An Overview of Tritium Fast Fission Yields

J. E. Tanner

March 1981

**Prepared for
the Environmental Control Technology Division,
U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute**

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

The views, opinions and conclusions contained in this report are those of the contractor and do not necessarily represent those of the United States Government or the United States Department of Energy.

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
Under Contract DE-AC06-76RLO 1830

Printed in the United States of America
Available from
National Technical Information Service
United States Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151

Price: Printed Copy \$ _____ *: Microfiche \$3.00

*Pages	NTIS
	Selling Price
001-025	\$4.00
026-050	\$4.50
051-075	\$5.25
076-100	\$6.00
101-125	\$6.50
126-150	\$7.25
151-175	\$8.00
176-200	\$9.00
201-225	\$9.25
226-250	\$9.50
251-275	\$10.75
276-300	\$11.00

3 3679 00056 1961

AN OVERVIEW OF TRITIUM FAST
FISSION YIELDS

J. E. Tanner

March 1981

Prepared for
the Environmental Control Technology Division,
U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

SUMMARY AND CONCLUSIONS

Tritium production rates are very important to the development of fast reactors because tritium may be produced at a greater rate in fast reactors than in light water reactors. This potential increase in tritium from fast reactors is due to: 1) the possibly higher yields of tritium from ^{239}Pu fissions than from ^{235}U fissions, 2) the higher yields of tritium from neutron capture in boron, and 3) the possibly higher yields of tritium from fast fission than from thermal fission. It is presently believed that tritium produced by ternary fission in fast reactors contributes 25 to 50% to the total tritium production. If ongoing studies confirm an order-of-magnitude increase in fast fission yields for tritium, the total tritium production will be significantly increased.

This report focuses on tritium production and does not evaluate the transport and eventual release of the tritium in a fast reactor system. However, if an order-of-magnitude increase in fast fission yields for tritium is confirmed, fission will become the dominant production source of tritium in fast reactors. Assuming that the fraction of tritium released to the reactor coolant and eventually to the environment is unchanged from previous evaluations (ref. EISs, LMFBR, GCFR), there would be a corresponding increase in estimated tritium releases to the environment from LMFBRs and GCFRs. For a GCFR, the current estimated tritium release is three orders of magnitude less than that for an LWR, so the possible increase would pose no apparent problem. An order-of-magnitude increase in the amount of tritium released from an LMFBR is more significant and would make it roughly equivalent to tritium releases from an LWR.

Increases in tritium fast fission yields are still being investigated experimentally. As the results of these experiments become available, a more extensive evaluation of tritium production from all sources should be undertaken. At that time, it would also be appropriate to reexamine tritium transport and release from LMFBR systems to determine whether additional environmental control measures are required.

CONTENTS

SUMMARY AND CONCLUSIONS	iii
INTRODUCTION	1
TERNARY FISSION YIELDS FOR TRITIUM	3
TRITIUM PRODUCTION IN FAST REACTORS	5
LMFBR TRITIUM GENERATION	6
GCFR TRITIUM GENERATION	6
REFERENCES	9

INTRODUCTION

Tritium production in nuclear reactors has long been an item of concern, since tritium can have significant biological impacts if released to the environment. For this reason, it is important to be able to predict how much tritium a reactor produces, and what processes are important in its production. For one important process, nuclear fission, some uncertainty exists as to how much tritium the process yields in fast reactors.

Until recently, tritium yields for fast fissions were estimated by calculations using the yields from thermal fission. Now, experimental results show significantly greater yields than previously calculated for fast neutron fission of the major fuel isotopes. The increase in the production of tritium could be as much as an order of magnitude. Given the uncertainties in fast fission yields, estimates of tritium releases from fast reactors could also be uncertain.

The calculated tritium yield estimates and the experimentally observed values from recently gathered data are presented in this report. Also included is a comparison of source terms for tritium generation in various fast reactors.

