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**FISSION PRODUCT INVENTORY
AND DECAY HEAT ASSOCIATED
WITH FTR FUEL**

FEBRUARY 1969

**AEC RESEARCH &
DEVELOPMENT REPORT**

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3 3679 00061 2327

BNWL-961
UC-80, Reactor
Technology

FISSION PRODUCT INVENTORY
AND DECAY HEAT ASSOCIATED
WITH FTR FUEL

By

W. L. Bunch and L. D. O'Dell
Reactor Physics and Operation Department
FFTF Project

February 1969

FIRST UNRESTRICTED
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PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON 99352

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

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ABSTRACT

The fission product inventory and the decay heat associated with driver fuel irradiated to goal exposure (45,000 MWd per metric ton) in the Fast Test Reactor is presented, based on calculations using the computer code RIBD with a nuclear data library prepared for the FTR environment. Curie inventories as a function of decay time are given for each of about 350 isotopes or isomeric states generated by the fast-neutron induced fission of either ^{239}Pu or ^{238}U , by down-chain decay, or by subsequent neutron capture. Beta, gamma, and total decay power are given in percent of operating power for decay times from 1 sec to about 10 years. Uncertainty in the decay heat calculations, based on propagation of the uncertainties associated with input nuclear data, is estimated. The uncertainty is calculated to be less than $\pm 10\%$ for the first 10 days, and less than $\pm 20\%$ over a 10-yr decay period.

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INTRODUCTION

It is necessary to establish the fission product inventory and the decay heat associated with irradiated driver fuel from the Fast Test Reactor (FTR) in order to provide a realistic and consistent basis for the design of a number of components and systems throughout the Fast Flux Test Facility (FFTF). The fission product inventory is required for evaluating the radiological consequences associated with the accidental release of the material. The fission product inventory in turn provides the required information for estimating shielding requirements for specific arrays of material, and for calculating the decay heat associated with this material. Decay heat rates are required information for the design of emergency or backup cooling systems for the reactor, for establishing shutdown cooling requirements and design bases for the fuel handling equipment, and for storage, examination, and shipping facilities. The objective of this report is to establish a reference position for the FFTF project in order that the design of all systems might adequately, realistically, and consistently accommodate the decay of fission products within the irradiated fuel.

Most of the available information concerning fission products was obtained from the study of those isotopes generated in the thermal-neutron induced fission of ^{235}U , whereas the FTR fuel will contain fission products produced primarily by the fast-neutron induced fission of ^{239}Pu . Because of the large number of radioisotopes produced by fission, it has been expedient in the past to treat the

fission products by statistical methods and not to differentiate between the fissioning nuclide. However, it is recognized that the most rigorous treatment can be achieved by considering individual isotopes. With existing computer programs, and with the large amount of nuclear data that exists, it is possible to calculate fission product inventories for specific fuels, reactor systems, and exposure conditions. This rigorous approach was used in the present study to calculate the fission product inventory and the decay heat associated with driver fuel irradiated to goal exposure in the FTR. An estimate was also made of the uncertainty in the decay heat calculations. This estimate was based on propagation of the experimental errors associated with the basic nuclear data employed in the calculations.

SUMMARY

It is assumed that driver fuel in the FTR will be irradiated to goal exposure during four 80-day long operating cycles interspersed with three 28-day outages. For a 400 MW system, one-fourth of the fuel would generate 100 MW. The curves in the Appendix present the curie content of FTR fuel that has generated 100 MW of power through four operating cycles to achieve an exposure of about 45,000 MWd per metric ton. These results can be scaled directly to estimate inventories in individual fuel sub-assemblies or pins. Inventories calculated using the computer program RIBD include the direct formation of each isotope or isomeric state by fission of both ^{239}Pu and ^{238}U , by chain decay of precursors, and by neutron induced transmutations.

Based on the calculated fission product inventories and the decay energy associated with each isotope, decay heat is obtained as a function of time with the results normalized to

percent of operating power. Thus, the results can be applied to any quantity of material. For the postulated operating history, the results should be valid within the uncertainty introduced by the nuclear data. It is estimated that the uncertainty in the calculated decay heat rate minimizes at about $\pm 5\%$ after a decay time of about an hour. The maximum calculated uncertainty at shorter decay times is about $\pm 6\%$, whereas the estimated uncertainty at long decay times increases to almost $\pm 20\%$ after 10 years. Decay heat rates as a function of time are summarized in Figure 1.

To calculate the inventories and decay heat rates summarized in this report, it was necessary to generate a nuclear data library for use with the code RIBD. This library can be used to estimate inventories and decay heat rates associated with various fuels for any postulated operating history in the FTR.

