comparable size (ternary fission) or may result in two massive fragments accompanied by a third low-mass charged-particle (third-particle emission). Third-particle emission is the most predominant mode of triple fission; however, it is a much rarer event than binary fission.

Low-mass charged-particles formed by the fission process have recently become of considerable interest. This interest has been generated

because of the relationship of this relatively rare process to the theory of fission, because development of particle-identification techniques have become sufficiently sophisticated that such processes are experimentally accessible, and because of the concern over the practical implications of third-particle yields to the development of fuel-cycle systems for fast reactors. It is the intent of this communication to review the area of low-mass charged-particle production by the fission process, primarily for the purpose of acquainting workers in reactor development with the current theoretical and experimental status of this field. Secondarily, we shall be concerned with interpreting the theoretical and experimental data in a manner which will enable some predictions of third-particle yields of particular interest to reactor technology. Thirdly, it is hoped that this summary will help to clarify the rather large quantity of work reported in this field and provide direction as to the need for further theoretical and experimental information.

The low-mass particles of concern to reactor development are tritium, alpha particles, and to a lesser extent ^1H , ^2H , ^3He and ^6He . Tritium as a fission product is of particular importance in power reactors because its half-life (12.3 yr) does not permit its practical reduction by allowing for radioactive decay. The present nuclear fuel-processing plants (aqueous process) dispose of tritium by environmental dispersion. It is estimated that by the year 2000, thermal reactors alone will be producing tritium at an annual rate in excess of 10 million Ci.¹ Fuel-processing schemes must be able to dispose of these quantities of tritium (estimated at between 130^2 to 260^1 Ci/ton of standard fuel) in a biologically and radiologically acceptable manner. If the tritium yields are of the order of 0.04% in future fuels (550 Ci/ton) the present fuel-processing procedures may become unacceptable.

Other gaseous hydrogen and helium isotopes are of concern to reactor development because of their interaction with fuel cladding material and the fuel matrix. Wier³ has described a mechanism in which helium produced within a metal can collect as bubbles in grain boundaries to form large cracks leading to fracturing of the metal. He found that helium in an atom ratio of 10^{-7} was sufficient to reduce the ductility of Type 304 stainless steel to one-half of its normal value. Such a collection of gas bubbles in reactor fuel is known to result in swelling, disruption of the fuel matrix, and embrittlement of cladding materials, all of which represent limitations to the attainment of high-burnup fuels.

In addition to the physical-mechanical damage by gaseous generation and the problems associated with radioactive waste disposal, the very high operating temperatures of future fast breeder reactors may result in chemical problems. Hydrogen and lithium atoms produced from fission or from fast-neutron reactions may result in deterioration of reactor-material properties which are not experienced in thermal reactors. The deleterious

effects of hydride formation on the mechanical properties of metallic fuels, zirconium, vanadium, and titanium are well documented.

The mechanism of formation of low-mass particles in fission is of fundamental importance in understanding the fission process and is related to the theory of matter in general. Present information strongly suggests that the light particles are released in the space between the two separating primary fragments very nearly at the time of the actual scission of the fissioning nucleus. Studies of the absolute and relative yields, the energy, and angular distribution of the emitted particles can be expected to provide information concerning the configuration of the fissioning nucleus at their time of origin, properties concerning the neck at the time of scission, and the clustering probabilities of the various nuclear configurations within the neck. One can also speculate that information concerning nuclear viscosity as a function of internal energy, tensile strength of nuclear matter, and nuclear charge distributions may be revealed by studies of these light particles.

B. Summary of Experimental Observations

The predominant type of low-energy fission is a breaking of the fissioning nucleus into two more or less equal fragments plus the emission of several neutrons. The possibility of ternary fission has been demonstrated both experimentally and theoretically by studies too numerous to mention. Several types of ternary fission have been classified according to the distribution of mass among the three fragments. The types of ternary fission of concern to this review involves the emission of two more or less equal massive fragments plus the emission of a third particle of low Z (generally 4 or less).

Third-particle emission in fission has not been studied as extensively as many of the other aspects of fission. One reason is that the relative rarity of the events of interest makes data collection by usual methods very slow and inexact. Until very recently mass-identification techniques were not sufficiently developed to provide the necessary discrimination for the wide range of intensities and energies of the emitted light particles.

The most abundant and well established third-particle event involves the emission of an energetic alpha particle. The first reported observation of long-range alpha particles was by Tsien, Ho, Chastel, and Vigneron⁴ in 1946. Since then, these particles have been studied by measurements utilizing nuclear emulsions,⁵⁻⁷ ionization chambers,^{8,9} magnetic analysis,¹⁰ and counter telescopes of several varieties.^{9,11-15} The literature has been reviewed by several authors,¹⁶⁻¹⁸ the most recent being an excellent review by Hyde,¹⁹ who gives a comprehensive summary of experimental values for the frequency of alpha emission for data reported up to 1963.

The available data for alpha-particle yields (α/f) in low-energy fission are numerous in quantity but considerable in spread. The spread in the data far exceeds the cited experimental errors quoted for individual measurements. For example, the most extensively studied reaction is thermal-neutron fission of ^{235}U , for which seven values are reported ranging from 4.35×10^{-3} to 1.95×10^{-3} , with individually quoted errors of typically less than 10%. Similarly, for spontaneously fissioning ^{252}Cf the average value for six measurements is 3.05×10^{-3} , but the spread in values exceeds 50% of the average.

Tables I and II summarize the alpha-particle yields (the number of alpha particles emitted per fission event) for all reported fissionable nuclei. Table I presents the neutron-induced and spontaneous-fission data, and Table II presents the results for charged-particle-induced fission. The data used in this compilation have been taken from Refs. 5-36. The yields have been listed as a function of excitation energy of the fissioning (compound) nucleus. The excitation energy was computed as the sum of the kinetic energy of the incident particle plus its binding energy. With two exceptions, the values given in Table I are averages of all available measurements for cases where more than one measurement has been reported. The values given for spontaneous fission of ^{252}Cf and the lowest-energy (thermal-neutron) ^{235}U results are considered the best of the reported data, and differ from the average values by 6% for ^{252}Cf and 15% for ^{235}U .

TABLE I Summary of Alpha-particle Yields (α/f) from Neutron-induced Fission

Excitation Energy (MeV)	Target	Fissioning Nucleus							
		^{252}Cf	^{242}Pu	^{240}Pu	^{239}U	^{236}U	^{235}U	^{234}U	
Spontaneous Fission	-	3.25×10^{-3}	2.75×10^{-3}	3.18×10^{-3}					1.93×10^{-3} 3.14×10^{-3}
4.8	^{238}U				3.3×10^{-3}				
6.3	^{241}Pu		2.5×10^{-3}						
6.4	^{239}Pu			2.3×10^{-3}					
6.4	^{234}U						2.1×10^{-3}		
6.5	^{235}U					2.0×10^{-3}			
6.9	^{233}U							2.4×10^{-3}	
7.3	^{238}U				1.7×10^{-3}				
7.4	^{239}Pu			2.2×10^{-3}					
7.5	^{235}U					1.9×10^{-3}			
9.0	^{235}U					1.7×10^{-3}			
9.5	^{235}U					1.3×10^{-3}			
18.8	^{238}U				0.9×10^{-3}				
20.5	^{235}U					2.0×10^{-3}			

TABLE II Summary of Alpha-particle Yields (α/f) from Charged-particle-induced Fission

Excitation Energy (MeV)	Incident Particle	Target	Fissioning Nucleus			
			^{242}Pu	^{236}U	^{239}Np	^{233}Pa
15.8	p	^{238}U			1.7×10^{-3}	
15.9	p	^{232}Th				1.5×10^{-3}
22.7	p	^{238}U			2.7×10^{-3}	
24.5	α	^{238}U	2.6×10^{-3}			
24.9	α	^{232}Th		2.0×10^{-3}		
37.0	α	^{238}U	3.5×10^{-3}			
37.4	α	^{232}Th		2.7×10^{-3}		

Coleman *et al.*,³² Schroder,^{34,43} Atneosen,¹⁵ and Nobles⁹ have concerned themselves with the production rate of alpha particles as a function of excitation energy. In Fig. 1, the data of Tables I and II are plotted to show this dependence. The general features of this plot indicate a rapidly decreasing α/f ratio with increasing excitation energy up to about 15 MeV, at which the ratio starts to rise rapidly with a further energy increase. Schroder *et al.*,^{34,43} carefully examined the α/f ratio of $^{235}\text{U} + n$ over the neutron energy range between 0.025 and 0.3 eV and found it constant within experimental error. They conclude that the spread in reported results from thermal-neutron fission of ^{235}U cannot be explained by any dependence on the neutron energy. Considering the inconsistency of the data, one can assume from Fig. 1 that the ternary-to-binary ratio decreases at a rate of about 25% per MeV increase in excitation energy for neutron-induced fission over the neutron energy range found in nuclear reactors. The indications are that the ratio becomes constant above an excitation energy of 40 MeV.¹⁵

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Fig. 1. The Number of Alpha Particles Emitted per Fission as a Function of the Excitation Energy of the Fissioning Compound Nucleus. The points represent averages where more than one measurement has been reported. The data are taken from Refs. 5-36.