TERNARY FISSION YIELDS FOR TRITIUM

Ternary fission is a nuclear process whereby fissioning nuclei split into three fission fragments, usually one low mass particle and two heavier particles. Isotopes of helium and isotopes of hydrogen are produced this way during ternary fission. The relative frequency of emission in low-energy ternary fission is ${}^4\text{He} > {}^3\text{H} > {}^6\text{He} > {}^1\text{H} > {}^2\text{H} > {}^3\text{He} > {}^8\text{He}$ (Dudey 1968). The yield of tritium in fission is a function of both the neutron energy and the type of fissioning nucleus. For example, the thermal neutron yield of tritium from ${}^{239}\text{Pu}$ exceeds that from ${}^{235}\text{U}$ by a factor of 2 (Table 1).

TABLE 1. Calculated Yields of Tritium from Thermal-Neutron Fission

<u>Isotopes</u>	<u>Calculated Yield^(a)</u> <u>(10^{-4} Tritons/Fission)</u>
${}^{233}\text{U}$	1.1
${}^{235}\text{U}$	1.3
${}^{238}\text{U}$	2.6
${}^{239}\text{Pu}$	2.3
${}^{241}\text{Pu}$	.26

(a) Based on data compiled by Argonne National Laboratory (1968).

Previous estimates of fast fission yields do not include the fertile isotopes such as ${}^{238}\text{U}$ and ${}^{232}\text{Th}$, which have fission threshold neutron energies of 1 MeV and 1.5 MeV, respectively. The tritium yields from these isotopes need to be known with greater accuracy to determine their impact on total tritium production. In a breeder reactor, the fertile isotopes may contribute as much as 20% of the fission fraction (Buzzelli, et al. 1976). Thus, the overall tritium production is sensitive to the fertile isotopes in a fast reactor.

An experiment performed by General Atomic personnel to establish empirical fast fission yield values for the heavy metal isotopes produced results far

different than expected. The samples (High-Temperature Gas-Cooled Reactor (HTGR) type fuel particles) were irradiated in the Experimental Breeder Reactor (EBR-II) at Idaho Falls until a burnup of 50 MWd/kg was achieved (Buzzelli 1976). The tritium content was then determined by liquid scintillation counting and corrections were applied for decay and volume dilution. The precision of the measurements is generally within $\pm 4\%$. Other experiments are being carried out, mainly at Oak Ridge National Laboratory (ORNL), to resolve uncertainties in the data or in the techniques used to observe the amount of tritium and whether there are any other source terms not taken into account. The results of this experiment are shown in Table 2 (Till, et al. 1980), which compares the calculated and experimental yields.

TABLE 2. Tritium Yields from Fast Neutron Fission

Isotope	10^{-4} Tritons/Fission	
	Buzzelli, et al. ^(a)	ENDF/BIV Library ^(b)
^{232}Th	6	2.0
^{233}U	15	1.4
^{235}U	1.5	1.1
^{238}U	9	2.3
^{239}Pu	20	1.8

(a) Based on irradiations in the fast flux of EBR-II (Buzzelli et al., 1976; Buzzelli and Langer, 1977).

(b) ENDG/BIV library-tapes 401-411 and 414-419, National Neutron Cross Section Center, Brookhaven National Laboratory (December 1974).

TRITIUM PRODUCTION IN FAST REACTORS

The preceding section concentrated attention only on the ternary fission yields in fast reactors. Some of these reactions, such as fission of the fertile isotopes, have greater cross sections at higher energies and may not play a major role in thermal reactors but need more consideration in fast reactors. In order to place these quantities in perspective, this section discusses the other sources in comparison with ternary fast fission yields.

Tritium is produced in several ways in fast reactors. Some of the more important sources are neutron activation of lithium and boron, in addition to ternary fission. Boron and lithium can contribute anywhere from 35 to 80% of the total tritium produced, depending on the type of reactor. Boron is used in the control rods in fast reactors and is also used as a chemical shim in the reactor coolant in fast reactors and some light water reactors. Lithium occurs as an impurity in the coolant and fuel.