BACKGROUND

Several hundred different isotopes or isomeric states are formed in reactor fuel as a result of the fission process. The inventory of fission products in a given fuel depends on the type of fuel, the reactor system, and the irradiation history. Because of the large number of fission products formed, it has been convenient in many cases to treat the gross decay characteristics using statistical techniques, and to neglect differences between fuels and reactor systems. With existing computers and available nuclear data, it is now possible to calculate fission product inventories for any type of fuel in any type of reactor following any given irradiation history. The uncertainty in the calculated results is dependent on the nuclear data, together with the rigor of the mathematical model employed. For this reason, considerable effort has been expended to generate an up-to-date nuclear data library for use in the present study.

FIGURE 1. Decay Heat Associated with Fission Products at Goal Exposure

A number of studies has been made to determine the decay characteristics of the fission products. One of the early reported studies is that of Borst⁽¹⁾ who measured the beta and gamma radiation from nongaseous fission products produced by the thermal neutron induced fission of ^{235}U . Measurements were made with electroscopes and Gieger-Mueller counters over the decay interval from 10 min to 220 days. Gold monitors irradiated with the uranyl nitrate samples were used to calculate the total number of fissions taking place. The data were presented in three parameter curves depicting activity per fission, time after irradiation, and the quantity of lead absorber over the sample. J. A. Wheeler⁽²⁾ has used these experimental results to calculate the energy liberation as a function of time of irradiation and cooling time. Results are directly applicable only for ^{235}U fission in a thermal neutron environment.

A direct calorimetric measurement of the fission product decay energy was made by Day and Cannon.⁽³⁾ Uranium was irradiated in the Clinton Reactor for periods from one to 122 days, and decay measurements were made with a calorimeter from 30 min to 60 days after discharge from the reactor. Decay curves were then plotted for each of the irradiations. Because results were not corrected for gamma radiation escaping from the calorimeter, the measured values are expected to be low. When compared to studies by others, the Day and Cannon results were higher at short decay times and lower at long decay times. As in the Borst experiment, the results are applicable only for ^{235}U fission induced by thermal neutrons.

Decay of the activity associated with fission products formed by the fission of ^{239}Pu was measured by Levochkin and Sokolov⁽⁴⁾ and compared to ^{235}U data. Enriched uranyl

nitrate and plutonium nitrate sources were prepared and irradiated in pairs in a neutron flux of about 10^{12} n/cm²-sec for 40 sec. Activity of the samples was measured over time periods ranging from about 2 hours to 1 year by means of a comparative method with standardized geometry and an end-window counter. These results indicated very little difference in the activity of the uranium and plutonium fission products in the 2-hr to 200-day time period.

A calorimetric determination of fission product heating associated with plutonium fuel irradiated in a fast reactor was made by Johnston.⁽⁵⁾ In these experiments, 40 mg samples of plutonium oxide were irradiated in the Dounreay Fast Reactor to an integrated exposure corresponding to 2.67×10^{17} fissions. The irradiation required several reactor operating cycles to complete. Decay heat was measured in a Calvet micro-calorimeter during the time period from about 40 to 125 days following discharge from the reactor. Corrections were applied for blank capsules, and uranium shields were used to separate the beta and gamma components. Consideration was also given to decay heat generated by the plutonium isotopes. These results were shown to compare favorably with ²³⁵U results obtained from thermal reactor systems.

A number of similar experiments comparing favorably with those described has been made and reported by others. Each experiment was limited in direct application to the specific conditions employed. For practical application, it was necessary to extrapolate the results to specific conditions of interest. One of the earliest attempts to do this was the classic work of Way and Wigner.⁽⁶⁾ They treated the fission products as a statistical assembly to derive relations to describe the decay rate. To derive their relations, they made a number of assumptions: a nucleus of mass number A and charge Z has a mass given by the relation $a Z_0(A) - Z^2 + b$, where

a and b are empirical constants; an empirical relation was used to relate the half life to the disintegration energy; the decay of fission products do not deviate significantly from the most probable modes of decay; and the chain lengths of the light and heavy fission fragments are equal. Their calculated values were shown to compare favorably with available experimental information.

Recognizing the limitations of the statistical treatment, Perkins and King⁽⁷⁾ calculated the decay of fission products using the known properties of 123 isotopes produced in the fission process. The isotopes were chosen on the basis of half life and cumulative yield and include all those with half lives greater than 1 min and with yield greater than 0.05%. The total chain yields were based primarily on the compilation of Katcoff,⁽⁸⁾ whereas independent fission yields were obtained from the work of Pappas.⁽⁹⁾ Calculations included the total disintegration rates, rates of beta and gamma energy release, and gamma-ray energy spectrum for the fission products produced by thermal-neutron induced fissioning of ^{235}U . The decay formulas used in the calculations took into account only one generation of decay. The results, while comparing favorably with other experimental and analytical results, fell somewhat below that reported by Way and Wigner.

