MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0382847 3

ORNL-5621

OAK RIDGE NATIONAL LABORATORY

UNION CARBIDE

ORIGEN2—A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code

A. G. Croff

OPERATED BY
UNION CARBIDE CORPORATION
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

OAK RIDGE NATIONAL LABORATORY
CENTRAL RESEARCH LIBRARY
CIRCULATION SECTION
4500N ROOM 175
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this report, send in name with report and the library will arrange a loan.

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
NTIS price codes: Printed Copy: A04; Microfiche: A01

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

ORNL-5621
Dist. Category UC-70

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

NUCLEAR FUEL AND WASTE PROGRAMS

Waste Management Analysis for Nuclear Fuel Cycles
(Activity No. AP 05 25 10 0; FTP/A No. ONL-WH01)

ORIGEN2 - A REVISED AND UPDATED VERSION OF THE OAK RIDGE ISOTOPE GENERATION AND DEPLETION CODE

A. G. Croff

Date Published: July 1980

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
DEPARTMENT OF ENERGY

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES

3 4456 0382847 3

CONTENTS

Page

ABSTRACT 1

1. INTRODUCTION 1

1.1 Background 2

1.2 Scope of Revisions and Updates 3

1.2.1 Revision of the ORIGEN computer code 3

1.2.2 Update of the cross section and fission product yield library 3

1.2.3 Update of the decay and photon data 4

1.2.4 Update of the reactor models 5

1.2.5 Update of miscellaneous input information . . . 5

1.3 Organization of ORIGEN2 and Its Data Libraries 5

1.4 Availability of ORIGEN2 6

2. OVERVIEW OF THE ORIGEN2 COMPUTER CODE 7

2.1 Description of the Capabilities of ORIGEN2 7

2.1.1 Function of ORIGEN2 7

2.1.2 ORIGEN2 input features 8

2.1.3 ORIGEN2 calculational features 9

2.1.4 ORIGEN2 output features 12

2.2 Computer-Oriented Description of ORIGEN2 16

3. DESCRIPTION OF THE MATHEMATICAL METHOD USED IN ORIGEN2. . . 20

3.1 Matrix Exponential Solution 21

3.1.1 General solution 21

3.1.2 Computation of the matrix exponential series 21

3.2 Use of Asymptotic Solutions of the Nuclide Chain Equations for Short-Lived Isotopes 24

3.2.1 Short-lived nuclide present initially 25

3.2.2 Short-lived daughter of a long-lived parent 27

3.3 Applications of the Matrix Exponential Method for Nonhomogeneous Systems 28

Page

3.4 Computation of Neutron Flux and Specific Power 29

3.4.1 Calculation of neutron flux given specific power 29

3.4.2 Calculation of specific power given neutron flux 31

3.4.3 Other considerations 31

3.5 Construction of the Transition Matrix 32

4. DESCRIPTION AND SOURCE OF MIXCELLANEOUS ORIGEN2 DATA . . . 37

4.1 Neutron Yield per Spontaneous Fission 37

4.2 Neutron Yield per Neutron-Induced Fission 37

4.3 Neutron Yields from (α,n) Reactions 38

4.4 Elemental Chemical Toxicities 41

4.5 Nuclide-Dependent Recoverable Energy per Fission 43

4.6 ORIGEN2 Flux Parameters THERM, RES, and FAST 44

4.6.1 Derivation of ORIGEN2 flux parameters THERM and RES 46

4.6.2 Derivation of the ORIGEN2 flux parameter FAST 48

4.6.3 Conversion of ORIGEN2 flux parameters to ORIGEN flux parameters 50

4.7 ORIGEN2 Nuclide and Element Identifiers 51

5. REFERENCES 52

ORIGEN2 - A REVISED AND UPDATED VERSION OF THE OAK RIDGE ISOTOPE GENERATION AND DEPLETION CODE

A. G. Croff

ABSTRACT

ORIGEN2 is a versatile point depletion and decay computer code for use in simulating nuclear fuel cycles and calculating the nuclide compositions of materials contained therein. This code represents a revision and update of the original ORIGEN computer code which has been distributed world-wide beginning in the early 1970s. Included in it are provisions for incorporating data generated by more sophisticated reactor physics codes, free-format input, the ability to simulate a wide variety of fuel cycle flowsheets, and more flexible and controllable output features. Included in this report are: (1) a summary description of the total effort to update ORIGEN and its data bases, (2) a summary description of ORIGEN2, its capabilities, and its relationship to ORIGEN, (3) a description of the mathematical methods used in ORIGEN2 to solve the equations describing the generation and depletion of nuclides, (4) a description of the mathematical methods used in ORIGEN2 to calculate the neutron flux and specific power, and (5) a description of the sources of specific data associated with the computer code ORIGEN2. Also included are directions for obtaining ORIGEN2, its data bases, and a separate user's manual.

---

1. INTRODUCTION

The purpose of this report is to give a summary description of a revised and updated version of the original ORIGEN[1] computer code, which has been designated ORIGEN2. The remainder of this section is concerned with describing the background, scope, organization, and availability of ORIGEN2 and its data bases. Section 2 gives a more detailed description of the computer code ORIGEN2. Section 3 describes the methods used by ORIGEN2 to solve the nuclear depletion and decay equations. Finally, Section 4 documents input information necessary to use ORIGEN2 that has not been documented in supporting reports.

1.1 Background

The ORIGEN computer code was written in the late 1960s and early 1970s by the ORNL Chemical Technology Division as a versatile tool for calculating the buildup and decay of nuclides in nuclear materials. At that time, the required nuclear data libraries (decay, cross section/fission product yield, and photon) and reactor models (PWR-U, PWR-Pu, LMFBR, HTGR, and MSBR) were also developed based on the information available at that time. The computer code was principally intended for use in generating spent fuel and waste characteristics (composition, thermal power, etc.) that would form the basis for the study and design of fuel reprocessing plants, spent fuel shipping casks, waste treatment and disposal facilities, and waste shipping casks. Since fuel cycle operations were being examined generically, and thus were expected to accommodate a wide range of fuel characteristics, it was only necessary that the ORIGEN results be representative of this range. A satisfactory result was obtained by using decay and photon data from the Table of Isotopes,[2] tabulated thermal cross sections and resonance integrals,[3,4] and chain fission product yields.[5] The resonance integrals of the principal fissile and fertile species were adjusted to obtain agreement with experiment and more sophisticated calculations.

Soon after the ORIGEN computer code was documented, it was made available to users outside ORNL through the Radiation Shielding Information Center (at ORNL). The relative simplicity of ORIGEN, coupled with its convenient and detailed output, resulted in its being acquired by many organizations. Some of these organizations began using ORIGEN for applications that required greater precision in the calculated results than those for which it had originally been intended. These applications were generally much more specific than the early ORNL generic fuel cycle studies, such as environmental impact studies that required relatively precise calculations of minor isotopes such as ^{3}H, ^{14}C, ^{232}U, and $^{242,244}Cm$. The initial responses to these requirements were attempts to update specific aspects of ORIGEN and its data bases.[6,7] However, inconsistencies and a large number of different data bases soon resulted from these efforts.

1.2 Scope of Revisions and Updates

In an effort to remedy the problems described above, a concerted program was initiated in 1975 to update the ORIGEN computer code and its associated data bases and reactor models. More specifically, the following five aspects of ORIGEN were examined and updated:

1. the computer code itself,
2. cross sections and fission product yields,
3. decay and photon data,
4. reactor models,
5. miscellaneous input information.

1.2.1 Revision of the ORIGEN computer code

One of the first aspects of the ORIGEN system to undergo modification was the computer code itself, yielding the code ORIGEN2. These modifications are the subject of most of this report and of a companion report[8] and will not be described in detail here. To summarize, the method for solving the nuclide generation and depletion equations is essentially unchanged. However, the input, output, and control aspect of ORIGEN have undergone substantial changes to improve its flexibility and capability. The computer code ORIGEN2 and its capabilities will be described in more detail in Sect. 2. A general description of the methods used to solve the differential equations representing the nuclear buildup and depletion processes is given in Sect. 3.

1.2.2 Update of the cross section and fission product yield library

The major activity of the effort to update ORIGEN and its data bases was involved in updating the cross sections and fission product yields. Relatively sophisticated reactor physics calculations were undertaken for many different reactor/fuel combinations leading to a calculated neutron energy spectrum. This multigroup spectrum was then used to weigh multigroup neutron cross sections and fission product yields and to calculate new values for the ORIGEN flux parameters THERM, RES, and FAST. Spectrum-weighed cross sections were calculated for about 230 different nuclides

for each reactor/fuel combination. Independent fission product yields were obtained for over 1100 nuclides. This was then reduced to a list of about 850 nuclides by adding the yields of the very short-lived nuclides to their longer-lived progeny. For those nuclides where multigroup cross-section data were not available, the 2200 m/s cross sections and resonance integrals were updated using the most recent information. The results of this effort and the reports where the results are or will be documented are summarized in Table 1.1. Other reactor models will be added in the future as the need arises.

Table 1.1. Summary of reactor/fuel combinations for which updated cross sections and reactor models were developed

Reactor/fuel type	Reference
Uranium-plutonium cycle PWRs and BWRs	9
Alternative fuel cycle PWRs	10
Uranium-plutonium and alternative-cycle LMFBRs	11
Uranium-cycle CANDUs	12

1.2.3 Update of the decay and photon data

The decay and photon libraries were updated for about 450 of the principal radioactive nuclides using evaluated data in the Evaluated Nuclear Structure Data File (ENSDF)[13,14] at ORNL. The information in ENSDF is normally published in the Nuclear Data Sheets.[15] Information concerning decay half-lives, branching ratios, energies, intensities, photons (gamma and x-ray), and beta particles was abstracted from the file. The beta-particle data was used as input to a computer code that calculated the amount of bremsstrahlung resulting from deceleration of the beta particles. Additional information was also developed concerning photons from spontaneous fission and (α,n) reactions and fission products that decay via neutron emission (i.e., delayed neutron precursors). All of this information was combined to yield updated decay and photon libraries for ORIGEN.[16]

1.2.4 Update of the reactor models

The updating of the reactor models includes such items as fuel enrichments, fuel specific power during irradiation, fuel burnup, amount and composition of fuel assembly structural materials (e.g., cladding) per unit of fuel, and the impurity concentrations in the fuel itself. This information was updated to include more recent reactor designs and fuel cycle concepts. The results of this update are or will be contained in the references listed in Table 1.1.

1.2.5 Update of miscellaneous input information

The category "miscellaneous input information" includes many types of data whose only relationship is that they are not associated with any type of the major ORIGEN data libraries. These data include

1. neutrons per spontaneous fission,
2. neutrons per neutron-induced fission,
3. neutrons per (α,n) reaction,
4. chemical toxicities of the elements,
5. recoverable energy per fission,
6. the ORIGEN flux parameters THERM, RES, AND FAST.

This information is discussed in Sect. 4 of this report.

1.3 Organization of ORIGEN2 and Its Data Libraries

The ORIGEN2 computer code requires three different computer-readable libraries for complete operation: decay, cross-section/fission product yield, and photon. These libraries are maintained at ORNL as master libraries. The primary characteristics of the master libraries are that (1) the data for each nuclide are listed only once, and (2) the data for all reactor/fuel combinations are listed in the libraries. This mechanism ensures that only the most recent data are being used in all cases since superseded data are deleted when the master libraries are updated. However, the large amount of data in these libraries (the cross-section/fission product yield library has over 25,000 cards) and the fact that they are not

organized into the requisite ORIGEN groupings (i.e., activation products, actinides and daughters, and fission products) makes these libraries unsuitable for use directly by ORIGEN2.

The solution to this was to write a series of small computer programs which access each of the three master libraries, select the data for the desired reactor/fuel combination (cross-section/fission product yield library only), organize it into the traditional ORIGEN three-group structure, assign each group a unique number, and then output the resulting libraries. These libraries are used by ORIGEN2 and distributed to outside users.

It should be noted that it is possible to have more than one photon library because the intensity of bremsstrahlung depends heavily on the medium in which the beta particle decelerates. This necessitates multiple bremsstrahlung libraries which are then combined with the single gamma-ray/x-ray master to yield the ORIGEN2-readable library. The bremsstrahlung library normally used with ORIGEN2 assumes a UO_2 matrix. This will be conservative (i.e., result in the maximum number of photons) for most applications.

1.4 Availability of ORIGEN2

A computer code package has been deposited with the Radiation Shielding Information Center at ORNL. Inquiries or requests for the code should be mailed to:

Codes Coordinator
Radiation Shielding Information Center
Bldg. 6025
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

or telephoned to:

(615)-574-6176 or

FTS 624-6176.