The lithium reactions for tritium production are:

The boron activation reactions are the following:

The ${}^{10}\text{B}(\text{n},\alpha){}^7\text{Li}$ reaction is the predominant reaction at fast energies and the reason for having the boron in the reactor. Most of the tritium, however, is produced by the ${}^{10}\text{B}(\text{n},2\alpha){}^3\text{H}$ reaction because the ${}^{11}\text{B}(\text{n},{}^9\text{Be}){}^3\text{H}$ reaction and the ${}^7\text{Li}(\text{n},\alpha){}^3\text{H}$ reaction have threshold energies of 9.6 MeV and 2.8 MeV, respectively. The amount of lithium-6 is very small relative to the amount of lithium-7 involved. The average value for the cross section of the ${}^{10}\text{B}(\text{n},2\alpha){}^3\text{H}$ reaction over the ${}^{235}\text{U}$ fission spectrum is about 31.85 millibarns (Sehgal 1971).

Two brief examples of tritium production in fast reactors are given in the following subsections, using the Liquid-Metal-Cooled Fast Breeder Reactor and the Gas-Cooled Fast Reactor concepts.

LMFBR TRITIUM GENERATION

The Liquid-Metal-Cooled Fast Breeder Reactor (LMFBR) design utilizes the fast neutron spectrum to generate fissions. It uses mixed oxide (Pu-U) fuel clad in stainless steel rods that are cooled by liquid sodium. The tritium in an LMFBR is produced by ternary fission in the fuel, activation of the boron in the control rods, and activation of the lithium present as an impurity in the fuel and the coolant. Kabele (1974) estimated that in the Fast Flux Test Facility (FFTF), the tritium produced from lithium activation would be about 50% of that produced from ternary fission and the tritium generated, due to the B_4C content of the control rods, would be 7.4 times that resulting from ternary fission.

In an LMFBR, it is assumed that 2.7×10^{21} fissions produce 1 MWD of thermal power (i.e., 200 MeV/fission) and that a 1000-MW(e) LMFBR is rated at 2500 MW(t) (Trevorrow, et al. 1974). Since the fuel is mixed oxide, a high fraction of the fissions will come from ^{239}Pu . Using a lower limit of 1.5×10^{-4} ^3H atoms/fission for fast fissioning of plutonium and an upper limit of 2.3 to 2.4 times that, the total generation rate of tritium by fission is estimated to be 2×10^4 to 4×10^4 curies/1000 MW(e)-year (Trevorrow, et al. 1974). This will be even higher if it is concluded by experimental measurement that fast fission yields of tritium for ^{239}Pu and the fertile isotopes of uranium are greater than previously calculated. Table 3 shows the breakdown calculated for tritium generation in an LMFBR.

GCFR TRITIUM GENERATION

The Gas-Cooled Fast Reactor (GCFR) concept is a combination of the High-Temperature Gas-Cooled Reactor (HTGR) helium coolant technology and the fuel and physics aspects of the Liquid-Metal Fast Breeder Reactor. The fuel would be mixed oxide and the fuel cladding would be type 316 stainless steel as in

TABLE 3. Estimated Rate of Tritium Generation in an LMFBR

Source	Expected Rate of Generation ^(a) Ci/1000 MWe-yr
Fission	2 to 4 x 10 ⁴
Activation of B ₄ C in Control Rods	65,000
Activation of B Impurity in Primary Coolant	700
Activation of B Impurity in Core and Blanket Fuel	600
Activation of Lithium in Primary Coolant	85
Activation of Lithium in Core and Blanket Fuel	1,400
	9 to 11 x 10 ⁴

(a) Trevorrow, et al., 1974.

an LMFBR. The unique feature of the GCFR fuel pin is its vented fuel rod with a pressure equalization system to eliminate pressure differentials between internal fission-product gas buildup and external gas coolant pressure. The surfaces of the fuel rods are also roughened to improve heat transfer capabilities.