Decay rates for ^{239}Pu fission products formed in a fast reactor environment were calculated by Burris and Dillon.⁽¹⁰⁾ They used the fast fission yield values compiled by Blomeke.⁽¹¹⁾ The work of Glendenin, Coryell, and Edwards⁽¹²⁾ was used as a basis for the distribution of the nuclear charge in a given mass chain. The calculations assumed 100% yield for the first isotope in a chain with half life greater than 1 hr, and assumed fissions occurring at a uniform rate throughout the irradiation period. In addition, all isotopes with half life less than 1 hr were neglected, as was the effect of neutron

absorptions in the fission products. These results would not be applicable at short decay times, nor for high flux reactors and high exposure material.

A critical review of available information on the energy release from ^{235}U fission products was made by Shure.⁽¹³⁾ He concludes that the energy release rates calculated by Stehn and Clancy⁽¹⁴⁾ should be used for cooling times less than 10^3 sec, whereas the results of Perkins and King⁽⁷⁾ should be used for longer times. Stehn and Clancy fused together relative observed time variations of gamma and beta decay with absolute determinations to provide absorbable decay power for times greater than 0.1 sec following a fission burst and following an infinite irradiation. Their results are normalized to an initial decay power of 7%. Beta-gamma activities provide the basis for shaping the decay heat curves out to about one day, at which time calculated summation values are employed.

A very recent calculation of the inventory and decay energy associated with the fast fission of ^{239}Pu is reported by Battat, Dudziak, and Hicks.⁽¹⁵⁾ These calculations were made using the computer code FPIC,⁽¹⁶⁾ which incorporates data for 200 fission product isotopes. Isotopes used in the calculation are those with half lives greater than 10 sec and ^{235}U fission yields greater than 0.001%. The omission of isotopes with half lives less than 10 sec would limit the application of the results. In addition, the FPIC calculations do not account for the production of shielded isotopes by neutron capture. The calculated results for ^{239}Pu are shown to agree satisfactorily with the Johnston⁽⁵⁾ experiments, which were limited to low exposure fuel and long decay times.

Based on the present review, it is concluded that decay heat can best be calculated by considering production and removal of all of the individual fission products in as

rigorous a manner as possible and then summing the results. Results obtained in this manner should be compared to experimental data to assure the generation of reliable values. However, there is no incentive to compare the results to previous, less rigorous analytical studies known to contain serious limitations. The work of Way and Wigner, among others, forms an important historical foundation for decay heat analyses, but does not provide a standard for evaluating subsequent, more rigorous analyses. The approach in the present study has been to assemble the most recent nuclear data available for the known fission products, and to employ these in realistically rigorous calculations to determine the decay heat associated with the Fast Test Reactor fuel.

COMPUTER PROGRAM RIBD

A number of computer codes have been written to calculate formation and decay of radioactive decay chains and time dependent concentrations of fission products. Because the processes can become quite involved, certain simplifying assumptions are usually made for the specific application of interest. One code developed specifically for calculating the inventory of fission products in a reactor is RIBD.⁽¹⁷⁾ This code was used in the present study because of its direct applicability to the problem, because it includes all important reactions strongly influencing the inventory, and because it was available and operational.

The RIBD code is a grid processor that calculates the isotopic concentrations of 450 isotopes or isomeric states resulting from two different fission sources, where the relative contribution of the two sources as a function of exposure is under input control. Normal down-chain decay by beta emission and isomeric transfers is included together with inter-chain coupling resulting from neutron capture. The calculations can follow the irradiation history through

an unlimited number of step power changes of unrestricted duration and variability including shutdown intervals, restarts at different power levels, and/or any other power level changes. The code organizes the various members described by the nuclear data library into a grid with the linkages established from chain branching data, yield data, and neutron capture cross sections. Radioactive decay includes not only the simple member-to-member cascade, but also the more complex forms where branching may partially or completely skip one or two intervening members. However, the code cannot treat inter-chain coupling resulting from delayed neutron emission nor by n,p and n,α reactions. This limitation is not considered to be significant.

Solutions of the differential equation for the buildup of a nuclide are programmed in a systematic manner providing the required quantities for the specific isotope under consideration. The solution is started at the lowest mass chain and each chain is processed in order. Computation within each chain proceeds from the lowest atomic number to the highest. Numerous checks are employed to assure that results converge within established limits.

Accuracy of the calculated results is primarily dependent on the nuclear data that are employed. Nuclear data required for the calculation include direct yield, half life, decay characteristics, and capture cross sections for each of the 450 isotopes or isomeric states. The nuclear data library incorporated in RIBD was developed for thermal reactors fueled with uranium. Before the program could be used for fast reactor studies, it was necessary to generate a new nuclear data library. The fast reactor library, as described in the following section, incorporates the latest available nuclear data supplemented, when necessary, by theoretical values.