The second most abundant low-mass ternary product is tritium. Considerably less data are available for this mode of third-particle emission than for the emission of alpha particles. Albenesius³⁷ first reported finding tritium in samples of natural uranium which had been irradiated in a thermal reactor. Albenesius and Ondrejcin³⁸ found convincing evidence that the tritium was due to fission and, based upon known reactor flux rates, estimated a yield of approximately 1×10^{-4} for natural uranium. Sloth *et al.*³⁹ radiochemically determined a tritium yield of 0.8×10^{-4} for ^{235}U fission by thermal-neutron irradiations of the sample contained in a platinum vial. These three measurements were made by a radiochemical extraction of the tritium from the target material and subsequent determination of the quantity of tritium by means of liquid scintillation counting. No

corrections were made for adsorption, migration, or diffusion of the tritium even though under certain conditions, such as reactor environments, tritium can readily diffuse through platinum. It can be assumed that these radiochemical results represent a lower limit for tritium yields. Horrocks⁴⁰ has radiochemically determined the tritium yield of spontaneously fissioning ^{252}Cf to be 2.2×10^{-4} tritium atoms per fission. This experiment was not subjected to the severe environmental conditions experienced in the reactor irradiations.

Tritium yields have been measured utilizing $dE/dX \times E$ particle identification techniques. Marshall and Scobie³⁵ used a gas proportional ΔE detector to find a value of 1×10^{-4} for thermal-neutron fissions of ^{235}U . Watson¹¹ used a scintillation-type ΔE detector to obtain a T/f ratio of 2.2×10^{-4} for spontaneous fission of ^{252}Cf , in good agreement with Whetstone and Thomas¹³ who report a value of 1.9×10^{-4} .

The most definitive experiments to date in which all low-mass charged particles emitted in fission are detected in a single experiment have exploited the superior mass resolution obtainable with semiconductor counter telescopes and two-dimensional data analysis.^{13,14,41} Whetstone and Thomas¹³ have reported ^{252}Cf yields for ^1H , ^2H , ^3H , ^3He , ^4He , ^6He , ^8He , Li, and Be, utilizing this particle-identification technique in which the identified particle is only recorded when in coincidence with a fission fragment. Cosper, Cerny, and Gatti¹⁴ have made similar measurements in an experiment designed to demonstrate the powerful mass, energy, and particle-identification capability of using a three-semiconductor telescope array in which identification is electronically determined based on relative energy loss in the three detectors. The results of these latter two experiments are given in Table III. The nine identified particles, their yields, maximum observed energies and most probable energies are summarized for spontaneous fission of ^{252}Cf .

TABLE III. Light-particle Data for Spontaneous Fission of $^{252}\text{Cf}^{(a)}$

Particle	X/f	Maximum Energy (MeV)	Most Probable Energy (MeV)
^1H	5.1×10^{-5}	17	9
^2H	2.0×10^{-5}	18	7
^3H	1.9×10^{-4}	20	8
^3He	$<2.9 \times 10^{-5}$	31	17
^4He	3.27×10^{-3}	34	16
^6He	7.8×10^{-5}	28	13
^8He	5.9×10^{-5}	24	<13
^{10}He	3×10^{-7}	-	-
Li	3.9×10^{-6}	32	-
Be	$>3 \times 10^{-7}$		

(a) Data taken from Refs. 13 and 14.

The angular distributions of emitted alpha particles have been studied in several laboratories.^{6,15,24,30,43-46} In one of the earliest experiments Titterton⁶ detected a strong peaking of the angular distribution of alpha particles at 82° with respect to the lighter of the massive fission fragments during ^{235}U fission. Muga²⁴ found similar results in ^{252}Cf fission. The results of Ramanna *et al.*³⁰ are the most unusual. They found in 14-MeV-neutron fission of ^{238}U that the alpha particles were emitted perpendicular to the massive fission fragments, but had an angular distribution peaked at forward and backward angles with respect to the beam and a $90/180^\circ$ differential-cross-section ratio of 1:10. In contrast, Atneosen *et al.*,¹⁵ and Perfilov⁴⁶ found the alpha particles to be peaked at near 90° with respect to the beam.

Atneosen *et al.*¹⁵ have made a detailed study of the angular correlations between the emitted alpha particles and the two massive fragments for spontaneous fission of ^{252}Cf and 17.5-MeV-proton fission of ^{238}U . The angular correlation was found to be independent of the angle with respect to the incident proton beam and independent of the azimuthal angle. However, the angular correlation for $p + ^{238}\text{U}$ was 14° broader than for ^{252}Cf . Since ternary-to-binary ratios are often determined by comparing the coincidence counting rate (alpha-fission fragment coincidence) of the unknown target to the coincidence counting rate of ^{252}Cf , corrections must be applied if the width of the angular correlations are different. In Atneosen's case, ^{239}Np , this correction resulted in a 29% increase in the ternary-to-binary ratio. Normalizing their angular-correlation results with respect to the direction of the lighter fragment, they find a full width at half-maximum between the alpha particles and the fission fragments to be 33 , 42 , and 47° for ^{252}Cf , thermal-neutron fission of ^{235}U , and 17.5-MeV-proton fission of ^{238}U , respectively.

The effects of alpha-particle emission upon the mass distribution of the heavier primary fragments has been examined in a few experiments.^{24,45,47-50} The results consistently showed that the low-mass side of both fragment peaks were identical in third-particle emission and binary fission. The high-mass sides of the peaks were displaced towards lower values in the case of alpha particles accompanying fission. In addition, the sum of the kinetic energies of the three particles in ternary fission appeared to be very close to the sum of the kinetic energies of the two fragments in binary fission.^{45,48}

Summarizing the results of studies of low-mass ternary fission, the following conclusions are reasonably well-substantiated and must be reconciled for a comprehensive understanding of the mechanism of formation:

1. The relative frequency of emission in low-energy fission is $^4\text{He} > ^3\text{H} > ^6\text{He} > ^1\text{H} > ^2\text{H} > ^3\text{He} > ^8\text{He}$.

2. The ternary-to-binary fission ratio decreases rapidly with increasing excitation energy up to about 15 MeV. At higher energies the ratio increases.

3. Alpha particles are emitted nearly perpendicular to the direction of emission of the two heavier fragments.

4. The most probable value for the energies of emission are in the order ${}^3\text{He} > {}^4\text{He} > {}^6\text{He} > {}^8\text{He} > {}^1\text{H} > {}^3\text{H} > {}^2\text{H}$.

5. The total kinetic energy release obtained by summing all particles is essentially identical for ternary and binary fission.

6. The high-mass side of the mass distribution peaks are displaced to lower values in ternary fission than in binary.

7. The yield of alpha particles is not strongly dependent upon the masses of the fissioning nucleus nor consistent with the fissionability parameter Z^2/A .

C. Discussion of Models

Many authors have speculated on the mechanism of low-mass ternary fission.^{30,45,47,48,50-54} Of the models of ternary fission which have been proposed, only those which relate to yields of emitted particles will be discussed in detail. The attempts to relate third-particle fission models to fundamental nuclear properties have been relatively unsuccessful to date and are not within the framework of this review.

Tsien⁵¹ first attempted to explain the angular distribution of ternary alpha particles in terms of a classical model. He assumed a configuration of three contiguous spheres in which the center of the alpha particle is not on a straight line connecting the centers of the two massive fragments, and that the initial velocity of the alpha particle is equal to zero. Tsien concluded that the alpha particles are emitted from a region between the fragments, thus explaining the kinetic energy and direction of motion of the alpha particle in terms of the effect of the Coulomb field of the three fragments. He also concluded that the alpha-particle emission takes place while the fragments are still close together.

Guylikman and Khlebnikov⁵⁴ refined Tsien's model by assuming the alpha particle is formed with a given probability (which they do not discuss) as soon as the diameter of the neck reaches the same magnitude as the diameter of the alpha particle. The alpha particle is thus located, by this interpretation, on a straight line connecting the centers of the two massive fragments. During deformation of the fissioning nucleus they calculate that the Coulomb barrier for alpha emission in the neck region is lowered

20-25%. Since the time of descent of the nucleus from the saddle point to the rupture point is considerably longer than the nuclear transit time, the deformation process of the nucleus is adiabatic, so the kinetic energy of the alpha particle remains constant. The departure of the alpha particle leads eventually to rupture and to formation of the three fragments.