In the GCFR, tritium would be produced by ternary fission in the fuel, boron activation in the control rods, and activation of the small amount of ³He in the helium coolant. Assuming a fission product yield of 1.4×10^{-4} ³H atoms/fission, the estimated tritium production rate for a 300-MW(e) demonstration plant (General Atomic 1974) is:

Ternary fission	5,460 Ci/yr.
Activation of ³ He	40 Ci/yr.
Activation of ¹⁰ B in control rods	<u>3,000 Ci/yr.</u>
Total	8,500 Ci/yr.

REFERENCES

- Argonne National Laboratory. 1968. Chemical Engineering Division Research Highlights, May 1967- April 1968. ANL-7450, Argonne, Illinois
- Buzzelli, G., et al. 1976. "Tritium: Fast Fission Yields of ^{238}U and ^{232}Th ." Trans. American Nuclear Society, 24:458.
- Buzzelli, G. and S. Langer. 1977. "Thorium Conversion and Tritium Fission Yields in an EBR-II Irradiation." Trans. American Nuclear Society. 27:283.
- Dudey, N. D. 1968. Review of Low-Mass Atom Production in Fast Reactors. ANL-7434, Argonne National Laboratory, Argonne, Illinois.
- General Atomic Company. 1974. Gas-Cooled Fast Breeder Reactor; Preliminary Environmental Report on 300-MWe Demonstration Plant, Vol. 1. GA-A12957, General Atomic Project 1304, San Diego, California.
- Horrocks, D. L. 1973. "Yields of Tritium During the Fissioning of Nuclides of Interest in the Operation of Nuclear Power Reactors." Tritium. Hoghissi, A. A. and Carter, M. W. eds. Messenger Graphics, Phoenix, Arizona.
- Jacobs, D. G. 1968. Sources of Tritium and Its Behavior Upon Release to the Environment. U.S. Atomic Energy Commission/Division of Technical Information, Oak Ridge, Tennessee.
- Kabele, T. J. 1974. Parametric Study of the Tritium Distribution in the FFTF. HEDL-TME 74-6. Hanford Engineering Development Laboratory, Richland, Washington.
- Sehgal, B. R. and R. H. Rempert, 1971. "Tritium Production in Fast Reactors Containing B_4C ." Trans. American Nuclear Society. 14:779.
- Till, J. E., et al. 1980. Tritium - An Analysis of Key Environmental and Dosimetric Questions. ORNL/TM-6990, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Trevorrow, L. E., et al. 1974. Tritium and Noble-Gas Fission Products in the Nuclear Fuel Cycle, I. ANL-8102, Argonne National Laboratory, Argonne, Illinois.

DISTRIBUTION

No. of
Copies

No. of
Copies

OFFSITE

A. A. Churm
DOE Chicago Patent Group
9800 South Cass Avenue
Argonne, IL 60439

J. Counts
DOE Division of Environmental
Control Engineering
Washington, D.C. 20545

27 Technical Information Center

ONSITE

DOE Richland Operations

H. E. Ransom

52 Pacific Northwest Laboratory

R. L. Aaberg
M. H. Arndt
W. J. Bailey
W. J. Bair
A. Brandstetter
C. L. Brown
D. B. Cearlock
L. L. Clark
M. O. Cloninger

C. Cowan
G. W. Dawson
D. A. Dingee
W. I. Enderlin
R. M. Fleischman
J. C. Fox
A. J. Haverfield
C. M. Heeb
D. L. Hessel
J. H. Jarrett
W. E. Kennedy, Jr.
H. V. Larson
R. C. Liikala
R. W. McKee
B. A. Napier
D. F. Newman
J. M. Nielsen
A. M. Nolan
A. M. Platt
T. B. Powers
R. J. Serne
J. E. Tanner (10)
B. E. Vaughn
E. C. Watson
L. D. Williams
J. K. Young
Technical Information (5)
Publishing Coordination (2)
EI