NUCLEAR DATA

Considering the problems involved, a tremendous amount of nuclear data has been accumulated in the relatively few years since the onset of the nuclear age. In the case of some of the fission products, the limited information available is understandable because of short half lives and the necessity for processing the multiplicity of radioisotopes before individual properties can be associated with a specific isotope.

As indicated previously, four specific types of nuclear data are required for use in RIBD to calculate the fission product inventory and the decay heat associated with this inventory. Of primary importance is the production probability or direct yield of an individual isotope or isomeric state from a fission event in a given fissionable material. Once formed, the isotope or isomeric state can either decay to become a different element, or can capture a neutron and move to another mass chain. The relative probability of these events depends on the relationship between the half life or decay constant and the capture cross section as weighted by the neutron flux level. For decay events, it is further necessary to know decay characteristics, such as branching ratios and the beta and gamma energy associated with the event. The following paragraphs indicate the sources of information used to compile the nuclear data library for use in fast reactor studies.

YIELD

The direct or independent yield of each isotope or isomeric state is required data for the calculation of the fission product inventory. In most cases the only available experimental determinations are the cumulative yield of the stable or long-lived states at or near the end of some of the decay chains. A two-step approximation is required to

obtain the direct yield values. First, the complete mass-yield curve must be generated from the available data, and second, the charge distribution within each mass chain must be deduced.

Cumulative yield data as a function of mass have been reviewed critically in a number of studies for the slow neutron induced fission of ^{235}U , with the most widely referenced work probably being that of Katcoff.⁽⁸⁾ However, more recent reviews are thought to contain a number of refined values.⁽¹⁸⁾ A number of isotopes other than ^{235}U have also been investigated, though in most cases, in much lesser detail. Of primary interest for the present study is the compilation by Zysin⁽¹⁹⁾ for the fast fission of ^{239}Pu .

The yield curves for all heavy isotopes exhibit the same general shape as that shown in Figure 2 for ^{239}Pu . Two general conclusions can be drawn from the available experimental data. First, a principal effect of the increase in mass of the fissioning nucleus is a shift in the light mass peak to higher values, with the heavy mass peak remaining relatively fixed. Second, the depth of the trough between the two peaks decreases with the energy of the neutron inducing the fission event. These differences are of sufficient magnitude to indicate that fission product information developed for uranium-fueled thermal reactors is not necessarily applicable for plutonium-fueled fast reactors. Based on these two general observations, a purely empirical library⁽²⁰⁾ was generated for use with RIBD.

Complete mass-yield curves, such as those shown in Figure 2, are constructed for fast-neutron induced fission of ^{239}Pu as well as for a number of other isotopes by interpolation of available data. In many cases, the direct experimental data have been augmented by mirroring techniques. The effect of

FIGURE 2. Fission Product Mass Chain Yields for ^{239}Pu Fast Fission
(From Reference 23)

neutron energy is included to the extent that experimental data are available. The curve is normalized in each case to a total yield of 200%.

Experimental yield data for individual mass chains are generally valid within an uncertainty of about $\pm 10\%$, based on the techniques employed and the spread in the reported results. For high yield mass chains, the interpolated values might also be estimated within this approximate uncertainty. For low yield mass chains, such as in the trough between the peaks of Figure 2, the interpolated values are much less certain. In addition, local perturbations in the yield-mass curve expected as a result of delayed neutron emission introduce small uncertainties in interpolated values. In estimating uncertainty in decay heat calculations, the total mass-yield values are assumed valid within an uncertainty of $\pm 20\%$.

As indicated, the mass-yield curves alone provide inadequate data for use in calculating the inventory of fission products within an operating reactor. For these calculations, the direct fission yield distribution, or the charge distribution, within each chain must be known. Because the experimental measurement of this distribution is very difficult, little data of this type exist. Empirical relations developed⁽¹²⁾ by means of an equal charge displacement hypothesis have satisfactorily described the division of charge in the available data obtained from slow-neutron induced fission events in ^{235}U . This empirical approach has been used widely because it correlates and predicts data more successfully than any theoretical models to date. Refinements such as those described by Wolfsberg⁽²¹⁾ provide adjustments to accommodate fission events in isotopes other than ^{235}U , and by neutrons at energies other than thermal.

Based on the total chain yields compiled by Katcoff⁽⁸⁾ and on the charge recipe developed by Glendenin et al,⁽¹²⁾ independent yields were calculated and tabulated⁽²²⁾ for a large number of fission processes. Based on the mass-chain yield data reviewed by Zysin⁽¹⁹⁾ and the charge distribution relations developed by Wolsberg,⁽²¹⁾ direct yield values were calculated for the fast-neutron induced fission of ^{239}Pu by Anderson.⁽²³⁾ The yield data presented in References 22 and 23 have been used to generate a number of nuclear data libraries for use with the computer program RIBD. For the present fast reactor studies, the ^{239}Pu yields developed by Anderson⁽²³⁾ and the ^{238}U yields of Weaver et al,⁽²²⁾ were employed. Since most of the fission events are calculated to take place in ^{239}Pu , the results of the RIBD calculations are most strongly dependent on the yield values developed by Anderson. These yield values were selected for use because they are based on the latest available data and incorporate more detailed refinements than do other compilations.