They calculate the energy and angular distribution of ternary alpha particles from the equations of motion of the two massive fragments and the alpha particle for the case of the most probable mass ratios of the two primary fragments, using experimentally derived fragment velocities. Under the assumption that the alpha particles are emitted when their kinetic energy is about 1.0 MeV above the barrier, they find reasonable agreement with Perfilov's⁴⁶ data for the energy and angular distribution of alpha particles emitted from spontaneous fission of ^{252}Cf and thermal-neutron fission of ^{235}U . This model makes no attempt to explain the observed alpha-particle yields nor does it permit such predictions to be calculated in any straightforward manner.

Interpretations of the mass distributions obtained from third-particle fission when compared to binary fission have been used to infer a third-particle fission mechanism.^{45,48,50,53} From a comparison of data for third-particle and binary-fission mass distributions of ^{235}U , Schmitt and coworkers⁴⁸ concluded that the alpha particle is emitted predominantly at the expense of the nucleons in the light fragment for more near-symmetric fission and at the expense of nucleons in the heavy fragment for more asymmetric fission. They further note a strong influence of closed shells on the mass distribution.

Schmitt and Feather⁵⁰ have analyzed the same mass-distribution data. They assumed a rapid two-step process in which the nucleus first breaks into two fragments, followed by emission of an alpha particle from one of the two massive fragments. The yield $Y_T(M-4)$ of the fragment of mass M which emits the alpha particle in third-particle fission is equal to the binary yield of the fragment times the probability of alpha-particle emission, $P_\alpha(M)$. For the case of the fissioning compound nucleus ^{236}U they derived

$$P_\alpha(M) = \frac{Y_T(236-M)}{Y_B(236-M)}. \quad (1)$$

By assuming a single-step, three-body breakup, they considered alpha-particle formation at the expense of $\underline{m}$ nucleons in one fragment and of $\underline{n}$ nucleons in the other, resulting in a series of probabilities $P_n(M)P_m(M)$. It is not clear that even, in principle, one can decide about these probabilities.

Doan, Carles, and Chastel⁵⁵ have made some quantitative calculations based upon Schmitt and Feather's model. From Eq. 1, the third-particle mass-distribution data of Ref. 48, and the $P_\alpha(M)$ values calculated from Ref. 50, they determined the alpha particle emission probability as

$$P_\alpha = P_\alpha(M_1) + P_\alpha(M_2), \quad (2)$$

where M_1 and M_2 are the complementary masses of the two fragments for binary fission of ^{235}U . They then obtained P_α as a function of mass ratio $R = M_2/M_1$. The third-particle to binary ratio was calculated to be

$$\frac{\alpha}{f} = K \frac{\int_0^{R_{\max}} F_B(R) P_\alpha(R) dR}{\int_0^{R_{\max}} F_B(R) dR}, \quad (3)$$

where $F_B(R)$ is the mass distribution of binary fission of ^{235}U as a function of the mass ratio R , and K is a normalized constant used to force $(\alpha/f)_{\text{calc}}$ to equal $(\alpha/f)_{\text{exp}}$ for ^{235}U . Using this K and $P_\alpha(R)$, they calculated the α/f ratio for ^{233}U , ^{239}Pu , and ^{241}Pu thermal-neutron fission to be 2.15×10^{-3} , 2.22×10^{-3} , and 1.93×10^{-3} , respectively, which, with the exception of the value for ^{241}Pu , is in very good agreement with experimental results.

The objection to this method as pointed out by the originators Schmitt and Feather,⁵⁰ is the redundancy between $P_\alpha(M_1)$ and $P_\alpha(M_2)$, which means $P_\alpha(M_2)$ can be generated from $P_\alpha(M_1)$. Furthermore, experimental values of α/f for these calculated nuclei are all very similar; therefore, by forcing K to fit one set, and varying only $F_B(R)$, which is also similar for all nuclei, one expects the other sets to give good agreement.

Fraenkel and Thompson⁴⁵ have examined the angular correlation of alpha particles in coincidence with two fission fragments for spontaneous fission of ^{252}Cf and observed a shift in the most probable direction of emission of the alpha particle towards the heavy fragment as the mass ratio increases. Assuming that the angular distribution of the alpha particle arises predominantly from Coulombic forces, they concluded that the most probable point of emission is close to the heavy fragment for near-symmetric fission and close to the light fragment for very asymmetric fission. This conclusion would appear to be in conflict with Schmitt's⁴⁸ conclusion if the alpha particle is emitted at the expense of the fragment to which it is nearer at the time of emission.

Fraenkel and Thompson further argue that if the alpha particles are emitted at the point of scission and if scission is close to a given fragment, then the average number of neutrons, $\bar{\nu}$, emitted by this fragment in binary

fission will be small and the angle of the alpha particle with respect to the direction of this fragment in ternary fission will be large. This anticorrelation between $\bar{\nu}$ and the angle of the emitted alpha particle supports the supposition that the configuration of the nucleus during the scission process is similar in binary and third-particle fission.

In order to examine the basis of the above arguments, we briefly turn to the problem of neutron emission in fission. The average number of neutrons, $\bar{\nu}$, emitted in binary fission as a function of mass have been measured in several experiments.⁵⁶⁻⁶⁰ The observations of relevance to this discussion can be summarized as follows:

- (1) The neutrons appear to be evaporated isotropically from the fragments after scission.
- (2) The shape of the energy spectra of emitted neutrons is rather independent of the mass of the fragment and the kinetic energy release in fission.
- (3) A smooth decrease is observed in the number and energy of neutrons as a function of the kinetic energy release for a given fragment mass.
- (4) A very strong variation in $\bar{\nu}$ with the mass of the fragment (sawtooth distribution) is found with as many as four neutrons being emitted for the largest of the light and heavy fragments, but less than one neutron for the smallest of the two fragments.

These observations led Whetstone⁵⁶ to visualize the fissioning nucleus prior to fission as two relatively large but unequal volumes connected by a long neck (asymmetric dumbbell). If the neck breaks in the middle, asymmetric fragments with approximately equal distortion energy results and the number of neutrons emitted from each fragment would be equal for this, the most probable mass distribution. If the neck breaks near the lighter fragment, the heavy fragment will retain most of the distortion energy and emit most of the neutrons resulting in very asymmetric fission. Conversely, for near-symmetric fission the break occurs near the heavy volume, and neutrons are emitted predominantly from the lighter fragment.

Whetstone's description may be used to resolve the apparent conflict as to which fragment pays the price of alpha emission. If the alpha particle is assumed to be emitted at the point of scission but from the long neck region (for illustration visualize backlash from the neck), then Schmitt's conclusion that the alpha particle is emitted at the expense of the heavy fragment in asymmetric fission is consistent with Fraenkel's conclusion that it is emitted near the light fragment. This implies that the alpha particle will be emitted predominantly at the expense of the fragment with the greatest distortion energy, as are the prompt scission neutrons.

Further, one might expect that the most probable mass division would lead to the fewest number of alpha particles. The alpha-particle yield would be greatest for very asymmetric or nearly symmetric divisions for which the neck is the largest. Thus, one would expect a correlation between the alpha-particle yield and $\bar{\nu}$.

The correlation can be seen on an integrated scale in Fig. 2, where the alpha-particle yield α/f is plotted versus $\bar{\nu}$ for five thermal-neutron-fissioning species. The total probability of alpha-particle emission (see Eq. 2) may be compared to the number of neutrons emitted by examining the correlation of these two parameters as a function of mass ratio. This comparison is shown in Fig. 3 for thermal-neutron fission of ^{235}U . The two minima at R equal to 1.27 and 1.65 have been attributed to shell effects.^{50,55} For comparative purposes the $\bar{\nu}$ versus R for ^{252}Cf is also shown.

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Fig. 2. The Number of Alpha Particles Emitted per Fission vs the Average Number of Prompt Neutrons Emitted per Fission for Thermal-neutron-induced Fission. Data taken from Refs. 56-60.

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Fig. 3. The Calculated Probability of Alpha-particle Emission as a Function of the Heavy Mass to the Light Mass Primary Fission Fragment Ratio. For comparative purposes the average number of prompt neutrons as a function of primary fragment mass ratio for spontaneously fissioning ^{252}Cf and thermal-neutron fission of ^{235}U is also shown. The prompt-neutron data is taken from Ref. 48.

Feather⁵¹ has made a similar analysis of data to conclude that the alpha particles are liberated in a two-step process of scission, followed

by emission from the heavy fragment exclusively. He concluded that the emission probability from low-yield fragments with small neutron excess is near unity. Ramanna *et al.*³⁰ have argued that third-particle fission is less abundant than binary fission because of a decrease in excitation energy of the system at the scission point. They propose that alpha-particle emission occurs during statistical equilibrium and that alpha-particles are evaporated prior to fission but after neutron evaporation. In an attempt to explain the angular distribution of the alpha particles, they had to assume that the barrier is considerably lowered at the neck and that alpha-particles are emitted with very little kinetic energy. It is difficult for a statistical model to explain the relative yields of other low-mass particles as well as the peaking of the angular distribution perpendicular to the direction of the massive fragments.