2. OVERVIEW OF THE ORIGEN2 COMPUTER CODE

The purpose of this section is to give a more detailed description of the ORIGEN2 computer code and its capabilities. Although the details of the mathematical methods used in ORIGEN2 (see Sect. 3) remain substantially unchanged from the original version of ORIGEN,[1] the outward characteristics appear to be substantially different. This detailed description is divided into two components: (1) a conceptual description of the capabilities of ORIGEN2, and (2) a description of the computer-related features of ORIGEN2.

2.1 Description of the Capabilities of ORIGEN2

2.1.1 Function of ORIGEN2

The general function of the ORIGEN2 computer code is to calculate the nuclides present in various nuclear materials and output the results in common engineering units and in a lucid format. The principal calculational task involved in doing this is to determine the buildup and depletion of nuclides in these materials during irradiation and decay. Additional functions necessary to realistically simulate nuclear fuel cycles include the reprocessing (i.e., chemical separation) of nuclear materials and the continuous feed, removal, and accumulation of nuclear materials. In general, these features were present in the original version of ORIGEN although their exact scope has undergone some changes in ORIGEN2.

The buildup and depletion of nuclides during irradiation is calculated by ORIGEN2 using zero-dimensional (i.e., point) geometry and quasi-one-group neutron cross sections (see Sect. 2.1.3). This means that ORIGEN2 cannot account for spatial or resonance self-shielding effects or changes in the neutron spectrum other than those encoded initially. Thus ORIGEN2 (or ORIGEN) is not suitable for performing depletion calculations on materials until the appropriate cross sections have been obtained from more sophisticated reactor physics codes.

The ORIGEN2 Computer code's internal operations use g-atoms as the measure of the amount of a specific nuclide that is present in some mixture.

The variety of common engineering units available in the ORIGEN2 output is obtained by multiplying the g-atoms of each nuclide by constants and by nuclide-dependent values from the decay library to convert to more useful units, such as grams, curies, photons, or watts. The nuclide values are then summed to form element values and tested to generate summary tables.

2.1.2 ORIGEN2 input features

ORIGEN2 has three principal input features other than the standard reading of libraries, instructions, and material compositions: free-format input, listing of input data, and substitute data library cards. The free-format input feature means that all of the input to ORIGEN2, including the data libraries, can appear any place on a card (or card image) as long as the data are in the correct order, of the correct type (i.e., real or integer), and separated by a comma and/or at least one space. Real numbers can be in either floating point or exponential notation. This feature makes input preparation much simpler and reduces the number of bad computer runs due to a value being in the wrong position on the card. It also permits comments to be placed on virtually every card since ORIGEN2 ceases to scan the card once the last datum expected on the card is encountered.

A listing of the input data to ORIGEN2 is desirable so that the user can determine what input was used to generate the output on hand at a later date. The data libraries (decay, cross-section/fission product yield, and photon) have always been listed and are not included in this discussion. The input listing now also contains all of the card input to ORIGEN2, which generally includes the controlling instructions and the material compositions. These data are read, printed on paper, and written to a scratch file. The scratch file is then rewound and ORIGEN2 reads the data from the scratch file.

The substitute data library cards feature will read a limited number of substitute data cards from the card input unit. The data on these cards will then override the data in the primary library, which is most likely on a direct-access device or a tape. This feature eliminated the need to read in an entire library on cards simply to change a few data values.

2.1.3 ORIGEN2 calculational features

The changes in the calculational features of ORIGEN that have been incorporated in ORIGEN2 constitute a much larger and more significant set than either the modifications to the input or output aspects. A list of the more significant features is as follows:

1. The user can now define the flowsheet to be simulated in much more detail.
2. The cross sections of the principal fissile and fertile actinides vary with burnup in ORIGEN2.
3. The recoverable energy per fission (e.g., 200 MeV per fission) is now nuclide-dependent.
4. The calculated composition of materials in ORIGEN2 can be output and read back in at a later date.
5. A mechanism is included to account for the fission product yield of actinides that do not have explicit fission product yields (e.g., ^{236}U).
6. Provisions have been made for including "nonstandard" neutron-induced reactions [e.g. $(n,{}^{3}He)$].
7. The fractional recoveries (i.e., separation factors) for the ORIGEN2 reprocessing operation can be specified by individual element or by element group.

The most visible change in going from ORIGEN to ORIGEN2 is in the method of defining the case to be calculated. The ORIGEN2 case is specified by a series of individual "commands," each of which defines a single operation (such as reading the data libraries), a single irradiation step, or a single output. The commands are relatively simple, generally consisting of a three- or four-character keyword and a few numbers defining the conditions of the command. By using the commands, the user, in effect, writes a small computer program that defines the case. The program is read by ORIGEN2, "compiled" (i.e., translated and stored), and then executed. Within the bounds of logic, the user can simulate a diverse assortment of situations using the ORIGEN2 and the presently existing 30 commands. For example, the user has detailed control over the movement, summation, and placement of material compositions in the columns

visible in the ORIGEN2 output and in storage areas, which are similar in appearance. Commands are available which make the simulation of recycle situations straightforward. A different command will blend two material compositions so that the product's infinite multiplication factor (IMF) is equal to a specified value or to the IMF of a third material. Other commands, which control the ORIGEN2 output, will be discussed in Sect. 2.1.4.

Two far less visible, but extremely important, features of ORIGEN2 are the variable actinide cross sections and the nuclide-dependent recoverable energy per fission (REPF). Early in the program to update ORIGEN and its data bases, it was discovered that the use of a constant, average cross section from a sophisticated reactor physics code for the principal actinide nuclides such as $^{235,238}U$ and $^{239-242}Pu$ would not give correct depletion results. The cause of this was determined to be (1) the cross sections of some of these isotopes did not vary linearly with burnup, and (2) the cross section near the end of irradiation is more important in determining the discharge fuel composition than the cross section near the beginning of irradiation. The solution to this problem was to obtain cross sections as a function of fuel burnup for the principal actinides and use these in ORIGEN2. These cross sections are in DATA statements in ORIGEN2 in the form of discrete interpolation points. At the beginning of each irradiation step the burnup is estimated, the variable cross sections are calculated by interpolation, and the values substituted in the matrix of differential equations being solved by ORIGEN2. If a fission cross section of a nuclide having direct fission product yields is altered (e.g., ^{235}U), then the array containing the product of the fission product yields and the fission cross section is also adjusted to reflect this change. Each different reactor/fuel combination can have either a different set of variable cross sections, a different set of actinides with variable cross sections, or both. The variable cross sections for each of the reactor/fuel combinations are listed in the reports discussed in Sect. 1.2.2. A second aspect of the original ORIGEN had to be changed before the cross sections from the sophisticated reactor physics codes could be used to predict the correct results — the previously constant

REPF value of 200 MeV per fission had to be replaced by a REPF appropriate for each nuclide. If this was not done, the flux required to sustain a given amount of power was not accurate and, thus, the amounts of neutron capture products in the discharged material was in error. For plutonium-enriched systems, the error in the flux could be on the order of 5%. A more detailed discussion of the nuclide-dependent REPF is given in Sect. 4.5.

The ability of ORIGEN2 to output the calculated compositions of various materials and to read them at a later time has already proven to be a very useful option. The composition is in g-atoms and is written in the same format as that for manually punched compositions. The mass of each nuclide is tested before being written and, if it is less than some cutoff value (10^{-25} being typical), the nuclide is not output. This feature's principal use thus far has been in allowing irradiation calculations to be separated from decay calculations. An irradiation calculation is performed with ORIGEN2, and the charge and discharge compositions are written to some data storage device. A separate ORIGEN2 run then reads this information and performs the decay and reprocessing calculations, which are generally the desired product. This procedure eliminates the necessity for repeating the irradiation calculation, which is very time consuming, every time a different decay time or output table is desired. Additionally, the set of equations to be solved in the decay calculations is significantly smaller than that for the irradiation calculations since flux-dependent reactions are not included.

A mechanism has been included in ORIGEN2 to account for fission product yields from those actinides that have nonzero fission cross sections but which are not explicitly included in the data base as are the yields for the principal actinides ($^{233,235,238}U$, $^{239,241}Pu$, ^{232}Th). At the beginning of each irradiation step ORIGEN2 sums the fission rates of all of the actinides that do <u>not</u> have explicit fission product yields (i.e., unconnected actinides) and also determines the largest contributor in this category. The identity of the largest unconnected actinide contributor is used to find the nearest neighboring actinide that does have explicit fission product yields (i.e., the nearest connected actinide). The fission

product yields of the nearest connected actinide are adjusted upward to compensate for the yield of all of the unconnected actinides. In thermal reactors, this correction is usually small (<0.5%). However, in a fast reactor the correction factor is on the order of 7%, principally due to ^{240}Pu.

Provisions have been made in ORIGEN2 for the inclusion of nonstandard, flux-dependent reactions in the calculation. A nonstandard reaction is defined as a reaction for which there is no provision in the cross-section/fission product yield library. Although an extremely large number of reactions could fall into this class, only a very few are of any significance. Examples of this type of reaction are $^{239}Pu(n,4n)\ ^{236}Pu$ and $^{16}O(n,^{3}He)^{14}C$.

Finally, the fractional recoveries that are used by ORIGEN2 to calculate the products of a reprocessing (chemical separation) operation can be specified by element group as well as by individual element. In the element group method, each element is assigned to a group based on its chemical behavior during separation. For example, all of the noble gases would be assigned to the same group. Then each group has associated with it a fractional recovery which is appropriate for all of the elements in the group. This approach greatly reduces the number of fractional recoveries that have to be specified or changed since 10 to 15 groups are usually adequate and the element membership of a group is relatively constant.

2.1.4 ORIGEN2 output features

The basic appearance of the ORIGEN2 output is the same as that in the original version of ORIGEN. However, several features have been added to make the output more versatile and easy to use:

1. Additional output table types have been incorporated.
2. "Road map" features have been added to enable the user to more easily find the desired information.
3. Dual output units have been incorporated to minimize the amount of output on paper.

4. The user can control the labeling of the ORIGEN2 output columns.
5. A more flexible testing procedure for summary tables has been incorporated.
6. Auxiliary information is printed on separate output units.

ORIGEN2 has provisions for writing 22 different types of output Tables. These table types are listed in Table 2.1. The 22 available table types are much larger than the 7 available in the original version, principally because of the addition of the "fractional" tables which calculate the fraction of a table total constituted by each nuclide and element. Aside from these tables, the new types of tables are as follows:

1. Isotopic composition of each element. Gives the isotopic composition of each element in a table in atom fraction or weight fraction (i.e., each element totals to 1.0, and the table total is equal to the number of elements).
2. Chemical ingestion toxicity. Gives the amount of water required to dilute a material to chemically acceptable levels. Obtained by dividing the grams of each element by the maximum permissible concentration in g/m^3 water; see Sect. 4.4 for further details.
3. Neutron absorption rate. Gives the absorption rate of neutrons in each nuclide and element in neutrons/s.
4. Neutron-induced fission rate. Gives the fission rate of the actinide nuclides and elements in fissions/s.
5. Radioactivity (alpha). Gives the alpha decay rate for each actinide nuclide and element in curies.

These table types were incorporated to alleviate the need to perform extensive manual calculations to obtain certain types of information. Provisions for controlling the output tables are also included in ORIGEN2 so only those tables that are desired need to be output.

"Road map" features have been added to ORIGEN2 to aid the user in finding the information of interest. These features are the following:

1. Each output page is labeled "Activation Products," "Actinides + daughters," or "Fission Products," according to the contents of

Table 2.1. ORIGEN2 output table description

Table description	Units
Isotopic composition of each element	atom fraction
Isotopic composition of each element	weight fraction
Composition	g-atoms
Composition	atom fraction
Composition	g
Composition	wt fraction
Radioactivity (total)	Ci
Radioactivity (total)	fractional
Thermal power	watts
Thermal power	fractional
Radioactive inhalation toxicity	m^3 air
Radioactive inhalation toxicity	fractional
Radioactive ingestion toxicity	m^3 water
Radioactive ingestion toxicity	fractional
Chemical ingestion toxicity	m^3 water
Chemical ingestion toxicity	fractional
Neutron absorption rate	neutrons/s
Neutron absorption rate	fractional
Neutron-induced fission rate	fissions/s
Neutron-induced fission rate	fractional
Radioactivity (α)	Ci
Radioactivity (α)	fractional
(α,n) neutron production	neutrons/s
Spontaneous fission neutron production	neutrons/s
Photon emission rate	photons/s, MeV/s, MeV/watt-s

the page.

2. Each output page is numbered sequentially.
3. A table of contents is printed during execution that lists each table printed and the page on which it begins.

The first of these greatly aids in distinguishing the activation product tables from the fission product tables since both have many of the same nuclides. Hopefully, the combination of the latter two features will enable the user to scan the table of contents, identify the table of interest and its page number, and then proceed directly to the correct page.

Dual output units have also been incorporated into ORIGEN2. The principal objective of having dual output units is to allow a reduced amount of output to be printed on the first unit while writing a much larger amount of output to a storage device or on microfiche using the second unit. This procedure is currently in use at ORNL and results in considerable information being available with only the most commonly used information (i.e., mass, radioactivity, thermal power, neutron activity, and photon emission rate) appearing on paper.