The uncertainty in the direct yield factors is based on estimates calculated by Wolsberg for the empirical relation that he employed.⁽²¹⁾ The calculated uncertainties are related to the magnitude of the fractional yield in such a manner that, if the direct yield is large, the associated uncertainty is small and, if the yield is small, the associated uncertainty is relatively large. Calculated uncertainties in the direct yield factors were used in the present study to estimate the uncertainty associated with decay heat calculations.

HALF LIFE

The half life of each of 450 isotopes or isomeric states is required for the RIBD calculation of the fission product inventory. Half life values were obtained from both the

Nuclear Data Sheets⁽²⁴⁾ and the Table of Isotopes.⁽²⁵⁾ Both of these sources list reported experimental values together with a recommended value for each isotope. In general, the agreement between the recommended values is quite good. Based on the information available from these two sources, half lives greater than 1 hr are concluded to be known within about $\pm 3\%$, whereas shorter values are known only to about $\pm 6\%$. These uncertainties have been used to estimate the uncertainty in the present decay heat calculations.

The shortest reported half life associated with fission products is of the order of 1 sec. The problems associated with isolating, measuring, and identifying the short half-lived isotopes that lie at the beginning of a given mass chain is obvious. Because a relatively large number of these isotopes generated in the fission process quite likely have not been identified, they cannot be included in the nuclear data library or in the RIBD calculations. At decay times of a few seconds or less, calculated values probably will underestimate the fission product inventory and the decay heat rate because of this necessary omission.

DECAY CHARACTERISTICS

In addition to interchain coupling resulting from neutron capture, the computer program RIBD also treats the normal down-chain decay of radioactive isotopes by beta emission. The code cannot account for decay of one isomeric state to another isomeric state, nor decay by neutron emission into a lower mass chain. These limitations are not expected to be significant because of the rarity of these events. For example, delayed neutrons emitted in about 0.6% of the fission events induced in ^{239}Pu by fast neutrons results in disruption of about 0.3% of the chains. Most of these events are thought to take place in the isotopes $^{87-90}\text{Br}$ and $^{137-139}\text{I}$. Hence,

many of the events are compensated, at least in part, by incoming events from an adjacent chain. Neglect of these transitions is not expected to bias the results significantly.

Input decay data (in addition to half life) required for the RIBD calculation includes the branching ratio, together with the average beta and gamma energy per disintegration. The branching ratios were obtained from the Nuclear Data Sheets⁽²⁴⁾ and the Table of Isotopes.⁽²⁵⁾ In most cases, these branching ratios have been measured with a high degree of accuracy. In a limited number of cases, it was necessary to estimate the branching ratio from incomplete or limited data. In three cases, the branching ratio was arbitrarily adjusted to compensate for the inability of the code to treat decay from one isomeric state to another. These adjustments insure return of the decay product to the most probable path following an event that cannot be treated rigorously.

The average beta and gamma energies per disintegration were taken from three different sources.^(24,25,26) An examination of these three compilations indicates that the beta and gamma decay energies are known for only about three-fourths of the 450 isotopes and isomeric states included in the RIBD library. For the remaining isotopes, the information varies from none to a listing of beta and gamma energies with no indication of fractional release rates. The available data were used to the extent possible to generate the required decay energies.

The minimum available information upon which to base decay energy is the energy level between the isotope and its daughter, even though these levels might be generated theoretically. In these cases, it was assumed that half of the energy would be given up to neutrinos, with the other half split equally between beta and gamma radiation. This classic split has long been applied and is realistic for measured isotopes.

CAPTURE CROSS SECTIONS

Neutron-induced transmutations can disrupt normal down-chain decay and thereby influence the inventory of fission products in reactor fuel. The probability of neutron capture is given by the product $\sigma_a \phi$, where ϕ is the total neutron flux intensity and σ_a is the spectrum weighted capture cross section. The probability for decay is given by the decay constant λ , which is equal to 0.693 divided by the half life in seconds. The effect of capture becomes significant in high flux reactors where the product $\phi \sigma_a$ for a number of isotopes is comparable to, or larger than, the corresponding λ for those isotopes. The process is also quite important in transmuting stable isotopes for which λ is zero and no decay can take place. In addition, neutron capture is the only process whereby certain shielded radioisotopes can be formed. A shielded isotope is one that cannot be formed by beta decay because the isobaric nuclide of the next lower atomic number is stable. For these reasons, neutron-induced transmutations can significantly influence the fission product inventory in high exposure fuel irradiated in high flux reactors.