Halpern,⁵³ in a comprehensive analysis of alternatives, considers the problem of third-particle fission as consisting of two more or less independent parts. The first part relates to the mechanism leading to the release of the alpha particle into the region between the fragments, and the second to the motion of the alpha particle in the Coulomb field of the fragments. It is the release mechanism that is of most concern to predictions of low-mass particle yields.

Halpern's explanation is based on the sudden approximation or a nonadiabatic change of nuclear potential. In this model the particle X is originally contained in a potential well. At scission a very rapid potential change occurs in a time which is much shorter than the time required for the particle to traverse the width of the nucleus. This can be viewed from a liquid-drop description as a drop with large deformation energy stretching into a dumbbell-like structure which eventually breaks at the neck region. From classical hydrodynamics, Hill and Wheeler⁶² have shown that such a drop breaking into two primary fragments could be expected to form small droplets in the neck region at the point of scission.

For a third particle to be released in a nonadiabatic process, it must acquire a large amount of energy to be released between the fragments. The energy associated with fission is usually considered to be in the form of nuclear distortion prior to fission and released as distortion energy to the individual fragments immediately after fission. On rare occasions this distortion energy can be made available to a third particle. Because such an event requires considerable energy, it is expected that third-particle emission is more probable when there is an abnormally long stretch before scission. The energy transfer to the third particle probably takes place through the sudden snap of the neck stubs after scission rather than a thinning of the neck on either side of the particle by a retraction of the stubs. Thus, one would expect third particles to be associated with low kinetic energy of the two primary fragments. If Whetstone's postulation of an asymmetric dumbbell is correct, Halpern's model would predict third particles to be more probable from symmetric and very asymmetric fission rather than from events leading to the most probable mass distribution.

Halpern has made estimates of the release energy E_R required for third-particle emission by comparing the energy of a pair of fragments from which particle X has been removed and placed midway between the fragments. The removal energy is calculated as being equal to the energy difference between these two situations and is given by

$$E_R = B_X + V_X + K, \quad (4)$$

where B_X is the average binding energy of any particle X to each of the two fragments, V_X is the Coulomb potential of the centrally placed particle with respect to the fragments, and K is the amount of kinetic energy with which X is initially released. Details of actual calculation will be given later, but in general is about 23 MeV for alpha-particle release.

Fuller⁵² has treated, in detail, the problem of a nonadiabatic nuclear potential change for neutron emission. He considers the original nucleus as a square well containing a Fermi sea of nucleons. During scission an inverted square well, or "volcano," rises and divides the original well into two idealized fission fragments. From the shape of the rising well, the rate of rise, and depth of the true nuclear well, he calculates the probability of imparting sufficient amounts of excitation energy to a neutron to emit it from the nucleus. He compares an adiabatic (no energy imparted during scission) to a sudden-impulse approximation in which scission must occur in less than 1.5×10^{-23} sec. From this, the probability of a neutron acquiring sufficient energy to be promoted to a nonbound state and emitted can be calculated. He finds that by assuming the shape of a square well with rounded edges and a scission time of approximately 2.5×10^{-23} sec (intermediate time between adiabatic and sudden impulse) he can qualitatively account for the number of neutrons emitted during scission for the spontaneous fission of ^{252}Cf . Although a further development of the nonadiabatic calculation for charged particles would be of great interest, the uncertainty of parameters makes specific particle-yield predictions difficult.

Considering the many alternative explanations of third-particle fission, it is clear that few positive conclusions can be drawn but some consensus of opinion may be arrived at. The following represent points of general agreement concerning the third particle accompanying fission:

1. Third particles are formed between the two primary fragments very near the point of scission.
2. Third particles result from abnormally long stretching; the magnitude can be estimated to be about a 15% increase based on kinetic energy release and energy conservation.
3. Absolute yields are related to the total distortion energy and the position of the scission point in the neck region.

4. The energy and angular distributions are understandable in terms of the Coulombic fields.

Further conclusions must await more detailed experiments and the development of a more general theory of fission. The more obvious experimental data which would be desirable are summarized below:

a. Comparisons of ternary and binary mass distributions such as that of Schmitt and coworkers⁴⁸ are of considerable interest. Comparison of alpha, tritium, and ⁶He fission mass-distributions would be useful to elucidate which primary fragment pays the price of the third particle, the influence of distortion energy and point of scission on particle yield, and the influence of shell effects in third-particle fission.

b. Measurements of the angular distribution of particles in addition to alpha particles would contribute to elucidating the point of origin of third particles and gross features of the mechanism, such as the adiabatic or nonadiabatic nature of the release and possibilities of scission followed by evaporation. Angular correlations of various particles with the simultaneous measurement of primary-fragment kinetic energies, such as in Fraenkel and Thompson's⁴⁵ experiment, could be extremely revealing.

c. Perhaps the simplest and presently most necessary experiment is simply to measure absolute particle yields, such as carried out by Whetstone and Thomas¹³ for several fissioning species at different bombarding energies. The dependencies upon the bombarding energy at present are very difficult to understand, largely because only alpha-particle emission has been examined.

This list is by no means exhaustive, and at this point virtually any experimental information is desirable. As Halpern⁵³ has pointed out, one can speculate about the mechanism of third-particle fission; however, the data are "unfortunately still too meagre to permit one to deduce the dependency of even α/f upon more than one variable."

II. LOW-MASS CHARGED-PARTICLE YIELD CALCULATIONS

A. Third-particle Yield Calculations in Fission

The discussion of the previous section concerning mechanisms of third-particle emission in fission has revealed that a theory for such processes is far from complete. At the present time Halpern's description, although intended to be of a qualitative nature, does encompass many of the considerations required to make estimates of relative third-particle yields. The importance of the tritium yield in fission to future fast reactor fuels and to the development of fast reactor fuel-reprocessing schemes has led us to pursue these calculations even though such calculations may be premature at this time.

From Halpern's model, the energy $E_R(X)$ required for release of any particle X is equal to the sum of the separation energy $B(X)$ required to remove X from a fission fragment, the Coulomb potential energy $V(X)$ of particle X placed between the two primary fragments, and $K(X)$, the amount of kinetic energy with which X is initially released. To the release energy $E_R(X)$ is added an empirical term $P(X)$ which correlates the relative yields of each particle X to the values measured in ^{252}Cf by Whetstone and Thomas.¹³ Although the correlation term has been determined on strictly an empirical basis, it will be shown later that the values determined for $P(X)$ can be directly determined from fundamental nuclear properties which are completely consistent with the model. By assuming that $P(X)$ is dependent only upon the emitted particle and not upon the fissioning nucleus, we can arrive at the total emission energy $E_T(X) = E_R(X) + P(X)$ for any particle from any fissioning nucleus. The yield of X can be determined relative to the yield of an alpha particle by a direct comparison of the total emission energies of the two particles.

1. Determination of Parameters

The separation energy $B(X)$ has been determined by averaging the binding energy of particle X to the heavy and to the light primary fragments. Several combinations of the most probable mass divisions reported by Milton and Fraser⁶³ and by Smith⁶⁴ which are thought to represent the primary fission fragments at the time of scission for the various fissioning nuclei were used in calculating each $B(X)$. Mass-defect values of the primary fragments were taken from Cameron⁶⁵ and for each X particle masses were taken from Bainbridge⁶⁶ and from Mattauch *et al.*⁶⁷

The Coulomb potential-energy change $V(X)$ required to give the assumed release configuration has been calculated as

$$V(X) = zV_0(X) \left(\frac{3}{2Z_L} + \frac{3}{2Z_H} - \frac{7z}{4Z_L Z_H} \right), \quad (5)$$

where z is the charge of particle X , Z_H and Z_L are the most probable charge of the primary heavy and light fragments, respectively, and $V_0(X)$ is the Coulomb potential energy of the two massive fragments at scission. Equation 5 has been derived by assuming particle X takes its charge equally from each of the two most probable fission fragments which would have been produced had binary fission taken place. $V_0(X)$ has been determined by subtracting the most probable kinetic energy of each particle (see Table III) from the total kinetic energy released in binary fission for the fissioning nuclei under consideration. As in the calculation of $B(X)$

values, several combinations of Z_H , Z_L pairs have been used to calculate $V(X)$, with the average $V(X)$ value being used in the determination of $E_R(X)$.

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Fig. 4. The Yields of Low-mass Charged Particles from Spontaneously Fissioning ^{252}Cf vs the Calculated Release Energy $E_R(X)$. Yield data are from Ref. 13. See text for description of the calculation.

The average kinetic energy of the particle at the time of release has been assumed to be equal to 2 MeV for all particles.⁶⁸ This compares favorably with a value of 1 MeV determined by Geylikman and Khlebnikov⁵⁴ in fitting angular-distribution data.