The user can also control the heading of the ORIGEN2 output columns by using a command that allows a column heading of up to 10 characters to be specified. This heading will remain associated with the column during most of the operations carried out in ORIGEN2. Commonly used alphanumeric headings are CHARGE, DISCHARGE, HLW, etc.

A more flexible testing procedure for the output summary tables has been included. The summary tables contain only those nuclides or elements that contribute more than a certain fraction of the table total. In the past, the contribution of each nuclide in a single ORIGEN column was compared to a specified cutoff value. If it was greater, the nuclide was included in the summary table; if it was less than the cutoff value, it was excluded. In ORIGEN2, the cutoff value is not a specific number, but rather a fraction of the column total. Thus the contribution of a nuclide or element is tested against the cutoff value (fraction) times the column total. Commonly used cutoff values are 0.1% and 1%. A second change in the summary tables is that the user can specify whether one column is to

be tested, all columns except one are to be tested, or all columns are to be tested. If multiple columns are tested, a nuclide is printed in the summary table if it is larger than the cutoff value times the column total for <u>any</u> of the columns tested.

The final major output change is the printing of various auxiliary information used internally in ORIGEN2. One type of auxiliary information printed concerns the variable cross sections that are being used in each ORIGEN2 irradiation step and the adjustment of the fission product yields to account for actinides without explicit fission product yields (see Sect. 2.1.3). Another type of auxiliary information output is a one-line message printed at the beginning of execution of each ORIGEN2 command. This serves to inform the user of exactly where in the stream of commands an error occurred. Also included in this output stream is internal information concerning the calculation of the neutron flux, specific power, and other parameters not directly specified by the user. A final type of auxiliary information, which is printed at the beginning execution of each output command, is the amount of space required thus far in the calculation. This type of information is required to make proper use of the variable dimensioning feature of ORIGEN2 (see Sect. 2.2.1).

2.2 Computer-Oriented Description of ORIGEN2

The ORIGEN2 computer code is currently comprised of about 7300 source statements. It is written entirely in the FORTRAN computer language. Certain IBM-specific features (e.g., partial-length words) have been used in the version at ORNL to conserve space during execution. However, these features are specified in such a way that their elimination is not particularly difficult. A CDC-compatible version of ORIGEN2 is also available from the ORNL Radiation Shielding Information Center (see Sect. 1.4).

There are currently about 60 subroutines in the ORIGEN2 source deck. The number of subroutines will continue to increase slowly since the addition of new reactor models necessitates the addition of cross sections contained in a new subroutine. Since there are so many subroutines and the ORIGEN2 internal logic can be somewhat complicated, a list of the

subroutines and their general function(s) is given in Table 2.2. The subroutine scopes have been defined so as to facilitate the use of the OVERLAY function which places only the necessary subroutines in core at a given time and thus minimizes the amount of space required during execution.

The subroutines are also set up to facilitate the use of variable dimensions. The dimensions of all major arrays can be varied in the primary calling routine (MAIN), which is comprised of about 80 cards. The other primary function of MAIN is to call the subroutines MAIN1, MAIN2, MAIN3, and LISTIT. The dimensions of the arrays in MAIN for a wide variety of cases are given in the ORIGEN2 user's manual[8] along with an estimate of the amount of space required for a particular case.

Finally, it might be useful for the user to have some feeling for the order in which the ORIGEN2 subroutines are called to facilitate input specification and debugging. MAIN is the primary routine which calls LISTIT, MAIN1, MAIN2, and MAIN3 sequentially. MAIN2 calls the appropriate XSECnn sybroutine. MAIN3 calls a large number of subroutines with the order depending significantly on the user-specified commands. However, in general, the first subroutines called from MAIN3 are NUDAT1 (plus DECRED), NUDAT2 (plus SIGRED), NUDAT3, and NUDOC, which read the data libraries, and ANSF, which sets up the spontaneous fission and (α,n) neutron data for later use. PHOLIB would then be called to read the photon library. Next, initial material compositions would be read by MAIN3. Then the irradiation/decay subroutines FUDGE, FLUXO, DECAY, TERM (plus MATREX), and EQUIL are called, in the order listed, to calculate the nuclide generation and depletion. The final operation is to output the results. This involves calls to OUTPUT, OUT1, OUT2, NUTRON, and GAMMA. The other subroutines in Table 2.2 are called in a wide variety of places at various times.

Table 2.2. Functional description of ORIGEN2 subroutines

Subroutine or function subprogram	Description
MAIN1	Initializes and/or reads data such as neutrons per spontaneous and neutron-induced fission, chemical toxicities, and fractional reprocessing recoveries
MAIN2	Reads and stores the information on the ORIGEN2 commands; obtains the correct set of variable cross sections from a XSECnn subroutine (see below) if required
MAIN3	Executes the ORIGEN2 commands read by MAIN2; this is actually a control subroutine since most of the calculations are done in the subroutines called by MAIN3
OUTPUT	Controls output of most tables
OUT1	Sums columns to generate totals for use in printing fractional tables and summary tables
OUT2	Writes most tables
NUDAT1	Reads and temporarily stores the decay library
NUDAT2	Reads and temporarily stores the cross-section/fission product yield library
NUDAT3	Accesses the temporarily stored information from NUDAT1 and NUDAT2 and rearranges it to form the matrix of equations solved by ORIGEN2 (see Sect. 3.5)
NUDOC	Prints of ORIGEN2 statements pointing to the documentation
DECRED	Reads the decay data for a nuclide; called by NUDAT1
SIGRED	Reads the cross-section/fission product yield data for a nuclide; called by NUDAT2
ANSF	Accesses, combines, and stores internal information related to the production of spontaneous fission and (α,n) neutrons for future use

Table 2.2 (Continued)

Subroutine or function subprogram	Description
PHOLIB	Reads, organizes, and stores the photon library
NUTRON	Outputs the spontaneous fission and (α,n) neutron production rate tables based on information stored by ANSF
GAMMA	Outputs the photon tables based on information stored by PHOLIB
HEAD	Writes the mixture of ORIGEN2-generated and user specified column headings; called mostly by OUTPUT, OUT2, GAMMA, and NUTRON
TOC	Prints the table of contents
FLUXO	Calculates the neutron flux from specific power or vice-versa; nuclide-dependent recoverable energy per fission and the fission product yield adjustments are also accomplished here
FUDGE	Calculates and incorporates the correct variable actinide cross sections
DECAY, TERM, MATREX, EQUIL	Solve buildup and depletion equations
NOAH	Converts six-digit integer nuclide identifiers (see Sect. 4.7) into an alphanumeric element symbol, three-digit atomic mass, and one character ground/excited state identifier
RMASS	Returns the atomic mass of a nuclide to the calling subroutine
RTIME	Returns a factor for converting the specified time units to seconds to the calling subroutine
IPAGE	Returns the current page number to the calling subroutine
LISTIT	Prints the card input data on paper and writes it to a temporary data set; called from MAIN

Table 2.2 (Continued)

Subroutine or function subprogram	Description
ADDMOV	Adds and moves ORIGEN2 columns; used extensively internally
BLOCK DATA	Initializes a large number of variables and labels
AREAD, DREAD, QQPACK, QQREAD, READ, IREAD	Read free-format input; QQPACK is machine-dependent
XSEC01 TO XSECnn	Contain the variable cross sections with one reactor/fuel type per subroutine

3. DESCRIPTION OF THE MATHEMATICAL METHOD USED IN ORIGEN2

This section presents a summary description of the mathematical methods used to solve (1) the differential equations describing the build-up and depletion of nuclides, and (2) the equations used to calculate the neutron flux level and the specific power during irradiation. A relatively detailed description of the methods used to generate and store the matrix to be solved is also given. These descriptions are based heavily on the original write-up given in ref. 1 because the mathematical methods used in ORIGEN2 are fundamentally the same as those used in ORIGEN.

A general expression for the formation and disappearance of a nuclide by nuclear transmutation and radioactive decay may be written as follows:

$$\frac{dX_i}{dt} = \sum_{j=1}^{N} \ell_{ij}\lambda_j X_j + \bar{\phi} \sum_{k=1}^{N} f_{ik}\sigma_k X_k - (\lambda_i + \bar{\phi}\sigma_i)X_i \quad (i = 1,\ldots N) \qquad (1)$$

where X_i is the atom density of nuclide $\underline{i}$, λ_i is the radioactive disintegration constant for nuclide $\underline{i}$, σ_i is the spectrum-averaged neutron

absorption cross section of nuclide $\underline{i}$, and ℓ_{ij} and f_{ik} are the fractions of radioactive disintegration and neutron absorption by other nuclides which lead to the formation of species $\underline{i}$. Also in Eq. (1), $\bar{\phi}$ is the position- and energy-averaged neutron flux. Rigorously, the system of equations described by Eq. (1) is nonlinear since the neutron flux and cross sections will vary with changes in the composition of the fuel. However, the variation with time is slow and, if they are considered to be constant over short time intervals, the system of Eq. (1) is a homogeneous set of simultaneous first-order ordinary differential equations with constant coefficients, which may be written in matrix notation as

$$\dot{\underset{\sim}{X}} = \underset{\approx}{A}\,\underset{\sim}{X} \; . \tag{2}$$

3.1 Matrix Exponential Solution

3.1.1 General solution

Equation (2) has the known solution

$$\underset{\sim}{X}(t) = \exp\,(\underset{\approx}{A}t)\;\underset{\sim}{X}(0), \tag{3}$$

where $\underset{\sim}{X}(0)$ is a vector of initial atom densities and $\underset{\approx}{A}$ is a transition matrix containing the rate coefficients for radioactive decay and neutron capture. The function exp $(\underset{\approx}{A}t)$ in Eq. (3) is the matrix exponential function, a matrix of dimension N^2, which is defined as

$$\exp\,(\underset{\approx}{A}t) = \underset{\approx}{I} + \underset{\approx}{A}t + \frac{(\underset{\approx}{A}t)^2}{2!} + \ldots = \sum_{m=0}^{\infty} \frac{(\underset{\approx}{A}t)^m}{m!} \tag{4}$$

If one can generate this function accurately from the transition matrix, then the solution of the nuclide chain equations is readily obtained.

3.1.2 Computation of the matrix exponential series

Two principal difficulties are encountered in employing the matrix exponential technique to solve large systems of equations: (1) a large amount of memory is required to store the transition matrix

and the matrix exponential function, and (2) computational problems are encountered in applying the matrix exponential method to systems of equations with widely separated eigenvalues. The generation and storage of the transition matrix are explained in Sect. 3.5. The computation and storage of the matrix exponential function have been facilitated by developing a recursion relation for this function which does not require storage of the entire matrix. Thus it is possible to derive an expression for one nuclide in Eq. (3) which is given by

$$x_i(t) = \sum_{n=0}^{\infty} C_i^n \ , \tag{5}$$

where C_i^n is generated by use of a recursion relation

$$C_i^0 = x_i(0) \ , \tag{6a}$$

$$C_i^{n+1} = \frac{t}{n+1} \sum_{j=1}^{N} a_{ij} C_j^n \ . \tag{6b}$$

Here, a_{ij} is an element in the transition matrix that is the first-order rate constant for the formation of species $\underline{i}$ from species $\underline{j}$. This algorithm requires storage of only one vector $\underset{\sim}{C}^n$ in addition to the current value of the solution.

In performing the summation indicated by Eq. (5), it is necessary to ensure that precision in the answer will not be lost due to the addition and subtraction of nearly equal large numbers. In the past, this objective has been accomplished by scaling the time step by repeatedly dividing by two until the norm of the matrix is less than some acceptable small value, computing the matrix exponential function for the reduced time step, and repeatedly squaring the resulting matrix to obtain the desired time step.[17-19] Such a procedure would be impracticable for a computation involving large numbers of nuclides (many of which have short

half-lives) corresponding to large norms of the $\underset{\approx}{A}t$ matrix. However, it is just for these short-lived isotopes that the conditions of secular and transient equilibrium are known to apply. Thus in the computations performed by ORIGEN2, only the compositions of those nuclides whose diagonal matrix elements are less than a predetermined value are computed by the matrix exponential method. The concentrations of the isotopes with large diagonal matrix elements are computed using an analytical expression for the conditions of secular or transient equilibrium, as described in Sect. 3.2.