Neutron capture cross sections as a function of energy have been measured for a number of stable isotopes. The experimental cross section data can be appropriately weighted for the calculated neutron spectrum within a given reactor. Because experimental capture cross sections have not been measured for most of the radioisotopes resulting from the fission process, it was necessary to develop theoretical energy-dependent cross sections using an optical potential model. Appropriate spectrum-weighted capture cross sections have been calculated for all fission product isotopes of interest by Schenter.⁽²⁷⁾

The uncertainty associated with capture cross sections of stable isotopes based on experimental data is of secondary importance because of the overriding uncertainty in the yield

data. The large but unknown uncertainty associated with the theoretical capture cross sections is also unimportant in most cases because of the individual relationship between capture and decay probabilities. For example, if the decay probability is large (half life short), capture is improbable and the uncertainty in the capture cross section will not be reflected in the results. The capture cross sections of the stable or long-lived isotopes that transmute to shielded isotopes are of primary importance, since these isotopes would not otherwise be produced in the fuel. The uncertainty in capture cross sections has not been included in the estimated uncertainty in the calculated decay heat. However, this omission is not considered to be significant.

The previously discussed data are assembled in a nuclear data library⁽²⁸⁾ for use with RIBD.

RESULTS

REFERENCE ASSUMPTIONS

In order to estimate the fission product inventory and the decay heat associated with fast reactor fuel irradiated to goal exposure, the fuel and the operating conditions must be defined. In RIBD calculations, eight basic input parameters can be controlled to describe the environment, fuel, and operating history. These include the length of time at power during each operating period, the length of each outage, the number of operating periods required to achieve goal exposure, the flux level, the fraction of fission events occurring in each fissionable isotope, the total energy release associated with each fission event in the particular reactor system, and the conversion ratio or number of grams of ^{239}U produced per MWd.

The Fast Test Reactor is being designed to provide irradiation space for fuels and materials in an environment typical of that in a liquid-metal-cooled, fast-breeder reactor. The flux within the test space should be as high as practical to accelerate the testing programs. To achieve these goals, the concept includes an oxide-fueled, sodium-cooled reactor of about a thousand liters operating at a power level of 400 MW. Design and safety considerations require the fuel within the core to be contained in about 80 individual subassemblies. Goal exposure for the fuel is set at about 45,000 MWd per metric ton. Reactivity and fuel management considerations require exposure to be achieved in four operating cycles, each of about 80 days duration. To achieve the goal operating efficiency of 75%, the outages would be about 28 days long.

The mixed oxide fuel will contain about four parts of depleted uranium oxide for each part plutonium oxide. This approximate ratio is established by safety considerations to provide an adequately large negative Doppler coefficient. With this fuel composition, and with the required structural and coolant materials, the neutron spectrum will be such that about 90% of the fission events will be induced in plutonium and 10% in uranium. Although the isotopic composition of the plutonium and uranium has not been established firmly, it is assumed that all of the fissions will be induced in ^{239}Pu and ^{238}U . This assumption is necessary because RIBD can only accommodate the yield distribution of two isotopes. It is unlikely that this assumption will influence the decay heat calculations significantly because of the statistical nature of the results. However, inventories of specific isotopes might be biased slightly by this simplifying assumption.

A direct relation between the power level and the fission rate is provided by the heat of fission (or total energy release per fission event). The energy release per fission

depends on the type of fuel and the reactor system. The kinetic energy of the fission products, the kinetic energy of the neutrons, and the beta and gamma decay energy are dependent primarily on the fuel. The binding energy released when excess neutrons are absorbed in nonfission events depends on the composition of the reactor system as a whole. Preliminary estimates for the FTR yield a value of about 215 MeV per fission event.⁽²⁹⁾ In uranium-fueled, thermal reactors, a value of about 200 MeV is more typical. Thus, the fission rate and fission product production rate will be about 7% lower in the FTR than in a uranium-fueled thermal reactor operating at the same power level.

In summary, the following input information has been used to calculate the fission product inventory in FTR fuel. A power level of 100 MW has been used. This is the value associated with the one-quarter of the fuel discharged at each outage. For decay heat applications, the results are given in percent of operating power and can be normalized to any quantity of material. Eighty-day operating periods at a flux level of 5×10^{15} n/cm²-sec were assumed, with 28-day outages. Four operating cycles were assumed to achieve goal exposure. The heat of fission was assumed to be 215 MeV, with 90% of the fissions being induced in ²³⁹Pu and 10% in ²³⁸U. To account for the decay heat produced by heavy isotopes, it was assumed that 0.5 g of ²³⁹U was produced per MWd of operation. Since the decay of ²³⁹U and ²³⁹Np accounts for about 10 to 20% of the decay heat during the first few days, the conversion ratio must be considered in greater detail as the design becomes more firm.