A plot of $E_R(X)$ versus particle yield from the spontaneous fission of ^{252}Cf is shown in Fig. 4. As might be expected, the release energy alone does not accurately predict the relative yields. Some of the reasons might include the following. We have assumed that each particle X has been emitted equally at the expense of each of the two primary fragments at the most probable mass division, whereas evidence previously discussed strongly suggests that the X particles are emitted from the lighter fragment for nearly symmetric fission and from the heavier fragment for very asymmetric fission.

One might expect that the initial kinetic energy with which particle X is emitted depends upon how strongly the particle is absorbed in nuclear matter. Thus, it is conceivable that K is a function of X and depends upon the effective volume of nucleons from which it is emitted⁵³ rather than the assumption of a constant value of 2 MeV. Finally, no consideration has been given to clustering or the probability of preformation of X within the neck.

On general grounds one might expect the particle yields $Y(X)$ to have an exponential dependence upon the release energy, being of the form

$$Y(X) = A \exp\left[\frac{-E_T(X)}{T}\right], \quad (6)$$

where $E_T(X) = E_R(X) + P(X)$ if $P(X)$ represents all of the energetic variables not considered in $E_R(X)$; T represents the dependence upon internal excitation energy analogous to a nuclear temperature, and A is a normalization factor. The $P(X)$ values for each third particle may be determined empirically from the ^{252}Cf data of Whetstone and Thomas¹³ by forcing a linear semilog fit in Fig. 4. However, a unique set of $P(X)$ values cannot be determined in this manner.

Halpern⁵³ has pointed out that it is reasonable to assign a momentum spread to an emitted particle corresponding to the nuclear volume from which the particle is emitted, and Fuller⁵² has calculated that non-adiabatic snapping of the neck can contribute energy to neutrons released at scission. On this basis one can calculate the energy associated with a momentum spread of particle X when emitted from a nuclear volume con-

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Fig. 5. The Yields of Low-mass Charged Particles from Spontaneously Fissioning ^{252}Cf vs the Total Calculated Release Energy (see text). Yield data are from Ref. 13.

taining the number of nucleons in the heavy fragment to be 8.0, 8.0, 5.0, 3.0, 3.0, 2.0, 1.0, and 1.0 for the $P(X)$ values of n , ^1H , ^2H , ^3H , ^3He , ^4He , ^6He , and ^6Li , respectively. A plot of particle yields versus $E_T(X)$ based on this set of $P(X)$ values is shown in Fig. 5. The correlation of particle yields from ^{252}Cf with $E_T(X)$ is quite good. This calculation predicts 0.4 scission neutron per fission, which is consistent with the experimental data of Bowman *et al.*^{52,60} and the calculations of Fuller.⁵² The results of Bowman and Fuller indicate a time scale for scission as slower than 2.5×10^{-22} sec,⁶⁰ which makes the assumption of the momentum spread of the emitted particle over a rather large nuclear volume appear more plausible. The value of T derived from the yield data for ^{252}Cf is 1.7 MeV^{-1} which, if T can be interpreted as a nuclear temperature, is somewhat larger than expected from observed neutron spectra in low-energy fission.

It is of interest to consider the implications of some more extreme assumptions. One finds that with very few exceptions the separation energy of X from the heavy fragment is less than from the light fragment. Since the sum of the kinetic energy of the three fragments in ternary fission is essentially the same as the sum of the kinetic energy of the two

fragments in binary fission, one must conclude that the average separation distance between the two primary fragments involving a third particle is 10-15% greater than for binary fission. We have calculated the release energy $E_R(X)$ with the assumptions of a 12% increase in the length of the neck, the X particles are emitted completely at the expense of the heavy fragment, a mass ratio $M_H/M_L = 2.0$, and an initial kinetic energy of 2.0 MeV. The correlation of $Y(X)$ with $E_R(X)$ shown in Fig. 6 is remarkably good. This set of parameters gives $T = 1.3 \text{ MeV}^{-1}$, which is nearer the expected nuclear temperature of about 1 MeV; however, it predicts a scission neutron yield of nearly 10 neutrons/fission, which is an order of magnitude too large.

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Fig. 6. The Yields of Low-mass Charged Particles from Spontaneously Fissioning ^{252}Cf vs Calculated Release Energies Based on Assumptions Discussed in the Text. Yield data are from Ref. 13.

The physical interpretation of T and $P(X)$ is at best speculation; however, the correlation of $E_T(X)$ with third-particle yields from ^{252}Cf does provide a means for estimating yield values for other fissioning nuclei. By assuming that the slope T

and the set of $P(X)$ values are independent of the fissioning nucleus, it becomes possible to calculate $E_T(X)$ values for any particle X. Absolute yields can be determined by normalizing Eq. 6 to the reported alpha-particle yields given in TABLES I and II, and solving for $Y(X)$. Table IV contains a summary of calculated values of $P(X)$, $V(X)$, $E_R(X)$, and $E_T(X)$ for seven low-mass particles emitted from six fissioning nuclei. From these parameters, tritium yields have been calculated for six neutron-induced fissioning materials at several different neutron energies. The calculated tritium yields are compared to experimental results in Table V. The agreement is quite satisfactory.

TABLE IV. Summary of Particle-yield Parameters for Thermal-neutron Fission

Target Nucleus	Parameter	n	^1H	^2H	^3H	^3He	^4He	^6He	^6Li
$^{233}\text{U} + \text{n}$	B(X)	4.9	12.1	14.9	13.2	20.2	4.0	12.1	20.6
	V(X)	-	9.2	9.3	9.2	15.0	15.1	15.3	18.3
	$E_{\text{R}}(\text{X})$	6.9	23.9	26.2	24.2	37.2	21.1	29.4	40.9
	$E_{\text{T}}(\text{X})$	14.9	31.3	31.2	27.2	40.2	23.1	30.4	41.9
$^{235}\text{U} + \text{n}$	B(X)	4.6	12.6	15.1	13.2	20.6	4.5	13.6	21.0
	V(X)	-	9.2	9.3	9.2	15.0	15.1	15.3	18.3
	$E_{\text{R}}(\text{X})$	6.6	23.8	26.4	24.4	37.6	21.7	30.9	41.3
	$E_{\text{T}}(\text{X})$	14.6	31.8	31.4	27.4	40.6	23.7	31.9	42.3
$^{238}\text{U} + \text{n}$	B(X)	4.0	13.4	15.2	13.4	21.7	5.2	11.7	24.6
	V(X)	-	9.2	9.3	9.2	15.0	15.1	15.3	18.3
	$E_{\text{R}}(\text{X})$	6.0	24.6	26.5	24.6	38.7	22.3	28.0	44.9
	$E_{\text{T}}(\text{X})$	14.0	32.6	31.5	27.6	41.7	24.3	29.0	45.9
$^{239}\text{U} + \text{n}$	B(X)	4.9	11.2	13.9	12.3	20.5	4.4	11.9	19.3
	V(X)	-	9.4	9.5	9.4	15.3	15.4	15.7	18.9
	$E_{\text{R}}(\text{X})$	6.9	22.6	25.4	23.7	37.8	21.8	29.6	40.2
	$E_{\text{T}}(\text{X})$	14.9	30.6	30.4	26.7	40.8	23.8	30.6	41.2
$^{241}\text{Pu} + \text{n}$	B(X)	5.6	11.7	15.2	12.3	22.0	4.6	12.0	18.8
	V(X)	-	9.4	9.5	9.4	15.3	15.4	15.7	18.9
	$E_{\text{R}}(\text{X})$	7.6	22.1	26.7	23.7	39.3	22.0	29.7	39.7
	$E_{\text{T}}(\text{X})$	15.6	30.1	31.5	26.7	42.3	24.0	30.7	40.7
^{252}Cf Spontaneous Fission	B(X)	6.1	12.5	16.5	14.0	22.2	5.2	12.3	20.1
	V(X)	-	9.6	9.8	9.7	15.9	16.0	16.3	19.3
	$E_{\text{R}}(\text{X})$	8.1	24.1	28.3	25.7	40.1	23.2	30.6	41.4
	$E_{\text{T}}(\text{X})$	16.1	32.1	33.3	28.7	43.1	25.2	31.6	42.4
All	P(X)	8.0	8.0	5.0	3.0	3.0	2.0	1.0	1.0
	K	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0

TABLE V. Tritium Yields (T/f)

Fissioning Nucleus	Neutron Energy (MeV)	Calculated Yield	Experimental Yield
$^{233}\text{U} + \text{n}$	0	1.1×10^{-4}	
$^{235}\text{U} + \text{n}$	0	1.3×10^{-4}	$1 \times 10^{-4}, ^{(a)} 0.99 \times 10^{-4}, ^{(b)} 0.8 \times 10^{-4} ^{(c)}$
	1.0	1.2×10^{-4}	
	2.5	1.1×10^{-4}	
	3.0	8.2×10^{-5}	
$^{238}\text{U} + \text{n}$	0	2.6×10^{-4}	
	2.5	1.4×10^{-4}	
$^{239}\text{Pu} + \text{n}$	0	2.3×10^{-4}	
	1.0	2.5×10^{-4}	
^{240}Pu	Spon. Fiss.	3.2×10^{-4}	
$^{241}\text{Pu} + \text{n}$	0	2.6×10^{-4}	
^{242}Pu	Spon. Fiss.	3.1×10^{-4}	
^{252}Cf	Spon. Fiss.	2.3×10^{-4}	$1.9 \times 10^{-4}, ^{(d)} 2.2 \times 10^{-4}, ^{(e,f)}$

(a)Ref. 35. (d)Ref. 13.