Lapidus and Luus[19] have shown that the accuracy of the computed matrix exponential function can be maintained at any desired value by controlling the time step such that the norm of the matrix $\underset{\approx}{A}t$ is less than a predetermined value which is fixed by the word length of the digital computer used in the calculations. They define a norm of the matrix $\underset{\approx}{A}$, denoted by [A], as the smaller of the maximum-row absolute sum and the maximum-column absolute sum:

$$[A] = \min \left\{ \max_j \sum_i \left| a_{ij} \right| , \max_i \sum_j \left| a_{ij} \right| \right\} , \tag{7}$$

where $|a_{ij}|$ denotes the absolute value of the element a_{ij}. They show that the maximum term in the summation for any element in the matrix exponential function cannot exceed $\frac{n^n}{n!}$, where n is the largest integer not larger than [A]t. Consideration of the word length of the computer used to perform the calculations will indicate the maximum value of n that can be used while obtaining a desired degree of significance in the results. Using double precision arithmetic, the IBM 360 operating system can perform operations retaining 16 significant decimal figures. In the ORIGEN2 code, the norm of the transition matrix is restricted to be less than $[A] \leq -2 \ln 0.001 = 13.8155$, so that the maximum term that will be calculated will be approximately 49,000. Thus a value as small as $\exp(-13.8155) = 10^{-6}$ can be computed, while retaining five significant figures. A sufficient number of terms must be added to the infinite summation given by Eq. (5) to ensure that the series has converged. The

mth term in the series for $e^{[A]}$ is equal to $\frac{[A]^m}{m!}$, which, for large values of m, can be approximated by $(\frac{[A]\ e}{m})^m (2m\pi)^{-1/2}$, using Stirling's approximation. The value of the norm, [A], is calculated by the code; and m is set equal to the largest integer in $\frac{7}{2}$ [A] + 5, which has been determined as a "rule of thumb" for the number of terms necessary to limit the error to <0.1%. Thus for [A] equal 13.8155, 53 terms will be required in the summation. The absolute value of the last term added to the summation in this case will be $<6.4 \times 10^{-10}$, which is sufficiently small compared with 10^{-6}. It has been observed that the norm is usually less than its maximum value, and, in most cases, 30 or fewer terms are required to evaluate the series.

It has been mentioned that, in previous applications of the matrix exponential method, the restriction of the size of the norm of the transition matrix necessary to treat nuclides with large eigenvalues was accomplished by repeatedly dividing the matrix by 2, and the final value of the matrix exponential function was obtained by repeatedly squaring the resulting intermediate matrix exponential function. In the present application, the suggestion of Ball and Adams[20] that the transitions involving isotopes with large decay constants be considered "instantaneous" was adopted; that is, if A → B → C and if the decay constant for B is large (i.e., B is short-lived), the matrix is reformulated as if C were formed from A directly, and the concentration of B is obtained by an alternative technique. Similarly, if the time constant for A is very large, the transition matrix is rewritten as if the amount of isotope B initially present were equal to A + B, and only the transition B → C is obtained by the matrix exponential technique. This reduction of the transition matrix and the generation of the solution by the matrix exponential method are performed by the subroutine TERM.

3.2 Use of Asymptotic Solutions of the Nuclide Chain Equations for Short-Lived Isotopes

The numerical techniques described in Sect. 3.1 are applied only to obtain the solutions for isotopes that are sufficiently long-lived to satisfy the criterion that the norm of the transition matrix be <2 ln 1000.

Short-lived isotopes are treated by using linear combinations of the homogeneous and particular solutions of the nuclide chain equations that are computed using alternative procedures.

3.2.1 Short-lived nuclide present initially

The quantity of a short-lived nuclide (originally present at the beginning of an interval) that remains at the end of the interval is computed in subroutine DECAY using a generalized form of the Bateman equations which treats an arbitrary forward-branching chain. The generalized treatment is achieved by searching through the transition matrix and forming a queue of all short-lived precursors of an isotope. The Bateman equation solution is then applied to this queue. The queue is terminated when an isotope having no short-lived precursors is encountered. The algorithm also has provisions for treating two isotopes with equal eigenvalues and for treating cyclic chains.

Bateman's solution for the ith member in a chain at time t may be written in the form[21]

$$N_i(t) = N_1(0)e^{-d_i t}$$

$$+ \sum_{k=1}^{i-1} N_k(0) \left[\sum_{j=k}^{i-1} \frac{\exp(-d_j t) - \exp(-d_i t)}{(d_i - d_j)} a_{j+1,j} \prod_{\substack{n=k \\ n \neq j}}^{i-1} \frac{a_{n+1,n}}{d_n - d_j} \right], \quad (8)$$

where $N_j(0)$ is the amount of isotope j initially present and the members of the chain are numbered consecutively for simplicity. This method of solution used the convention that $\prod_{n=k}^{i-1} a_{n+1,n}$ is equal to the product $a_{k+1,k}, a_{k+2,k+1} \cdots a_{i,i-1}$, and that the empty product ($k \geq i$) is equal to unity. The notation $a_{i,j}$ for the first-order rate constant is the same as that described in Sect. 2.1, and $d_i = -a_{i,i}$. In the present application, Eq. (8) is recast in the form

$$N_i(t) = N_i(0)e^{-d_i t} + \sum_{k=1}^{i-1} N_k(0) \prod_{n=k}^{i-1} \frac{a_{n+1,n}}{d_n} \left[\sum_{j=k}^{i-1} d_j \frac{\exp(-d_j t) - \exp(-d_i t)}{(d_i - d_j)} \prod_{\substack{n=k \\ n \neq j}}^{i-1} \frac{d_n}{d_n - d_j} \right] \tag{9}$$

by multiplication and division by $\prod_{n=k}^{i-1} d_n$. The first product in the outer summation of Eq. (9) has significance because it is the fraction of atoms of isotope k that follow a particular sequence of decays and captures. If this product becomes $<10^{-6}$, contributions from nuclide k and its precursors to the concentration of nuclide i are neglected. The inner summation in Eq. (9) is performed in double precision arithmetic to preserve accuracy. This procedure is unnecessary for evaluating the outer summation because all the terms in this sum are known to be positive. The difficulties described by Vondy[21] in applying the Bateman equations for small values of $d_i t$ do not occur in the present application since, when this condition occurs, the matrix exponential solution is employed. The matrix exponential method and the Bateman equations complement each other; that is, the former method is quite accurate when the magnitude of the characteristic values of the equations to be solved is small, whereas the Bateman solution encounters numerical problems in this range. For the case where two isotopes have equal removal constants ($d_i = d_j$), the second summation in Eq. (9) becomes

$$\sum_{j=k}^{i-1} d_j t \, e^{-d_j t} \prod_{\substack{n=k \\ n \neq j}}^{i-1} \frac{d_n}{d_n - d_j} \, . \tag{10}$$

An analogous expression is derived for the case when $d_n = d_j$. These forms of the Bateman equations are applied when two isotopes in a chain have the same diagonal element or when a cyclic chain is encountered, in which case a nuclide is considered to be its own precursor.

3.2.2 Short-lived daughter of a long-lived parent

In the situation where a short-lived nuclide has a long-lived precursor, a second alternative solution is employed. In this instance, the short-lived daughter is assumed to be in secular equilibrium with its parent at the end of any time interval. The concentration of the parent is obtained from subroutine TERM, and the concentration of the daughter is calculated in a subroutine named EQUIL by setting Eq. (3) equal to zero:

$$\dot{x}_i = 0 = \sum_{j=1}^{N} a_{ij} \, x_j . \tag{11}$$

Equation (11), which is a set of linear algebraic equations for the concentrations of the short-lived isotopes, is readily solved by the Gauss-Seidel iterative technique.[22] The coefficients in Eq. (11) have the property that all the diagonal elements of the matrix are negative and all off-diagonal elements are positive. The algorithm involves inverting Eq. (11) and using assumed or previously calculated values for the unknown concentrations to estimate an improved value, that is,

$$x_i^{k+1} = - \frac{1}{a_{ii}} \sum_{\substack{j=1 \\ j \neq i}}^{N} a_{ij} \, x_j^k . \tag{12}$$

The iterative procedure has been found to converge very rapidly since, for these short-lived isotopes, cyclic chains are not usually encountered and the procedure reduces to a direct solution.

3.3 Application of the Matrix Exponential Method for Nonhomogeneous Systems

Certain problems that involve the accumulation of radioactive materials at a constant rate and are of engineering interest require the solution of a nonhomogeneous system of first-order linear, ordinary differential equations. In matrix notation, one writes

$$\dot{\underset{\sim}{X}} = \underset{\approx}{A}\,\underset{\sim}{X} + \underset{\sim}{B}. \tag{13}$$

This set of equations has the particular solution

$$\underset{\sim}{X}(t) = [\exp\,(\underset{\approx}{A}t) - \underset{\approx}{I}]\,\underset{\approx}{A}^{-1}\,\underset{\sim}{B}\ , \tag{14}$$

provided that $\underset{\approx}{A}^{-1}$ exists. Substituting the infinite series representation for the matrix exponential function, one obtains

$$\underset{\sim}{X}(t) = [\underset{\approx}{I} + \frac{\underset{\approx}{A}t}{2!} + \frac{(\underset{\approx}{A}t)^2}{3!} + \ldots]\,\underset{\sim}{B}t \tag{15a}$$

$$= \left(\sum_{m=0}^{\infty} \frac{(\underset{\approx}{A}t)^m}{(m+1)!} \right) \underset{\sim}{B}t. \tag{15b}$$

It should be noted that in many of the cases solved by ORIGEN2, $\underset{\approx}{A}^{-1}$ in Eq. (14) does not exist. However, the series solution [Eqs. (15a) and (15b)] does always exist; when substituted into Eq. (13), the correct solution results. Furthermore, since each term of Eq. (15) is smaller than the corresponding term in Eq. (4), Eq. (15) converges for each $\underset{\approx}{A}t$ for which Eq. (4) converges.

The particular solution may also be expressed as the sum of an infinite series

$$x_i(t) = \sum_{n=1}^{\infty} D_i^n\ , \tag{16}$$

whose terms are generated by use of a recursion relation

$$D_i^1 = b_i t \ , \tag{17a}$$

$$D_i^{n+1} = \frac{t}{n+1} \sum_{j=1}^{N} a_{ij} D^n_j . \tag{17b}$$

Once again, the algorithm is applied only to long-lived nuclides, and the concentrations of the short-lived nuclides are obtained by an alternative technique. In this situation, Eq. (11) is modified to the form

$$x_i = 0 = \sum_{j=1}^{N} a_{ij} x_j + b_i \tag{18}$$

and is solved by the Gauss-Seidel method. After the homogeneous and particular solutions have been obtained, they are added to obtain the complete solution of the system of equations.

3.4 Computation of Neutron Flux and Specific Power

To accurately compute changes in fuel composition during irradiation at constant power, it is necessary to take into account changes in the neutron flux with time as the fuel is depleted. The neutron flux is a function of the amount of fissile nuclides per unit of fuel, the fission cross section for each fissile nuclide, and the recoverable energy per fission for each nuclide.

3.4.1 Calculation of neutron flux given specific power

At the start of the computation, the known parameters are the initial fuel composition, the constant specific power that the fuel must produce during a time interval, and the length of the time interval.

The instantaneous neutron flux is related to the constant specific power at a fixed time by the equation

$$P = 1.602 \times 10^{-19} \, \phi \sum_i x_i \, \sigma_i^f \, R_i \quad , \tag{19}$$

where P is the specific power, in MW per unit of fuel; x_i is the amount of fissile nuclide i present in the fuel in g-atoms per unit of fuel; σ_i^f is the microscopic fission cross section for nuclide i; ϕ is the instantaneous neutron flux, in neutrons cm^{-2} s^{-1}; and R_i is the recoverable energy per fission for nuclide i, in MeV/fission. The constant in Eq. (19) converts MeV/sec into megawatts.

An approximate expression for the value of the neutron flux as a function of time during the interval is obtained by solving Eq. (19) for the instantaneous neutron flux ϕ and expansion of the resulting expression in a Taylor series about the start of the interval:

$$\phi(t) = 6.242 \times 10^{18} \, P \left[S(0)^{-1} - t \frac{\dot{S}(0)}{S(0)^2} + \frac{t^2}{2} \frac{2 \, \dot{S}(0)^2 - S(0) \, \ddot{S}(0)}{S(0)^3} + \ldots \right] \tag{20a}$$

or

$$\phi(t) = \phi(0) \left[1 - t \frac{\dot{S}(0)}{S(0)} + \frac{t^2}{2} \frac{2 \, \dot{S}(0)^2 - S(0) \, S(0)}{S(0)^2} + \ldots \right] \quad . \tag{20b}$$

In this expression, the parameter $S(0) = \sum_i x_i \, \sigma_i^f \, R_i$ at the start of the time interval. $\dot{S}(0)$ and $\ddot{S}(0)$ are the first and second derivatives of S evaluated at the start of the interval. The values of the derivatives of S(0) can be evaluated since $\dot{\underset{\sim}{X}}(0) = \underset{\approx}{A}\underset{\sim}{X}(0) + \underset{\sim}{B}(0)$ and $\ddot{\underset{\sim}{X}}(0) = \underset{\approx}{A}\dot{\underset{\sim}{X}}(0)$.