The design reference conditions used in these calculations provide base conditions for the design of FTR systems. It is obvious that actual operating conditions will determine

the fission product inventory and the decay heat encountered in FTR fuel. Unless the assumed reference conditions are exceeded significantly, any system designed to accommodate the inventories and decay heat loads calculated herein should perform satisfactorily. In the event lower power levels, lower goal exposures, or lower operating efficiencies are actually achieved, the systems will be overdesigned for the encountered loads.

FISSION PRODUCT INVENTORY

The fission product inventory associated with fuel operating at a power level of 100 MW through four 80-day operating cycles (separated by three 28-day outages at zero power) is given in the Appendix by isotope for the 39 elements included in the RIBD calculation. These inventories should be of value in considering not only containment problems associated with the accidental release of this material but processing techniques as well. These inventories are also being used to calculate gamma intensity associated with irradiated fuel and the effect of shields in attenuating this radiation. These shielding calculations will be the subject of a separate report.

DECAY HEAT

The calculated beta, gamma, and total decay heat rates as a function of time are shown in Figure 1, where decay heat is in percent of operating power. These results can be applied directly to any quantity of material, such as that for the entire reactor, a single fuel subassembly, a single fuel pin, a unit volume of fuel, a unit mass of fuel, etc.

ESTIMATED UNCERTAINTY IN DECAY HEAT RATE

The estimated uncertainty in the decay heat rate calculated by the computer code RIBD is based on the uncertainty associated with the nuclear data and assumes the calculation

to be rigorous. A consideration of the limitations of the calculational technique employed in RIBD indicates this to be a reasonable approach. Although the decay heat calculations actually include the complex down-chain decay, it has been assumed that the uncertainty could be approximated by assuming decay heat to be given by the relation:

$$H(t) = \sum_i k_i Y_i E_i \exp(-\lambda_i t)$$

where Y is the yield, E is the decay energy, λ is the decay constant, and the summation is over the 450 isotopes or isomeric states. The normalizing constants k_i were obtained from the RIBD results at zero decay time:

$$H(0) = k C_i E_i = k_i Y_i E_i$$

where C_i is the curie content of an individual isotope or isomeric state.

For the assumed relationship, the standard deviation, σ_H , in the decay heat value is given by:

$$\sigma_H^2(t) = \sum_i^N \left\{ \left[Y_i E_i (-t) e^{-\lambda_i t} \sigma_{\lambda_i} \right]^2 + \left(E_i e^{-\lambda_i t} \sigma_{Y_i} \right)^2 + \left(Y_i e^{-\lambda_i t} \sigma_{E_i} \right)^2 \right\} k_i^2$$

where the subscripted σ 's are the standard deviations in the decay constant, yield, and average energy release per disintegration, respectively.

As indicated previously, the uncertainty in the yield was based on both the magnitude of the yield and the position of the isotope in the decay chain. If the particular isotope yield exceeded 10% of the total mass chain yield, or if the isotope was in the last half of the chain (so that cumulative yield exceeded 50% of the total), then a limiting uncertainty of $\pm 20\%$ was assumed. If the yield was between 1% and 10% and in the first half of the chain, then the deviation was approximated by

$$\pm\sigma_Y = 56 \exp (-0.105x)$$

where x is the percent of the total chain yield. The relation is based on the data tabulated in Reference 21. If the yield was less than 1% of the total for that mass chain and the isotope was in the first half of the chain, the deviation was approximated by

$$\pm\sigma_Y = 83.4 \exp (-0.595x) \quad .$$

where the numerics are chosen to fit the data in Reference 21.

The uncertainty in the average energy per disintegration was based on data obtained from References 24 and 25. Uncertainties were calculated as a function of the position of the isotope in the mass chain, the half life, and the half lives of the isotopes preceding it in the mass chain. The maximum deviation assigned to the decay energy for any isotope was $\pm 90\%$. Large values were associated with isotopes at the beginning of a mass chain that have short half lives. The minimum uncertainty associated with any of the isotopes was about $\pm 20\%$.

The uncertainty in the half life (decay constant) was based on the large amount of available tabulated experimental data. These tabulations indicate the uncertainty in the half life of less than about an hour to be significantly larger than for those cases where the half life is longer than an hour. Based on the tabulations, an uncertainty of $\pm 6\%$ was ascribed to all half lives of 1 hr or less and $\pm 3\%$ to all half lives greater than 1 hr.

The relations described above were programmed in order to calculate the effect of all of the isotopes as a function of time. Results of the uncertainty estimates are given in Table 1, where the percent deviation is the calculated uncertainty in the total decay heat rate at the given decay time. The uncertainty is large at first because of the uncertainties associated with short half life isotopes. A minimum uncertainty is estimated after about an hour decay time, where high yield isotopes with relatively well established characteristics are dominant. Because fewer isotopes remain at long decay times, the statistical averaging process becomes less important and uncertainties in individual isotopes become significant.