(b)Ref. 37. (e)Ref. 40.

(c)Ref. 39. (f)Ref. 11.

It is apparent from this discussion that third-particle yields cannot be predicted entirely from theory. By application of energy considerations alone, we have attempted to rationalize existing data into a consistent, although not unique, framework in order to determine unknown yields relative to measured yields. The empirical basis of these calculations, the insensitivity of the calculated yields to the more extreme assumptions, and the good agreement of the calculated to observed tritium yields are encouraging. More exact calculations must await better understanding of the details of the third-particle release mechanism and a more generalized theory of fission.

2. Tritium Production Rates in Nuclear Power Reactors

From the calculated number of tritium atoms produced per fission event, the rate of tritium production in nuclear power reactors can be estimated.

a. Thermal-fission Reactors

For thermal reactors utilizing ^{235}U - ^{238}U as fuel we assume a specific power of 20 kW(t)/kg U for fuel of low ^{235}U content and an energy release of 200 MeV per fission of ^{235}U . For an oxide fuel the number of fissions F are

$$F = \left(\frac{20 \text{ kW}}{\text{kg U}} \right) \left(\frac{10^3 \text{ W}}{\text{kW}} \right) \left(\frac{3.12 \times 10^{-10} \text{ fissions}}{\text{W-sec}} \right) \left(\frac{10 \text{ kg UO}_2}{1} \right) \left(\frac{1}{10^3 \text{ cm}^3} \right) \left(\frac{238 \text{ g U}}{270 \text{ g UO}_2} \right)$$

$$= 5.5 \times 10^{12} \frac{\text{fissions}}{(\text{sec})(\text{cm}^3)}.$$

The specific production rate of tritium is

$$T = \left(5.5 \times 10^{12} \frac{\text{f}}{\text{sec-cm}^3} \right) \left(1.3 \times 10^{-4} \frac{\text{T}}{\text{f}} \right) = 7.2 \times 10^8 \left(\frac{\text{T atoms}}{\text{sec-cm}^3} \right)$$

$$= \frac{\left(7.2 \times 10^8 \frac{\text{T}}{\text{sec-cm}^3} \right) \left(\frac{1.78 \times 10^{-9}}{\text{sec}} \right)}{\left(3.7 \times 10^{10} \frac{\text{dis}}{\text{sec-Ci}} \right)} = 3.5 \times 10^{-11} \frac{\text{Ci(T)}}{\text{cm}^3\text{-sec}},$$

where $1.78 \times 10^{-9}/\text{sec}$ is the decay constant for tritium.

b. Release of Tritium in Fuel-reprocessing Plants

Tritium in irradiated fuel will be released during chemical processing. The quantity of tritium can be calculated from the reference processing load. For example, the reference load of the Nuclear Fuel Services Plant, Inc., in West Valley, New York, is one metric ton/day of fuel

having a burnup rating of 20,000 MW(t)-D/metric ton. The tritium activity is

$$T = \left(\frac{2.7 \times 10^{21} \text{ fissions}}{\text{MW(t)-D}} \right) \left(1.3 \times 10^{-4} \frac{\text{T}}{\text{f}} \right) = 3.5 \times 10^{17} \frac{\text{atoms}}{\text{MW(t)-D}} = 327 \frac{\text{Ci}}{\text{MT}}.$$

Recycling of thermal reactor fuels can result in a buildup of ^{239}Pu which is expected to approach 50% of the fissile atoms; future reactors may employ ^{233}U as the driver fuel. An estimate of the expected tritium production for ^{233}U , ^{235}U , and ^{239}Pu fuels in thermal reactors is 1.4×10^{-2} , 1.7×10^{-2} , and 2.8×10^{-2} Ci of tritium per MW(t)-D, respectively. A metric ton of reference fuel containing equal atom percent of ^{239}Pu and ^{235}U could be expected to yield 440 Ci of tritium.

c. Other Low-mass Charged-particle Production Rates in Thermal Reactors

In addition to tritium, we can use the same methods to calculate the fission yields of other low-mass charged-particles expected to be produced in thermal reactor fuels. The results of these calculations are given in Table VI, in which particle yields, specific production rates in units of atom/cc-sec of oxide fuel, and total atoms produced per MW(t)-D are given for the three most probable power reactor driver fuels: ^{233}U , ^{235}U , and ^{239}Pu . Also summarized in Table VI are the total number of gaseous atoms (hydrogen and helium) and the number of low-mass chemically reactive atoms (hydrogen and lithium) produced from fission.

TABLE VI. Summary of Thermal Reactor Production Rates due to Fission

A. Calculated Particle Yields for Thermal-neutron Fission							
Nucleus	Particles Produced per Fission Event						
	^1H	^2H	^3H	^4He	^6He	^6Li	
^{233}U	1.4×10^{-5}	1.5×10^{-5}	1.1×10^{-4}	2.4×10^{-3}	3.5×10^{-5}	3×10^{-8}	
^{235}U	1.3×10^{-5}	1.6×10^{-5}	1.3×10^{-4}	2.0×10^{-3}	1.6×10^{-5}	5×10^{-8}	
^{239}Pu	3.2×10^{-5}	3.5×10^{-5}	2.3×10^{-4}	2.3×10^{-3}	4.3×10^{-5}	1×10^{-7}	
B. Atoms/cc-sec for Specific Power of 20 kW(t)/kg in Oxide Fuel							
Fuel	Specific Production Rate (atoms/cc-sec)						
	^1H	^2H	^3H	^4He	^6He	^6Li	
^{233}U	7.7×10^7	8.3×10^7	6.1×10^8	1.3×10^{10}	1.9×10^8	2×10^5	Gaseous Atoms ~ 1.4×10^{10} Reactive Atoms ~ 7.7×10^8
^{235}U	7.0×10^7	8.9×10^7	7.2×10^8	1.1×10^{10}	8.9×10^7	3×10^5	~ 1.2×10^{10} ~ 8.8×10^8
^{239}Pu	1.7×10^8	1.8×10^8	1.2×10^9	1.2×10^{10}	2.3×10^8	5×10^5	~ 1.4×10^{10} ~ 1.6×10^9
C. Atoms/MW(t)-D							
Fuel							
	^1H	^2H	^3H	^4He	^6He	^6Li	
^{233}U	3.8×10^{16}	4.0×10^{16}	3.0×10^{17}	6.5×10^{18}	9.4×10^{16}	8×10^{13}	Gaseous Atoms ~ 6.9×10^{18} Reactive Atoms ~ 3.8×10^{17}
^{235}U	3.4×10^{16}	4.3×10^{16}	3.5×10^{17}	5.4×10^{18}	9.3×10^{16}	1×10^{14}	~ 5.8×10^{18} ~ 4.3×10^{17}
^{239}Pu	7.9×10^{16}	8.7×10^{16}	5.7×10^{17}	5.7×10^{18}	1.1×10^{18}	1×10^{14}	~ 6.3×10^{18} ~ 7.1×10^{17}

3. Tritium Production Rates in Fast Reactors

From the design and operating data reported for EBR-II,⁶⁹ it is possible to estimate tritium production rates for fast-breeder reactors. The rated heat output of EBR-II is reported to be 62.5 MW(t). A calculation based on the tritium yield for 1.0-MeV neutrons implies a tritium production rate for ^{235}U fuel of 340 Ci/yr. If ^{239}Pu were used exclusively as the driver fuel, the rate would be about 710 Ci/yr.

In order to calculate the amount of tritium produced in the ^{238}U blanket, it becomes necessary to integrate the fission cross section over the rather broad neutron-energy distribution in which the ^{238}U is located. The exact details of integrating a cross section over the EBR-II flux is described later. We calculate that 3.12 Ci/yr of tritium will be produced from the 188 kg of ^{238}U in the core. Performing the same kind of calculations over the entire EBR-II blanket, we estimate approximately 280 Ci/yr of tritium will be produced from the 28,000 kg of ^{238}U in the blanket.

One could estimate that a 400-MW(t) fast breeder reactor such as FFTF would produce a total of 4000 Ci/yr of tritium utilizing ^{235}U as driver fuel and ^{238}U as blanket material. The same reactor operating with ^{239}Pu fuel would produce about 6200 Ci/yr, and ^{233}U fuel with ^{232}Th blanket would produce about 2500-3000 Ci/yr of tritium.