The average neutron flux during the interval is obtained by integrating over the interval and dividing by the length of the interval t:

$$\bar{\phi} = \phi(0) \left[1 - \frac{t}{2} \frac{\dot{S}(0)}{S(0)} + \frac{t^2}{6} \left(\frac{2 \, \dot{S}(0)^2 - S(0) \, \ddot{S}(0)}{S(0)^2} \right) + \ldots \right] . \tag{21}$$

Equation (21) is used in subroutine FLUXO of ORIGEN2 to estimate the average neutron flux during a time interval based on the conditions at the start of the interval. The term involving the second derivative is only employed for the first time interval where, for some nuclides,

$\dot{X}$ is zero but $\ddot{X}(0)$ is nonzero.

3.4.2 Calculation of specific power given neutron flux

The average power produced during a time interval for a fuel in a fixed neutron flux is estimated from the initial composition using a similarly derived equation:

$$P = 1.602 \times 10^{-19} \phi S(0) \left[1 + \frac{t}{2} \dot{S}(0) + \frac{t^2}{6} S(0) + \ldots\right] . \qquad (22)$$

3.4.3 Other considerations

In order for the above procedures to estimate the average neutron flux or average specific power correctly, the changes in neutron flux during the interval must be relatively small. If the average value of either of these quantities differs from the initial value by more than 20%, a message will be printed out advising the user to employ smaller time increments.

It was noted at the beginning of this discussion that the neutron flux is a function of the amount of fissile nuclides present, the nuclide fission cross sections, and the recoverable energy per fission. The variation in the amount of each fissile nuclide is accounted for by the derivatives in the time series described above and will not be considered further here. The nuclide fission cross sections also vary during the time interval but do not necessarily correlate with the variation in nuclide mass. This variation is accommodated by calculating the parameter S(0) and its derivatives using fission cross sections appropriate for the estimated fuel burnup at the middle of the time interval (see Sect. 2.1.3). Finally, the variation in the recoverable energy per fission is accounted for by using a constant value appropriate for each nuclide instead of a single constant value for all nuclides.

3.5 Construction of the Transition Matrix

An extensive library of nuclear properties of radioactive isotopes has been compiled for use with the ORIGEN2 code.[9-12] The data are in the form of half-lives, fractions of transitions that produce a given nuclear particle, cross sections, and fractions of absorptions that yield certain particles. These data are read from tape, direct access device, or cards and processed into a form for use by the mathematics routines in subroutines NUDAT1, NUDAT2, and NUDAT3 (hereafter called subroutines NUDATn).

It is possible to compute the concentrations of as many as 1700 nuclides using the present code. However, straightforward construction of a generalized transition matrix would require the storage of a 1700 by 1700 array, which would tax the storage capacity of the largest computers available today. On the other hand, the transition matrix is typically very sparse, and storage requirements can be reduced substantially by storing only the nonzero elements of the matrix and two relatively small vectors that are used to locate the elements. Subroutines NUDATn are also used to generate the compacted transition matrix and the two storage vectors.

Subroutines NUDATn process the libraries by reading a six-digit identifying number, NUCL(I), for each nuclide, followed by the half-life and the fraction of each decay that occurs by several competing processes from the decay library. Neutron absorption cross sections for (n,γ), (n,α), (n,p), (n,2n), (n,3n), (n,fission) reactions, and fission product yields are then read from the cross-section library. The six-digit identifying number is equal to Z * 10,000 + W * 10 + IS, where Z is the atomic number, W is the atomic weight (in integral atomic mass units), and IS is either 0 or 1 to indicate a ground state or a metastable state, respectively. This information is processed into a compacted transition matrix, as described below.

First, the half-life is used to calculate the radioactive disintegration constant, λ. First-order rate constants for various competing decay processes are calculated by multiplying λ by the fraction of

transitions to that final state. The product nuclide resulting from each nuclear transition is next identified by the addition of a suitable constant to the six-digit identification number for the parent nuclide. (For example, for a β^- decay, 10,000 is added to the parent identifier; for neutron capture, 10 is added; or for isomeric transition the quantity -1 is added.) Two arrays are constructed: the first, NPROD(J,M), contains all the products which can be directly formed from any nuclide J by the transitions considered in the library; and the second, COEFF(J,M), contains the first-order rate constants for the corresponding transitions. When these arrays have been constructed, a search of the NPROD array is conducted to identify all the parents of a given nuclide I. [Nuclide J is a parent of I if NPROD(J,M) equals NUCL(I) for any reaction of type M.] When a parent of nuclide I has been located, the value of the corresponding coefficient a_{ij} in the transition matrix is equal to COEFF(J,M). However, direct storage of a_{ij} in a square array would require an excessive amount of storage. Hence this procedure is avoided by incrementing an index, N, each time a coefficient is identified. The coefficients are stored sequentially in a one-dimensional array, A(N); the value of J is stored in another one-dimensional array, LOC(N); and the total number of coefficients for production of nuclide I are stored in a third array, NØNO(I). When all of the coefficients for every nuclide have been stored, the NØNO(I) array is converted to indicate the cumulative number of matrix coefficients for all the isotopes up to and including I [i.e., NØNO(I + 1) = NØNO(I) + NØNO(I + 1) for all values of I greater than 1]. After this procedure has been executed, the NØNO array is a monotonically increasing list of integers whose final value is the number of nonzero, off-diagonal matrix elements in the transition matrix. This final value is preserved separately as the variable NON. For computational convenience, the values of the diagonal matrix elements are stored in a separate vector, D(I). To perform the multiplication of the transition matrix by a vector (e.g., $\dot{x}_i = \sum_{j=1}^{N} a_{ij}x_j$), as is required to execute the algorithm described in Sect. 3.1, the operations described in the flowchart given in Fig. 3.1 are employed.

ORNL-DWG 72-7938

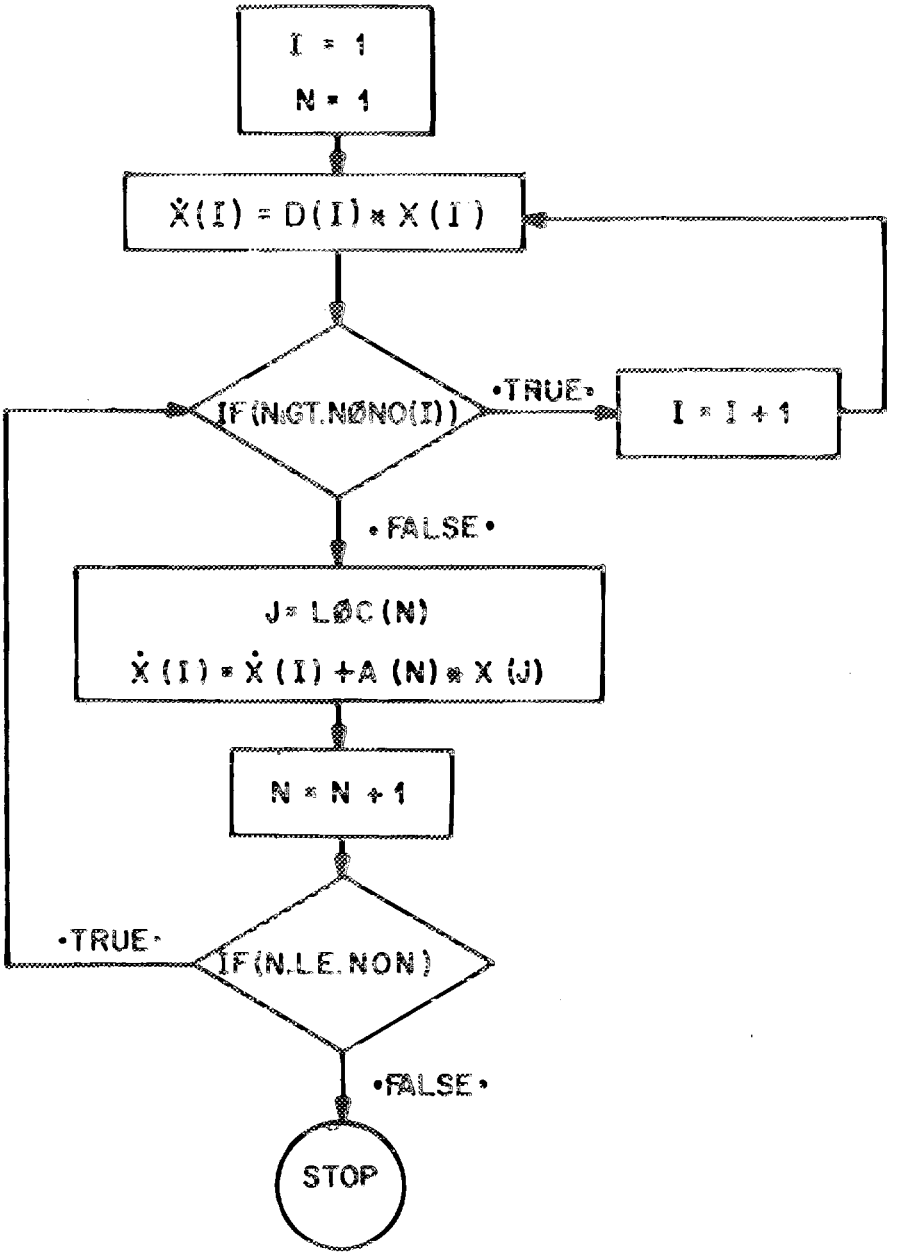

Fig. 3.1. Flowchart illustrating computational algorithm executed to perform the matrix calculation $\dot{\underset{\sim}{X}} = \underset{\approx}{A}\ \underset{\sim}{X}$

Two types of data in the nuclear library require a departure from the procedure just described. In the case of neutron-induced reactions, it is necessary to specify the neutron flux before first-order rate coefficients can be calculated as products of flux and cross sections. At the time the matrix is generated, the neutron flux is unknown. Also, to perform a fuel depletion calculation, the flux must be permitted to vary with time. Thus when the nonzero, off-diagonal matrix elements for isotope I are stored, all those for formation by radioactive decay are grouped first and are followed by those for formation of I by neutron capture. Another vector, KD(I), is also generated and used in a manner analogous to NONO(I). It initially is the number of radioactive parents of isotope I, and the difference NONO(I) - KD(I) represents the number of coefficients for formation of I by neutron capture. The variables A, LOC, NONO, and KD are all generated in subroutines NUDATn. They are used to perform calculations in subroutines FLUXO, DECAY, TERM, and EQUIL.

The second exception to the standard procedure for constructing the transition matrix involves the coefficients corresponding to the fission product yields. The nuclear data library contains direct fission yields for the formation of fission product isotopes from several fissionable nuclides. When these yields are multiplied by the fission cross section for the fissile nuclide and the neutron flux, the result is a first-order rate constant for production of fission product isotope I by fission of nuclide J. Hence for these data, the construction of the arrays NPROD and COEFF and the subsequent search procedure are not required. The coefficients are entered directly into the A vector, and the corresponding value of J that identifies the fissioning nucleus is recorded in the LOC array.

The preceding description summarizes the basic construction of the transition matrix in ORIGEN2. This construction process is very nearly identical to that used in ORIGEN.[1] However, there are three additional features in ORIGEN2 that result in modification of the transition matrix as compared to that in ORIGEN. The first is that provisions have been made in ORIGEN2 to incorporate "nonstandard," flux-dependent reactions. These are neutron capture processes that may be important but which are

significant for only one specific nuclide and therefore are not included in the cross-section library format [e.g., $^{16}O(n,^{3}He)^{14}C$]. Since the solution methods used in ORIGEN only require that the cross section and the parent and daughter nuclides be specified, these are specified separately and incorporated into the transition matrix. Thereafter, these reactions are treated the same as any other flux-dependent reaction.

A second feature of ORIGEN2 that substantially affects the transition matrix is the provision for variable cross sections for certain principal actinide nuclides (see Sect. 2.1.3). Since these values vary during ORIGEN2 irradiation calculations and after the transition matrix has been constructed, it is necessary to modify the appropriate elements of the transition matrix each time the cross sections are altered. This is accomplished by simply storing the locations of the variable cross sections in the transition matrix and altering those elements as required. A similar, but more extensive, problem occurs with the fission products since the variation of the actinide fission cross sections also affects the fission product yield. This effect is accounted for in the same manner as the variation in actinide cross sections: by storing the locations of the fission product yields and adjusting them to compensate for the variation in actinide fission cross sections.

The final feature requiring modification of the transition matrix also occurs after the matrix has been generated. ORIGEN2 has provisions to account for the production of fission products from those actinides which do not have explicit yields in the transition matrix (e.g., ^{237}Np and ^{240}Pu). This is accomplished by calculating the total fission rate in all of the actinides without explicit yields and adjusting the yields for an actinide that has explicit yields (e.g., ^{235}U) to account for these additional fissions. This procedure requires that the explicit fission product yields be adjusted at the beginning of every time interval. This adjustment is accomplished in a manner very similar to that used for accommodating the effect of the variable cross sections on the fission product yields; viz., the locations of the yields in the transition matrix are stored and modified to incorporate the unaccounted-for yields at the beginning of each time interval.