Although this uncertainty estimate does not rigorously propagate errors through the entire RIBD calculation, it should provide a realistic estimate of the uncertainty associated with the decay heat calculations because of the nuclear data limitations. It is concluded that the uncertainties associated with the nuclear data library employed for these calculations do not significantly limit the applicability of the results "for decay times greater than a few seconds."

TABLE 1. *Calculated Uncertainty Associated with Decay Heat Calculations Because of Uncertainties Associated with Nuclear Data*

<u>Time, sec</u>	<u>Deviation, %</u>
0.1	6.036
0.3	6.032
1.0	6.024
3.0	6.036
10.0	6.090
30.0	6.015
10^2	5.702
3×10^2	5.443
10^3	5.281
3×10^3	5.120
10^4	5.293
3×10^4	6.236
10^5	7.521
3×10^5	7.711
10^6	9.999
3×10^6	12.269
10^7	13.626
3×10^7	11.746
10^8	11.984
3×10^8	16.971
10^9	18.208
3×10^9	18.789

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APPENDIX

APPENDIX

CALCULATED FISSION PRODUCT INVENTORY IN FTR FUEL

The RIBD decay calculations were made at a finite number of times, spaced approximately exponentially, to provide inventory values out to about 10 years following shut down of the fuel. Figure A-1 contains the summed results for all radioisotopes associated with 100 MW of FTR fuel irradiated to goal exposure. Each subsequent figure illustrates the curie inventory by element for each individual isotope or isomeric state. Curie values at zero decay time are listed at the right of each figure together with the identifying symbol for each isotope. All elements and isotopes included in the RIBD calculation and nuclear data library are presented. For elements such as zinc, no inventory is calculated because the yield from the fast fission of both ^{239}Pu and ^{238}U is zero. The same scale was used in all figures to permit direct sight comparisons. In some cases, other isotopes exist but are not shown in the graphs because of the small quantity present. For those isotopes, the quantities present at zero decay time are included in the tabulated values and the reader can calculate the inventory at any decay time by using published half life information.

FIGURE A-1. Total Activity Associated with Fission Products

FIGURE A-2. Zinc Radioisotopes in FTR Fuel

FIGURE A-3. Gallium Radioisotopes in FTR Fuel

FIGURE A-4. Germanium Radioisotopes in FTR Fuel

FIGURE A-5. Arsenic Radioisotopes in FTR Fuel

FIGURE A-6. Selenium Radioisotopes in FTR Fuel

FIGURE A-7. Bromine Radioisotopes in FTR Fuel

FIGURE A-8. Krypton Radioisotopes in FTR Fuel

FIGURE A-9. Rubidium Radioisotopes in FTR Fuel

FIGURE A-10. Strontium Radioisotopes in FTR Fuel

FIGURE A-11. Yttrium Radioisotopes in FTR Fuel

FIGURE A-12. Zirconium Radioisotopes in FTR Fuel

FIGURE A-13. Niobium Radioisotopes in FTR Fuel

FIGURE A-14. Molybdenum Radioisotopes in FTR Fuel

FIGURE A-15. Technitium Radioisotopes in FTR Fuel

FIGURE A-16. Ruthenium Radioisotopes in FTR Fuel

FIGURE A-17. Rhodium Radioisotopes in FTR Fuel

FIGURE A-18. Palladium Radioisotopes in FTR Fuel

FIGURE A-19. Silver Radioisotopes in FTR Fuel

FIGURE A-20. Cadmium Radioisotopes in FTR Fuel

FIGURE A-21. Indium Radioisotopes in FTR Fuel

FIGURE A-22. Tin Radioisotopes in FTR Fuel

FIGURE A-23. Antimony Radioisotopes in FTR Fuel

FIGURE A-24. Tellurium Radioisotopes in FTR Fuel

FIGURE A-25. Iodine Radioisotopes in FTR Fuel

FIGURE A-26. Xenon Radioisotopes in FTR Fuel

FIGURE A-27. Cesium Radioisotopes in FTR Fuel

FIGURE A-28. Barium Radioisotopes in FTR Fuel

FIGURE A-29. Lanthanum Radioisotopes in FTR Fuel

FIGURE A-30. Cerium Radioisotopes in FTR Fuel

FIGURE A-31. Praseodymium Radioisotopes in FTR Fuel

FIGURE A-32. Neodymium Radioisotopes in FTR Fuel

FIGURE A-33. Promethium Radioisotopes in FTR Fuel

FIGURE A-34. Samarium Radioisotopes in FTR Fuel

FIGURE A-35. Europium Radioisotopes in FTR Fuel

FIGURE A-37. *Terbium Radioisotopes in FTR Fuel*

FIGURE A-38. Dysprosium Radioisotopes in FTR Fuel

FIGURE A-39. Holmium Radioisotopes in FTR Fuel

FIGURE A-40. Erbium Radioisotopes in FTR Fuel

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