Reprocessing plants for fast-reactor fuels can expect to handle approximately twice as much tritium from ^{239}Pu fuel as for ^{235}U fuel. Assuming that burnup ratings of fast reactor fuels will be 100,000 MW(t)-D/metric ton, a metric ton of ^{239}Pu fuel can be expected to produce 3150 Ci of tritium activity.

B. Integral Fast-neutron Reaction-rate Calculations for EBR-II

Fast-neutron interactions with structural and coolant materials represent a significant source of low-mass charged-particles in fast reactors in addition to the fission process. The principal reactions responsible for gaseous atom production in fast reactors are (n,p) and (n, α) processes.

Again, using the design and operating data reported for EBR-II,⁶⁹ we calculate the core of EBR-II to contain 1.03×10^5 g of Type 304L stainless steel (SS). From the elemental composition of 304L SS and the isotopic composition of each of the elements, and the total weight in grams of all of the principal isotopes in the core of EBR-II which contribute to charged-particle reactions have been determined.

The production rates of gaseous atoms were then calculated from the following equation:

$$N_X = n \int_{E_{j,L}}^{E_{j,H}} \sigma_X(E_j) \phi(E_j) dE_j, \quad (7)$$

where

N_X = the number of atoms of particle X which are produced per unit time;

n = the number of atoms of an isotopic constituent which is present in the core of EBR-II;

$\sigma_X(E_j)$ = the differential cross section averaged over the neutron-energy interval E_j ;

$\phi(E_j)$ = the neutron flux in the neutron-energy interval E_j .

The differential cross sections $\sigma(E)$ for hydrogen and helium generation have been taken from the neutron cross-section interpretations of Brookhaven⁷⁰ and Karlsruhe,⁷¹ as well as a compilation of experimental neutron excitation functions by Jessen, Bormann, Dreyer, and Newert.⁷² Where available, the recommended values have been used, and in a few cases it has been necessary to interpolate or extrapolate experimental data.

The neutron energy and flux distribution $\phi(E_j)$ has been calculated as follows. A 22-neutron-energy-group, multiregion code thought to represent the neutron spectrum of EBR-II was used as a starting point to obtain the number of neutrons in each energy group as a function of radial distance from the center of the core. The radial distribution was averaged over the entire core in order to obtain the average number of neutrons in each energy group. The averaged distribution was normalized to 3.7×10^{15} n/cm²-sec reported to be present at the center of the core.⁶⁹ The relative flux distribution at the center of the core is shown in Fig. 7.

For the reactions under consideration only the first four energy groups (0.821-10 MeV) contain neutrons with energies above most reaction thresholds. These four neutron energy groups are too broad to take full advantage of the available differential cross sections. Therefore, the number of neutrons in each of the groups were normalized to follow a fission-neutron energy distribution within the group in order to obtain $\phi(E_j)$. The contribution to the overall error by this approximation is thought to be considerably smaller than the errors in the reported differential cross sections.

Fig. 7. The Neutron Distribution Assumed to Represent the Neutron Spectrum of EBR-II at the Center of the Core. The portion of the 22-neutron-energy-group calculation above 10 keV is shown.

The gaseous-atom production rates were then calculated by means of Eq. 7. For calculational purposes, the integral in Eq. 7 was replaced by a summation and the energy increment dE_j was fixed at 0.5 MeV. The lowest energy calculated ($E_{j,L}$), was in all cases the reaction threshold energy, and the upper limit ($E_{j,H}$), was arbitrarily set at 10 MeV, which is the maximum energy in the 22-neutron-group code. A considerable error may be introduced by choosing an energy interval as wide as 0.5 MeV; however, for most reactions the available experimental differential cross-section data do not justify a more elaborate treatment. No corrections have been applied for atom burnup since all calculations have been performed for specific production rates of atoms/sec or atom/MW(t)-D.

The calculated hydrogen and helium production rates are summarized in Table VII. The helium results may be compared to a similar calculation reported by De Pino⁷³ for the neutron spectra of ATR and FFTF reactors. The discrepancies between the two calculations are partially due to the initial assumptions concerning the composition of the stainless steel. For this reason we have included in Table VII the production rate per atom of isotope (columns 3 and 7 for hydrogen and helium, respectively) for the assumed EBR-II neutron-energy distribution. With the exceptions of ^{54}Fe , ^{14}N and ^{10}B , De Pino performed his calculations using fission-spectrum-averaged effective cross sections rather than differential cross sections which were used for all of these calculations. His differential cross-section data for ^{54}Fe were approximately 50% greater than the BNL-325 recommended value. These factors plus the differences between the energy spectra of FFTF and EBR-II reasonably well account for the apparent disagreement.

TABLE VII Fast-neutron-reaction Production Rates in EBR-II

Nuclide	Fraction in 304 SS (w/o)	Hydrogen Production Rate				Helium Production Rates			
		$\int \sigma_j \phi_j$ (a) (sec ⁻¹)	$\int n_j \sigma_j \phi_j$ (atoms/sec)	Atoms MW(t)-D in EBR-II	Production in 304L SS (%)	$\int \sigma_j \phi_j$ (a) (sec ⁻¹)	$\int n_j \sigma_j \phi_j$ (atoms/sec)	Atoms MW(t)-D in EBR-II	Production in 304L SS (%)
⁵⁴ Fe	4.01	7.04×10^{-11}	3.24×10^{15}	4.5×10^{18}	28.2	8.92×10^{-13}	4.10×10^{13}	5.7×10^{16}	5.0
⁵⁶ Fe	63.2	8.55×10^{-13}	6.06×10^{14}	8.4×10^{17}	5.2	1.15×10^{-13}	8.16×10^{13}	1.1×10^{17}	9.6
⁵⁷ Fe	1.51	(b)	-	-	-	(b)	-	-	-
⁵⁸ Ni	6.06	1.13×10^{-10}	7.35×10^{15}	1.0×10^{19}	62.5	$3.41 \times 10^{-13(e)}$	2.22×10^{13}	3.1×10^{16}	2.7
⁶⁰ Ni	2.361	2.00×10^{-12}	5.0×10^{13}	6.9×10^{16}	0.4	1.79×10^{-13}	4.47×10^{12}	6.2×10^{15}	0.5
⁵⁰ Cr	0.82	2.31×10^{-12}	2.31×10^{13}	3.2×10^{16}	0.2	1.27×10^{-12}	1.27×10^{13}	1.8×10^{16}	1.6
⁵² Cr	16.11	3.20×10^{-13}	6.08×10^{13}	8.5×10^{16}	0.5	1.76×10^{-12}	3.34×10^{14}	4.6×10^{17}	40.0
⁵³ Cr	1.82	(b)	-	-	-	(b)	-	-	-
²⁸ Si	0.92	8.03×10^{-12}	1.61×10^{14}	2.2×10^{17}	1.4	3.33×10^{-12}	6.66×10^{13}	9.4×10^{16}	8.1
²⁹ Si	0.047	(b)	-	-	-	(b)	-	-	-
³¹ P	0.040	(b)	-	-	-	(b)	-	-	-
³² S	0.029	5.79×10^{-11}	3.18×10^{13}	4.4×10^{16}	0.3	7.15×10^{-11}	3.93×10^{13}	5.5×10^{16}	4.8
⁵⁵ Mn	2.00	(b)	-	-	-	(b)	-	-	-
¹² C	0.03	(b)	-	-	-	4×10^{-13}	6.27×10^{11}	8.77×10^{14}	0.1
¹⁴ N	0.05	5.85×10^{-11}	1.32×10^{14}	1.84×10^{17}	1.2	1.02×10^{-10}	2.28×10^{14}	3.17×10^{17}	27.6
¹⁰ B	(F)(c)	(b)	-	-	-	3.46×10^{-10}	-	-	-
Total (for Type 304 Stainless Steel)			1.17×10^{16}	1.59×10^{19}			8.31×10^{14}	1.15×10^{18}	
²³ Na	$7.6 \times 10^{26(d)}$	1×10^{-12}	7.6×10^{14}	1.05×10^{18}		2.99×10^{-13}	2.27×10^{14}	3.16×10^{17}	

(a) Equivalent to the atom production rate per second per atom of isotope

(b) Inadequate data available

(c) F denotes an impurity whose composition varies but in general is less than 0.002%

(d) Atoms of ²³Na in EBR-II core(e) Value of 2.18×10^{-12} is equally probable

III. DISCUSSION

A. Tritium Yields

The principle motivation for estimating tritium yields results from the importance of tritium activities to reactor fuel-reprocessing schemes. The curies of tritium in discharged reactor fuel predicted from the above calculations are summarized below.

Reactor	Burnup [MW(t)-D/MT]	Tritium per Metric Ton (Ci)
LWR, ^{235}U fuel	20,000	327
LWR, ^{239}Pu fuel	20,000	540
LWR, 1/2 U-1/2 Pu fuel	20,000	440
LMFBR, ^{235}U core	100,000	1510
LMFBR, ^{239}Pu core	100,000	3150
LMFBR, ^{239}Pu - ^{238}U cycle core and blanket	40,000	1900

Detailed calculations of tritium generation from low-Z elements such as lithium or boron present as impurities in fuel, structural, cladding, and coolant materials have not been attempted because of the uncertainty in their concentration levels. Boron contributions are certainly negligible in thermal reactors and very small in fast reactors.