4. DESCRIPTION AND SOURCE OF MISCELLANEOUS ORIGEN2 DATA

This section describes the nature and sources of a wide variety of miscellaneous data associated with ORIGEN2. The types of data described are as follows:

1. neutron yield per spontaneous fission;
2. neutron yield per neutron-induced fission;
3. (α,n) neutron yields;
4. chemical toxicities;
5. the recoverable energy per fission;
6. the methods for calculating values for the ORIGEN2 flux parameters THERM, RES, and FAST;
7. the six-digit ORIGEN2 nuclide and element identifier.

The common basis for describing these data in this report is that (1) they are not associated with a particular reactor or fuel cycle, and (2) they are contained within the ORIGEN2 computer code itself and not in one of the data libraries read by ORIGEN2.

4.1 Neutron Yield per Spontaneous Fission

The calculation of the spontaneous fission neutron emission rate required that the spontaneous fission decay branching ratios and the neutron yields per spontaneous fission be specified. The values of the branching ratios are contained in the updated ORIGEN2 decay library[16] and will not be described here. The spontaneous fission neutron yields are contained in BLOCK DATA in ORIGEN2. The data being used are given in Table 4.1 and are a combination of measured and calculated values.[23,24]

4.2 Neutron Yield per Neutron-Induced Fission

Neutron yields per neutron-induced fission have been incorporated in the BLOCK DATA subroutine of ORIGEN2 for use in calculating the infinite multiplication factor of the materials output by ORIGEN2. These values

were generated by weighing energy-dependent ENDF/B[25,26] neutron yields with multigroup neutron fluxes calculated as a part of the ORIGEN update effort. Two generic sets of neutron yields have been incorporated into ORIGEN2: (1) one for thermal reactors, based on a PWR-U spectrum;[9] and (2) one for fast reactors, based on an advanced oxide, plutonium recycle LMFBR.[11] The two sets of neutron yields are listed in Table 4.2. The ORIGEN2 user can control which of the sets of neutron yields are used with a parameter in the MAIN subroutine. However, it is evident by inspection of the values in Table 4.2 that the thermal and fast neutron yields differ little for the nuclides of significance in most cases. Thus very little error would result if the thermal neutron yields were used.

4.3 Neutron Yields from (α,n) Reactions

The neutrons resulting from (α,n) reactions of light nuclides comprise a second important source of decay-induced neutrons. These neutrons can result from the interaction of energetic alpha particles with a wide variety of light elements such as beryllium and fluorine. In the commercial nuclear fuel cycle, these target materials are seldom encountered in concentrated form, and the principal source of (α,n) neutrons is usually ^{18}O. Since it is impossible for a single set of (α,n) neutron yields to be valid for all cases of a media-dependent reaction, values appropriate to a heavy-metal oxide matrix have been developed and included in ORIGEN.

These (α,n) neutron yields have been incorporated in two ways. First, the yields of seven principal (α,n) contributors are given explicitly in the BLOCK DATA subroutine of ORIGEN2. These values were obtained from ref. 27 and are given in Table 4.3. For those nuclides which generally contribute to a lesser extent, the (α,n) neutron yield is determined using a semiempirical equation that uses the alpha particle energy as the independent variable. The constants in this equation (Table 4.3) were based on the measured (α,n) neutron yields for ^{239}Pu and ^{242}Cm as given in ref. 27. It should be noted that this equation has been significantly altered as compared to the equation in ORIGEN[1] in order to improve the agreement of the equation with measured values. The results predicted

Table 4.1. Neutron yields per spontaneous fission used in ORIGEN2

Nuclide	Neutrons per spontaneous fission	Reference	Nuclide	Neutrons per spontaneous fission	Reference
^{235}U	1.695	2	^{241}Am	2.383	2
^{236}U	1.650	2	^{242}Am	2.475	2
^{237}U	1.872	2	^{242m}Am	2.590	2
^{238}U	2.000	2	^{243}Am	2.520	2
^{239}U	2.048	2	^{244}Am	2.657	2
^{236}Np	1.783	2	^{244m}Am	2.665	2
^{236m}Np	1.790	2	^{241}Cm	2.500	2
^{237}Np	1.873	2	^{242}Cm	2.590	2
^{238}Np	1.963	2	^{243}Cm	2.687	2
^{239}Np	2.053	2	^{244}Cm	2.760	2
^{236}Pu	2.220	2	^{245}Cm	2.872	2
^{237}Pu	1.886	2	^{246}Cm	3.000	2
^{238}Pu	2.280	2	^{248}Cm	3.320	3
^{239}Pu	2.240	2	^{250}Cm	3.560	3
^{240}Pu	2.160	2	^{249}Bk	3.720	3
^{241}Pu	2.250	2	^{249}Cf	3.440	3
^{242}Pu	2.150	2	^{250}Cf	3.560	3
^{243}Pu	2.430	2	^{252}Cf	3.725	3
^{244}Pu	2.300	2	^{254}Cf	3.900	3
^{240}Am	2.290	2	^{253}Es	3.920	3

Table 4.2. Spectrum-averaged neutron yields per neutron-induced fission ($\bar{\nu}$)

Nuclide	PWR-U	LMFBR	Nuclide	PWR-U	LMFBR
^{232}Th	2.418	2.396	^{242m}Am	3.162	3.311
^{233}Pa	2.663	2.631	^{243}Am	3.732	3.653
^{233}U	2.499	2.520	^{242}Cm	3.746	3.868
^{234}U	2.631	2.555	^{243}Cm	3.434	3.496
^{235}U	2.421	2.468	^{244}Cm	3.725	3.743
^{236}U	2.734	2.614	^{245}Cm	3.832	3.898
^{238}U	2.807	2.776	^{246}Cm	3.858	3.870
^{237}Np	3.005	2.935	^{247}Cm	3.592	3.680
^{236}Pu	2.870	2.946	^{248}Cm	3.796	3.866
^{238}Pu	2.833	3.009	^{249}Bk	3.760	3.671
^{239}Pu	2.875	2.946	^{249}Cf	4.062	4.130
^{240}Pu	3.135	3.024	^{250}Cf	3.970	3.813
^{241}Pu	2.934	2.978	^{251}Cf	4.140	4.227
^{242}Pu	3.280	3.075	^{252}Cf	4.126	4.364
^{241}Am	3.277	3.402	^{253}Cf	4.150	4.151
^{242}Am	3.360	3.361			

Table 4.3. Neutron yields from (α,n) reactions

Nuclide	Neutron yield (neutrons sec^{-1} g^{-1})
^{238}Pu	2.00×10^{4}
^{239}Pu	45
^{240}Pu	170
^{242}Pu	2.7
^{241}Am	4.00×10^{3}
^{242}Cm	2.67×10^{7}
^{244}Cm	5.72×10^{5}
All others	2.152×10^{-18} $(E)^{14.01}$, where E = alpha particle energy, MeV

by the equation in Table 4.3 agree well with experimental values for alpha particles in the 5.0 to 6.2 MeV range, which is of interest in most commonly encountered situations. For light nuclide targets other than ^{18}O or for alpha particle energies significantly outside the 5 to 6 MeV range, a different set of equation constants must be derived and inserted in the ORIGEN2 BLOCK DATA subroutine.

4.4 Elemental Chemical Toxicities

Elemental chemical toxicity values have been incorporated into the ORIGEN2 BLOCK DATA subroutine. The values, which were obtained from ref. 28 and are listed in Table 4.4, are given in units of maximum desired ambient concentration in water (i.e., g/m^3). These values are used to calculate a measure of the chemical toxicity of an element

Table 4.4. Elemental chemical toxicities incorporated in ORIGEN2

Element	Toxicity (g/m^3)	Element	Toxicity (g/m^3)	Element	Toxicity (g/m^3)
H	3500	Se	0.01	Ho	1.0
He	0.2	Br	3.0	Er	0.1
Li	5.0	Kr	40	Tm	0.2
Be	1.0	Rb	50	Yb	0.1
B	1.0	Sr	10	Lu	0.1
C	400	Y	0.001	Hf	0.05
N	0.01	Zr	1.0	Ta	1.0
O	945,500	Nb	0.02	W	100
F	1.0	Mo	0.5	Re	10
Ne	1.0	Tc	100	Os	1.0
Na	1000	Ru	1.0	Ir	0.8
Mg	10	Rh	0.05	Pt	0.3
Al	0.01	Pd	0.05	Au	0.02
Si	5.0	Ag	0.001	Hg	0.002
P	0.01	Cd	0.01	Tl	0.005
S	50.0	In	0.02	Pb	0.01
Cl	0.15	Sn	0.05	Bi	0.1
Ar	10	Sb	0.05	Po	0.2
K	1000	Te	0.2	At	10
Ca	30	I	10	Rn	500
Sc	0.5	Xe	150	Fr	5.0
Ti	0.1	Cs	5.0	Ra	0.001
U	0.1	Ba	0.5	Ac	0.02
Cr	0.02	La	1.0	Th	0.0005
Mn	0.01	Ce	2.0	Pa	0.005
Fe	0.05	Pr	1.0	U	0.5
Co	0.05	Nd	0.2	Np	0.003
Ni	0.05	Pm	1.0	Pu	0.0008
Cu	0.01	Sm	0.2	Am	0.04
Zn	0.05	Eu	0.2	Cm	0.5
Ga	0.2	Gd	0.2	Bk	0.005
Ge	0.5	Tb	0.5	Cf	0.01
As	0.01	Dy	1.0	Es	0.01

mixture by dividing the mass of each element by its chemical toxicity and summing the resulting water volumes. The total water volume thus calculated is the volume required to dilute the mixture to desired ambient levels and can therefore serve as a measure of the toxicity of the mixture.

4.5 Nuclide-Dependent Recoverable Energy per Fission

One of the principal features of ORIGEN2 is that the cross sections it uses are calculated by more sophisticated reactor physics codes and thus are "actual" cross sections. If the neutron flux (or specific power) calculated from these cross sections is to be accurate, then the other parameters used in the calculation must also represent reality. The two other parameters involved in the calculation (see Sect. 3.4) are the concentrations of the various fissile nuclides and the recoverable energy per fission. The fissile nuclide concentration (in g-atoms per unit of fuel) has always been on a "real" basis and need not be considered further. However, in the past the recoverable energy per fission was assumed to be 200 MeV per fission for all fissile nuclides in ORIGEN.[1] This assumption worked well because the fission cross sections being used at that time had been adjusted to account for the difference between the 200 MeV value and the actual value. However, since the cross sections being used in ORIGEN2 are not adjusted, it became necessary to incorporate realistic values of the recoverable energy per fission into ORIGEN2.

As a result of the large number of actinides in ORIGEN2 and the lack of recoverable fission energy data on many of them, it was decided to use a semiempirical equation to calculate the recoverable energy per fission. The form of the selected equation is $REPF = C_1(Z^2A^{0.5}) + C_2$, where Z is the nuclide's atomic number, A is the nuclide's atomic weight, C_1 and C_2 are constants, and REPF is the recoverable energy per fission in MeV per fission. Equations of this general form are widely used to correlate fission product kinetic energy,[29,30] although most have the exponent of the atomic weight as 0.33 instead of 0.5. However, work by Okolovich et al.[31] and the results of comparing the general form given above with evaluated data indicate that the 0.5 exponent gives slightly better results.

The constants in the general equation were determined by using the REPF values for ^{235}U and ^{239}Pu as given in ref. 29. The resulting equation, which has been incorporated in ORIGEN2 subroutine FLUXO, is

$$REPF(MeV/fission) = 1.29927 \times 10^{-3} \ (Z^2 A^{0.5}) + 33.12.$$

This equation predicts REPF values with a maximum error of 1% for nuclides between ^{232}Th and ^{242}Pu as compared to the evaluated data given in ref. 29.

4.6 ORIGEN2 Flux Parameters THERM, RES, and FAST

The flux parameters THERM, RES, and FAST are needed to allow thermal cross sections, resonance integrals, and fission-spectrum-averaged threshold cross sections to be incorporated into ORIGEN2. This feature is highly desirable since multigroup cross sections are only available for about 200 of the 1300 distinct nuclides considered in ORIGEN2 and the only other source of cross-section information is compilations of standard cross sections such as BNL-325.[32] Values for these flux parameters were originally developed for ORIGEN[1] based on the Westcott[33] formalisms and generic flux shapes for thermal reactors. However, modern reactor physics codes supply sufficient information to allow these parameters to be derived from the calculated neutron spectrum instead of a generic spectrum. It is the purpose of this section to derive the recipes for calculating these parameters from information available in the output of a code that calculates a static neutron energy spectrum and spectrum-averaged neutron cross sections.

Before launching into the details of the derivation, it is necessary for the reader to understand the neutron energy groups of interest. These groups are depicted schematically in Fig. 4.1 along with the variables associated with each group. The variables will be defined in detail when they are used in the derivations. The diagram in Fig. 4.1 shows four energy groups which are comprised of two pairs. In addition, there is a single, all-encompassing group characterized by the total flux and the total spectrum-averaged cross section.

ORNL DWG 79-1368

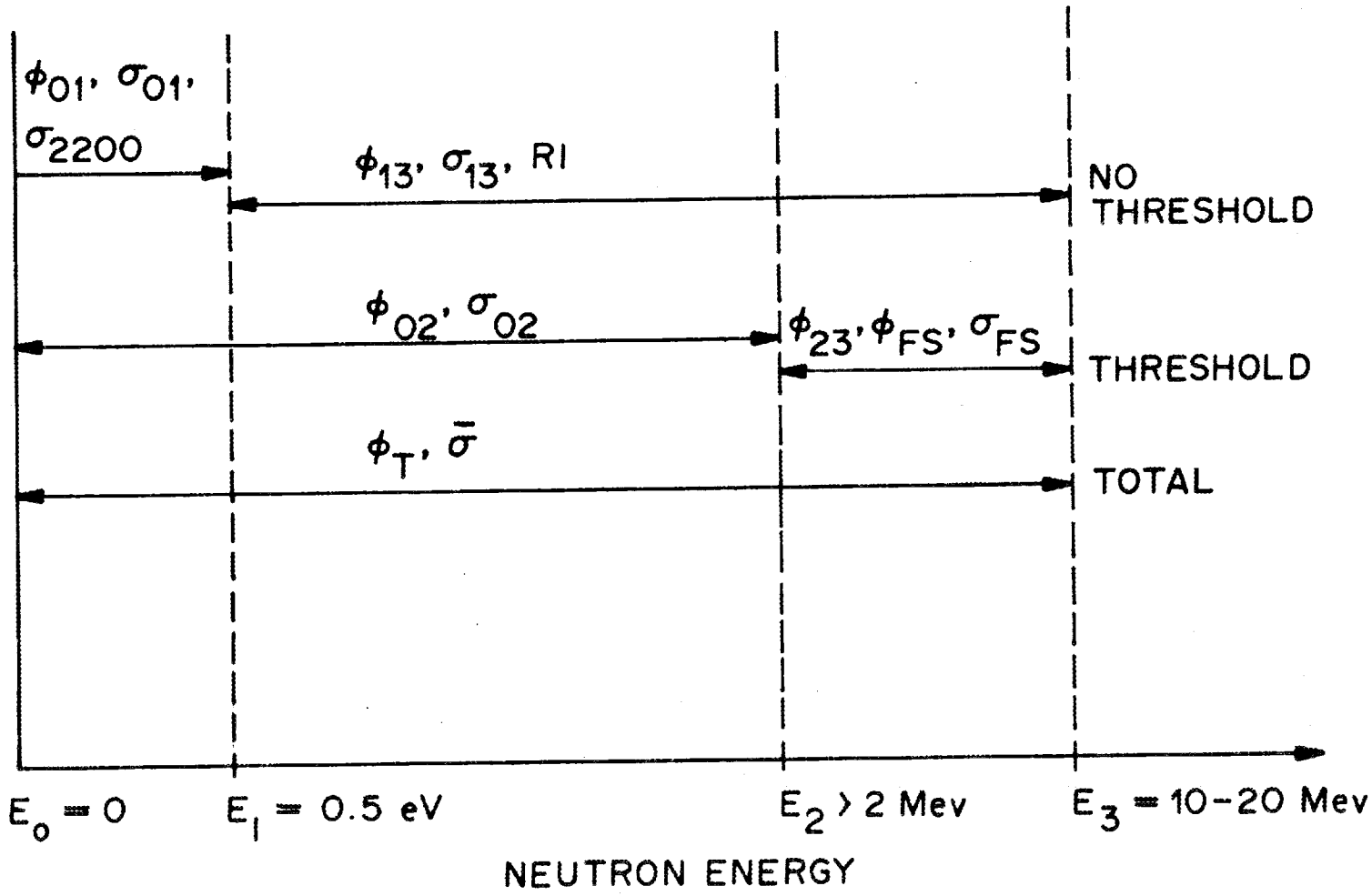

Fig. 4.1. Schematic diagram of energy group structure for the calculation of ORIGEN2 flux parameters.

The ORIGEN2 flux parameters are constants applicable to a specific reactor-fuel combination. The "standard" cross sections (e.g., infinite dilution resonance integral), when multiplied by the appropriate flux parameter, yield an average cross section which can be appropriately multiplied by the total flux to yield the correct nuclide reaction rate. This procedure is necessary because the flux-parameter-averaged cross sections are stored in the ORIGEN2 transition matrix in the same manner as the multigroup-spectrum-averaged cross sections and thus must be compatible with them.

4.6.1 Derivation of ORIGEN2 flux parameters THERM and RES

The basic approach used in deriving expressions for calculating THERM and RES is to write two reaction rate balances for an unspecified nuclide: one in terms of a thermal cross section and resonance integral and the other in terms of averaged cross sections. For this derivation, the top pair of energy groups and the single-group (bottom) in Fig. 4.1 will be used.

The neutron-induced reaction rate of an unspecified nuclide can be written in two ways:

$$\phi_T \bar{\bar{\phi}} = C_1 \, \phi_{01} \, \phi_{2200} + C_2 \, \phi_{13} RI \tag{1a}$$

or

$$\phi_T \bar{\sigma} = \phi_{01} \, \phi_{01} + \phi_{13} \, \phi_{13} \, , \tag{1b}$$

where

ϕ_T = Total flux from a reactor physics calculation, neutrons barn^{-1} s^{-1}.

ϕ_{01} = Thermal flux (0 to 0.5 eV) from a reactor physics calculation, neutrons barn^{-1} s^{-1}.

ϕ_{13} = Resonance/fast flux (0.5 eV to maximum) from reactor physics calculation, neutrons barn^{-1} s^{-1}.

ϕ_{2200} = Thermal (22 m/s) cross section from source such as ref. 32, barns.

RI = Infinite dilution resonance integral (0.5 eV to infinity) from source such as ref. 32, barns.

σ_{01} = Effective cross section between E_0 and E_1 (i.e., between 0 and 0.5 eV), from a reactor physics calculation, barns.

σ_{13} = Effective cross section between E_1 and E_3 (i.e., between 0.5 eV and maximum) from reactor physics calculation, barns.

$\bar{\sigma}$ = Effective cross section between E_0 and E_3 (i.e., between 0 and the maximum) from reactor physics calculation, barns.

C_1, C_2 = Constants accounting for effects within energy groups.

Dividing Eqs. (1a) and (1b) by the total flux ϕ_T yields

$$\bar{\sigma} = \frac{C_1 * \phi_{01} * \sigma_{2200}}{\phi_T} + \frac{C_2 * \phi_{13} * RI}{\phi_T} \tag{2a}$$

and

$$\bar{\sigma} = \frac{\phi_{01} * \sigma_{01}}{\phi_T} + \frac{\phi_{13} * \sigma_{13}}{\phi_T} \quad . \tag{2b}$$

The ORIGEN2 flux parameters THERM and RES are now defined as

$$\text{THERM} \equiv \frac{C_1 * \phi_{01}}{\phi_T} \tag{3a}$$

and

$$\text{RES} \equiv \frac{C_2 * \phi_{13}}{\phi_T} \quad . \tag{3b}$$

Using these definitions Eqs. (2a) and (2b) can be rewritten as

$$\bar{\sigma} = \text{THERM} * \sigma_{2200} + \text{RES} * \text{RI} \tag{4a}$$

and

$$\bar{\sigma} = \frac{\phi_{01} * \sigma_{01}}{\phi_T} + \frac{\phi_{13} * \sigma_{13}}{\phi_T} \quad . \tag{4b}$$

The final step in the derivation is to equate the first terms of Eqs. (4a) and (4b) with each other and the second terms with each other and solve for the flux parameters. In effect, this procedure equates the two expressions giving the effective cross section in the thermal range (E_0 to E_1) with each other and similarly for the resonance/fast neutron group. The result of this procedure is

$$\text{THERM} = \frac{\phi_{01}{}^{*}\sigma_{01}}{\phi^{*}\sigma_{2200}} \quad ; \tag{5a}$$

$$\text{RES} = \frac{\phi_{13}{}^{*}\sigma_{13}}{\phi_{T}{}^{*}\text{RI}} \quad . \tag{5b}$$

All of the parameters on the right-hand sides of Eqs. (5a) and (5b) are obtainable from either a reactor physics (i.e., static neutron spectrum) calculation with the output in two groups with a 0.5 eV cutoff or from standard literature references such as ref. 32.

The only difficulty that arises is in choosing which nuclide to use in the calculation since the varying shapes and resonance structures of different nuclides will alter the calculated values for the flux parameters. The solution to this problem was to use an artificial cross section that varies inversely with neutron velocity (i.e., inversely with the square root of neutron energy), which is the theoretical variation for all nuclides. This nuclide's thermal cross section is defined as 1.0 barn and the resonance integral is 0.45 barn. By spectrum-averaging the multigroup representation of this cross section so that the resulting cross sections and fluxes are in two groups, values of THERM and RES can readily be calculated. This is the procedure that has been followed in previously issued reports[9,10] describing updated ORIGEN2 reactor models.

4.6.2 Derivation of the ORIGEN2 flux parameter FAST

The derivation of an expression for calculating the ORIGEN2 flux parameter FAST is similar to that for THERM and RES. The energy groups used in this derivation are the middle pair and the single group (bottom)

in Fig. 4.1. However, it is possible to simplify the derivation by noting that the parameter FAST is only applicable to nuclear reactions with a threshold [i.e., the reaction cross section is effectively zero below a few MeV, such as (n,2n) and (n,3n)]. Thus the term for the low-energy reaction rate is always zero because the cross section is zero.

The neutron-induced reaction rate of an unspecified nuclide can be written in either of two ways:

$$\phi_T \bar{\sigma} = C_3 \, \phi_{23} \, \sigma_{FS} \tag{6a}$$

or

$$\phi_T \bar{\sigma} = \phi_{FS} \, \sigma_{FS} \quad , \tag{6b}$$

where

ϕ_{23} = fast neutron flux (from about 100 keV to maximum) from a reactor physics calculation, neutrons barn^{-1} s^{-1} ;

σ_{FS} = threshold cross section averaged over a fission neutron spectrum, neutrons s^{-1};

C_3 = constant accounting for effects within the fast energy group;

ϕ_{FS} = fission spectrum neutron flux (from about 100 keV to maximum) based on a reactor physics calculation, neutrons barn^{-1} s^{-1}.

and the other variables are as previously defined. An expression for calculating the flux parameter FAST is derived by (1) dividing such equation by ϕ_T, (2) defining FAST $\equiv C_3 * \phi_{23}/\phi_T$, and (3) equating the right-hand sides of these two equations. The result is:

$$\text{FAST} = \phi_{FS}/\phi_T \quad , \tag{8}$$

which is a relatively simple expression and is free of the nuclide-dependence characterizing the expressions for THERM and RES. There is, however, one difficulty with this equation: the flux ϕ_{FS} is not directly obtainable from a reactor physics calculation over the desired range (i.e., about 100 keV to 10 MeV) because the portion of the fission neutron flux below the peak value (about 2 MeV) is distorted due to high-energy

scattering processes. The solution used to avoid this problem is to obtain a partial fission neutron flux from the region above the fission neutron flux peak (i.e., the undistorted region) and divide it by a constant factor to convert it into the full fission neutron flux. If ϕ_{FS}' is defined as the undistorted fission neutron flux and f is defined as the constant ϕ_{FS}'/ϕ_{FS} (obtainable from sources such as ref. 34, the resulting expression for calculating FAST is then:

$$\text{FAST} = \phi_{FS}'/(f*\phi_T) \quad . \tag{9}$$

For example, if ϕ_{FS}' is defined as the fission neutron flux above 2.5 MeV, this value would correspond to 0.2927 of the total number of fission neutrons. Thus, the appropriate value of f is 0.2927. It should be noted that the exact energy used in this calculation is not important as long as the fission neutron spectrum is undistorted and the corresponding value of f is used.

4.6.3 Conversion of ORIGEN2 flux parameters to ORIGEN flux parameters

As is evident from the derivations in Sects. 4.6.1 and 4.6.2, the ORIGEN2 flux parameters are on a total flux (i.e., summed from 0 to maximum energy) basis. However, previous versions of ORIGEN[1] have generally been on a thermal flux basis for thermal reactors (e.g., PWRs). Thus, it may be desirable to convert the total-flux-bases parameters so older versions of ORIGEN can take advantage of more recent data.