Lithium represents the principal source of tritium other than fission. The tritium generation from lithium may be estimated for EBR-II, where the lithium concentration in the sodium coolant is approximately 0.5 ppm. This corresponds to about 3.8×10^{20} atoms in the core. An upper limit to the production rate of tritium from $^6\text{Li}(n,t)$ and $^7\text{Li}(n,tn)$ would be about 1×10^{14} atoms per MW(t)-D. This rate could be increased significantly in a thermal reactor. The only significant source of tritium in LMFBR's may be concluded to be fission.

The uncertainty of the tritium problem was illustrated in a 1964 review of the management of tritium in fuel-processing wastes. It was stated that "the extent to which present practices can be applied to the future management of tritium-bearing wastes from recovery of power reactor fuels is unclear."⁷⁴ The Purex Solvent Extraction (aqueous) process presently employed to reprocess reactor fuel results in conversion of over 90% of the tritium in the fuel into water and about 7% of the tritium is exchanged with organics. Special considerations are required for safe disposal of tritium-bearing water.

Liquid dispersal of the tritium activity extracted from a ton of spent fuel requires dilution factors which probably require a river. In the 1964 review,⁷⁴ it was estimated that 1200 Ci/day of tritium could be safely dispersed to the atmosphere through a 100-m stack under very unfavorable atmospheric conditions without exceeding one-tenth the maximum permissible ground-level concentrations. The report considered the 96,000 MW(t) or about 35,000 MW(e) of total power generation would be associated with that amount of tritium and that this level of power generation would be encountered by about 1985. However, calculations made in this paper indicate the power generation associated with 1200 Ci/day of tritium would correspond to only 73,000 MW(t) for ²³⁵U fuel or about 55,000 MW(t) for a mixed Pu-U fuel. Furthermore, larger amounts of tritium will be encountered sooner than expected. The AEC has recently estimated that by 1980 the total nuclear electric power generation will be about 150,000 MW(e).⁷⁵ This 1980 power generation would represent, based on the above calculations, a daily production rate of 7,000 Ci/day for ²³⁵U fuel or as high as 9,000 Ci/day if mixed U-Pu fuels predominate at that time. An earlier projection by the Federal Power Commission⁷⁶ estimated an installed nuclear system capacity of 734,000 MW(e) by the year 2000 [their estimate for 1980 was approximately 100,000 MW(e)], which corresponds to a daily production rate in the year 2000 of about 45,000 Ci/day for mixed U-Pu fuels or an annual production rate in excess of 14 million curies of tritium.

Positive conclusions concerning tritium generation in power reactors is, perhaps, premature until more experimental verification of theoretical values is obtained. It is of interest to refer to a 1966 experimental study of the tritium content of irradiated power reactor fuels.⁷⁷ Applying a reference tritium yield of 0.83×10^{-4} , this study actually found 150% of the expected tritium in the dissolved fuel. The indicated yield of 1.2×10^{-4} agrees very well with our calculated value of 1.1×10^{-4} .

B. Fast-neutron Interactions

It has been reported by Weir³ that a helium atom concentration of 10^{-7} is sufficient to reduce the ductility of Type 304 stainless steel to one-half of its normal value. Assuming that all of the helium atoms produced within the stainless steel are present in grain boundaries, the 10^{-7} concentration would be reached in about 2 days in the maximum EBR-II flux. Since many of the helium atoms will be present in the form of smaller bubbles that may be bound to precipitates or dislocations, and others will be lost by migration and diffusion from the stainless steel, only a fraction of the total helium produced will be effective in reducing the ductility. It should be expected, however, that the stainless steel will begin to show reduced ductility in a fairly short time (considerably less than one year) of in-core residence at full power.

If induced helium generation within structural and cladding material represents a problem, these calculations provide a means of identifying significant sources. The principal sources of helium in stainless steel result from (n,α) reactions of ^{52}Cr , ^{14}N , ^{56}Fe , and ^{28}Si , in order of decreasing importance. Chromium isotopes are calculated to contribute in excess of 40%, nitrogen over 25%, and iron about 15%. Boron-10 as trace impurities is known to be a principal source of helium in thermal reactors. The spectrum-averaged cross section for the $^{10}\text{B}(n,\alpha)\text{T}$ reaction for EBR-II is given in Table VIII. It may be assumed that boron represents a negligible source of helium in fast reactors; for example, a 10-ppm concentration in stainless steel would produce less than 3% of the helium generation in Type 304L stainless steel irradiated in EBR-II. An alloy containing less than 4 w/o chromium, 0.1 w/o silicon, and 0.005 w/o nitrogen would reduce the helium sources by over a factor of two and, therefore, could be expected to maintain its mechanical properties over a considerably longer period of time in a fast reactor. Boron concentrations of less than one part per million would greatly affect the operating lifetime in a thermal reactor, but is of little concern for fast reactor conditions.

TABLE VIII. Sources of Gaseous Atoms in EBR-II

Source	$\frac{\text{T Atoms}}{\text{MW(t)-D}}$ (10^{17} atoms)	% T in EBR-II	$\frac{\text{H Atoms}}{\text{MW(t)-D}}$ (10^{17} atoms)	% H Atoms in EBR-II	$\frac{\text{He Atoms}}{\text{MW(t)-D}}$ (10^{17} atoms)	% He in EBR-II
Fission	3.5	100	4.3	2.5	55.0	78.4
Fe			53	30.5	1.7	2.3
Ni			100	57.7	0.4	0.6
Cr			1.2	0.7	4.8	6.9
Si			2.2	1.3	0.9	1.3
N			1.8	1.0	3.2	4.6
Na			10.5	6.1	3.2	4.6
Other	0.001 ^(a)		0.5	0.2	0.9 ^(b)	1.3
TOTAL	3.5		173.5		70.1	

(a) Estimated upper limit from lithium impurity in sodium.

(b) Includes 0.6 from 0.001% ^{10}B impurity and 0.3 from ^{32}S .

These calculations indicate that hydrogen is generated at a rate more than ten times greater than is helium for 304L stainless steel in the EBR-II flux. Over 60% of the hydrogen is calculated to result from fast-neutron interactions with ^{58}Ni . The formation of bubbles by hydrogen is much less likely than from helium. Some hydrogen could be held in solid solution in stainless steel at high temperatures; upon rapid cooling, bubble formation could result. Because hydrogen is so mobile at high temperatures, hydrogen in excess of the solubility limit would most likely evolve at the surfaces or interfaces rather than remaining in the matrix to form bubbles. Hydride formation in stainless steel is also unlikely. Hydride formation may be quite important for exothermic occluders of hydrogen, such as metallic fuels, zirconium, vanadium, and titanium, because hydrides contribute to embrittlement.

Based upon the amount of sodium in the EBR-II core, the calculations indicate that considerably more gaseous atoms are produced from both fission and from neutron reactions of stainless steel than are produced from sodium. No attempt has been made to consider neutron reactions with low-mass impurities in the sodium; however, these impurities could be more significant than the sodium itself.

Another ramification of gaseous atom production in fast reactors is the effect of fuel swelling. Assuming that approximately 3×10^{21} fission per MW(t)-D occur and that $1/3$ of all fissions lead to gaseous atoms, one can estimate 1×10^{21} gaseous atoms/MW(t)-D due to fission. This represents over 10 liters at STP of gas generation per day in EBR-II. A summary of all calculated sources of hydrogen and helium in EBR-II for a ^{235}U oxide fuel is presented in Table VIII. Nickel represents the largest source of low-mass gaseous atoms with fission and iron, each producing about one-half as much as nickel in EBR-II.

C. Conclusions

We have attempted to review the area of low-mass charged-particle production as it relates to reactor technology. The calculations have been included only for a frame of reference, and any conclusion inferred by these results must be made with regard to their accuracies. The fission-yield values have been based on experimental results of considerable uncertainty. Based on theoretical considerations, we have attempted to extrapolate experimental data to predict yields which have not been verified by experiment. Errors in the fission-yield data may well exceed 50%. The lack of accurate and consistent differential (n,p) and (n, α) cross-section data is the principal source of error in the stainless steel production-rate calculations. Disagreements by as much as an order of magnitude can be found in the literature for the $^{58}\text{Ni}(n,\alpha)^{55}\text{Fe}$ reaction, and errors in other (n,p) and (n, α) reactions may be as large. Uncertainties in the energy distribution of the EBR-II flux could result in errors as large as a factor of two for some of the calculations. In the event that more accurate light-element production rates are required, an extensive experimental effort will be necessary to obtain the required monoenergetic and integral cross-section data.

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