The most straightforward method for making this change is to multiply the total-flux-based parameters by the ratio of the total flux to the thermal flux; i.e., by ϕ_T/ϕ_{01} [see under Eqs. (1a) and (1b) for more precise definitions]. However, in many instances the requisite reactor physics information for generating this conversion factor are not readily obtainable. In this case, the thermal-flux-based value of THERM can be calculated using the expression derived from the Westcott[33] formalism:

$$\text{THERM (thermal)} = \sqrt{\frac{\pi*293}{4*T}} \quad ,$$

where T is the effective moderator temperature in K. The conversion factor for the other flux parameters is then simply the ratio of THERM on a thermal-flux-basis to THERM calculated on a total-flux basis. Values for the ORIGEN flux parameters on a thermal-flux basis will be give in all of the reports describing updated models ORIGEN2 reactor models.

4.7 ORIGEN2 Nuclide and Element Identifiers

Most of the input and internal operations done in ORIGEN2 are based on the use of a six-digit integer nuclide or element identifier. The nuclide identifier is defined as

$$NUCLID = 10{,}000*Z + 10*A + M \quad ,$$

where

NUCLID = six-digit nuclide identifier,
Z = atomic number of nuclide (1 to 99),
A = atomic mass of nuclide (integer),
M = state indicator; 0 = ground state; 1 = excited state.

It should be noted that only one excited state is allowed in ORIGEN2, as with the previous versions of ORIGEN. The principal functions of the NUCLID are to identify the data associated with it in the input libraries and internally in ORIGEN2 and to permit construction of the transition matrix using integer operations (see Sect. 3.5). It is also used to supply the atomic weight for converting g-atoms to grams. ORIGEN2 contains a single psuedonuclide, ^{250}Sf, which is used to collect the fission product mass resulting from spontaneous fission.

The six-digit identifier for an element follows the pattern set by the nuclide identifier

$$NELID = 10{,}000*Z \ ,$$

where NELID is the element identifier and Z is as described previously.

5. REFERENCES

1. M. J. Bell, ORIGEN - The ORNL Isotope Generation and Depletion Code, ORNL-4628 (May 1973).
2. C. M. Lederer, J. M. Hollander, and S. Perlman, Table of Isotopes, 6th ed., Wiley, New York, 1967.
3. J. R. Stehn, M. D. Goldberg, B. A. Magurna, and R. Weiner-Chasman, Neutron Cross Sections, 2nd ed., Suppl. 2, BNL-325 (1964).
4. M. K. Drake, A "Compilation of Resonance Integrals," Nucleonics, 24(8), 108-111 (August 1966).
5. M. E. Meek and B. F. Rider, Summary of Fission Product Yields for ^{235}U, ^{238}U, ^{239}Pu, and ^{241}Pu at Thermal, Fission Spectrum, and 14 MeV Neutron Energies, APED-5398-A (Rev.) (1968).
6. C. W. Kee, A Revised Light Element Library for the ORIGEN Code, ORNL/TM-4896 (May 1975).
7. C. W. Kee, C. R. Weisbin, and R. E. Schenter, Processing and Testing of ENDF/B-IV Fission Product and Transmutation Data, Trans. Am. Nucl. Soc. 19, 398-99 (1974).
8. A. G. Croff, A User's Manual for the ORIGEN2 Computer Code, ORNL/TM-7175 (in preparation).
9. A. G. Croff, M. A. Bjerke, G. W. Morrison, and L. M. Petrie, Revised Uranium-Plutonium Cycle PWR and BWR Models for the ORIGEN Computer Code, ORNL/TM-6051 (September 1978).
10. A. G. Croff and M. A. Bjerke, Alternative Fuel Cycle PWR Models for the ORIGEN Computer Code, ORNL/TM-7005 (February 1980).
11. A. G. Croff and J. W. McAdoo, LMFBR Models for the ORIGEN2 Computer Code, ORNL/TM-7176 (in preparation).
12. A. G. Croff and M. A. Bjerke, CANDU Models for the ORIGEN Computer Code, ORNL/TM-7177 (in preparation).
13. W. B. Ewbank, M. R. Schmorak, F. E. Bertrand, M. Feliciano, and D. J. Horen, Nuclear Structure Data File: A Manual for Preparation of Data Sets, ORNL-5054 (June 1975).
14. W. B. Ewbank, "Evaluated Nuclear Structure Data File (ENSDF) for Basic and Applied Research," paper presented at the Fifth International CODATA Conference, Boulder, Colo., June 1976.

15. Nuclear Data Sheets, published by Academic Press, Inc. Subscription information available on request to Academis Press, Inc., 111 Fifth Ave., New York, N.Y. 10013.

16. A. G. Croff, R. L. Haese, and N. B. Gove, Updated Decay and Photon Libraries for the ORIGEN Code, ORNL/TM-6055 (February 1979).

17. B. H. Duane, in Physics Research Quarterly Report, Oct.-Dec. 1963, HW-80020 (1964).

18. H. E. Krug, J. E. Olhoeft, and J. Alsina, Chap. 8 in Supplementary Report on Evaluation of Mass Spectrometric and Radiochemical Analyses of Yankee Cor I Spent Fuel, Including Isotopes of Elements Thorium through Curium, WCAP-6086 (August 1969).

19. L. Lapidus and R. Luus, Optimal Control of Engineering Processes, pp. 45-49, Bleisdell Publishing Co., Waltham, Mass., 1967.

20. S. J. Ball and R. K. Adams, MATEXP, a General Purpose Digital Computer Program for Solving Ordinary Differential Equations by the Matrix Exponential Method, ORNL/TM-1933 (August 1967).

21. D. R. Vondy, Development of a General Method of Explicit Solution to the Nuclide Chain Equations for Digital Machine Calculations, ORNL/TM-361 (Oct. 17, 1962).

22. L. Lapidus, Digital Computation for Chemical Engineers, pp. 259-62, McGraw-Hill, New York, 1962.

23. S. Raman, "General Survey of Applications Which Require Actinide Nuclear Data," presented at the IAEA Advisory Group Meeting on Transactinium Nuclear Data, Karlsruhe, FRG (November 1975).

24. L. J. King, J. E. Bigelow, and E. D. Collins, Transuranium Processing Plant Semiannual Report of Production Status, and Plans for the Period Ending June 30, 1973 (March 1974).

25. ENDF/B-IV Library Tapes 401-411 and 414-419, available from the National Neutron Cross Section Center, Brookhaven National Laboratory (December 1974).

26. ENDF/B-V Library Tapes 521 and 522, available from the National Neutron Cross Section Center, Brookhaven National Laboratory (July 1979).

27. D. L. Johnson, "Subcritical Reactivity Monitoring: Neutron Yields from (α,n) Reactions in FTR Fuel," section in Core Engineering Technical Progress Report April, May, June 1976, HEDL-TME 76-36 (March 1977).

28. G. W. Dawson, The Chemical Toxicity of Elements, BNWL-1815 (June 1974).

29. J. P. Unik and J. E. Gindler, A Critical Review of the Energy Released in Nuclear Fission, ANL-7748 (March 1971).

30. V. E. Viola, Jr., "Correlation of Fission Fragment Kinetic Energy Data," Nuclear Data 1(5), pp. 391-410 (July 1966).

31. V. N. Okolovich, V. I. Bol'shov, L. D. Gordeeva, and G. N. Smirenkin, "Dependence of the Mean Kinetic Energy of Fragments on the Fissionable Nucleus Mass," Soviet Atomic Energy 15(5), pp. 1177-78 (November 1963).

32. S. F. Mughabghab and D. I. Garber, Neutron Cross Sections, Volume 1, Resonance Parameters, BNL-325, 3d ed. (June 1973).

33. C. H. Westcott, Effective Cross Section Values for Well-Moderated Thermal Reactor Spectra, CRRP-960, 3d ed. corrected (November 1960).

34. H. Etherington, Nuclear Engineering Handbook, 1st ed., pp. 7-91, McGraw-Hill, New York (1958).

ORNL-5621
Dist. Category UC-70

INTERNAL DISTRIBUTION

1. C. W. Alexander
2. E. J. Allen
3. W. B. Arthur
4. D. E. Bartine
5. M. A. Bjerke
6-10. J. O. Blomeke
11. E. S. Bomar
12. W. D. Burch
13. D. O. Campbell
14. J. C. Cleveland
15. W. H. Corcoran (consultant)
16-20. A. G. Croff
21. W. Davis, Jr.
22-26. D. E. Ferguson
27. G. F. Flanagan
28. C. W. Forsberg
29. E. J. Frederick
30. R. W. Glass
31. M. J. Haire
32. F. E. Harrington
33. O. W. Hermann
34. P. R. Kasten
35. C. W. Kee
36. L. J. King
37. R. S. Lowrie
38. F. C. Maienschein
39-40. B. F. Maskewitz
41. A. D. Mitchell
42. G. W. Morrison
43. E. Newman
44. K. J. Notz
45. L. M. Petrie
46-50. C. H. Shappert
51. L. B. Shappert
52. H. F. Soard
53. J. W. Snider
54. M. G. Stewart
55. S. N. Storch
56. W. E. Thomas
57. V. C. A. Vaughen
58. C. R. Weisbin
59. Radiation Shielding Information Center
60-61. Central Research Library
62. ORNL Patent Section
63-64. Laboratory Records
65. Laboratory Records, ORNL RC
66. ORNL-Y-12 Technical Library Document Reference Section

EXTERNAL DISTRIBUTION

DOE-ORO, Oak Ridge, TN 37830
67. Office of Assistant Manager for Energy Research and Development
68. D. E. Large

DOE, Washington, DC 20545
69. G. K. Oertel
70. R. D. Walton

Battelle Pacific Northwest Laboratory, P.O. Box 999, Richland, WA 99352
71. A. M. Platt
72. E. J. Merrill

Argonne National Laboratory, 9700 South Cass Ave., Argonne, IL 62439
73. Y. I. Chang
74. M. Steindler

Sandia Laboratories, P.O. Box 5800, Albuquerque, NM 87115
75. S. A. Dupree

E. I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, SC 29801
76. R. W. Benjamin

Bechtel National, Inc., P.O. Box 3965, San Francisco, CA 94119
77. G. H. Dyer

Los Alamos Scientific Laboratory, Group T-2, Mail Stop 243, Los Alamos, NM 87545
78. T. R. England

Los Alamos Technical Associates, P.O. Box 410, 1553 Myrtle St., Los Alamos, NM 87544
79. S. E. Logan

General Electric, Mail Code S-34, P.O. Box 5020, Sunnyvale, CA 94086
80. S. L. Beaman

Battelle Columbus Laboratories, Office of Nuclear Waste Isolation, 505 King Ave., Columbus, OH 43201
81. J. Waddell

Hanford Engineering Development Laboratory, P.O. Box 1970, Richland, WA 99352
82. F. M. Mann
83. F. Schmittroth

Westinghouse Electric, Waltz Mill Site, P.O. Box 158, Madison, PA 15663
84. C. McGinnis

Allied-Gulf Nuclear Services, P.O. Box 847, Barnwell, SC 29812
85. W. H. Carr

Science Applications, Inc., 5 Palo Alto Square, Suite 200, Palo Alto, CA 94304
86. R. R. Fullwood

Atomic Energy Research Establishment, Harwell, Didcot, Oxon., OX11 ORA, England
87. J. R. Grover

National Radiation Protection Board, Harwell, Didcot, Oxon., OX11 ORA, England
88. G. N. Kelley

Centre d'études nucléaiers de Fontenay-aux-Roses, B. P. No. 6, Fontenay-aux-Roses, France
89. Y. Sousselier

OECD-Nuclear Energy Agency, 38, Boulevard Suchet, F-75016 Paris, France
90. M. Takahashi

International Atomic Energy Agency, P.O. Box 590, A-1011 Vienna, Austria
91. E. R. Irish

Lab. Teoria e Calcolo Reottori, C.N.E.N.-C.S.N. Casaccia, CP 2400, I-00100 Rome, Italy
92. L. Tondinelli

Institut für Heisse Chemie Kernforschungszentrum, Postfach 3640, 75 Karlsruhe, Germany
93. H. O. Haug

NUKEM GmbH, Postfach 110080, D-6450 Hanau 11, West Germany
94. Michael Weber

Commission of the European Communities, CCR Euratom, I-21200 Ispra, Italy
95. F. Girardi

Chalmers University of Technology, Dept. of Nuclear Chemistry, S-402 20 Göteborg, Sweden
96. J. O. Liljenzin

CCR Euratom, I-21200, Ispra, Italy
97. W. Hage

Studsvik, AB Atomenergi, Fack, S-611 01, Nyköping 1, Sweden
98. E. Hellstrand

Cadarach, B. P. No. 1, 13115 Sant-Paul-lez-Durance, France
99. J. Bouchard

100-423. Given distribution as shown in TID-4500 under Nuclear Waste Management category (NTIS - 25 copies)

☆U.S. GOVERNMENT PRINTING OFFICE: 1980-640-245/167