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## Perturbation and Sensitivity Theory for Reactor Burnup Analysis

M. L. Williams

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PERTURBATION AND SENSITIVITY THEORY FOR REACTOR BURNUP ANALYSIS\*

M. L. Williams

Date Published: December 1979

\*Submitted to The University of Tennessee as a doctoral dissertation  
in the Department of Nuclear Engineering.

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## ABSTRACT

Perturbation theory is developed for the nonlinear burnup equations describing the time-dependent behavior of the neutron and nuclide fields in a reactor core. General aspects of adjoint equations for nonlinear systems are first discussed and then various approximations to the burnup equations are rigorously derived and their areas for application presented. In particular, the concept of coupled neutron/nuclide fields (in which perturbations in either the neutron or nuclide field are allowed to influence the behavior of the other field) is contrasted to the uncoupled approximation (in which the fields may be perturbed independently).

Adjoint equations are derived for each formulation of the burnup equations, with special attention given to the quasi-static approximation, the method employed by most space- and energy-dependent burnup codes. It is shown that, based on this formulation, three adjoint equations (for the flux shape, the flux normalization, and the nuclide densities) are required to account for coupled variations in the neutron and nuclide fields. The adjoint equations are derived in detail using a variational principle. The relation between coupled and uncoupled depletion perturbation theory is illustrated.

Solution algorithms are given for numerically solving the adjoint burnup equations, and the implementation of these procedures into existing computer codes is discussed. A physical interpretation is given for the burnup adjoint functions, which leads to a generalization of the principle

of "conservation of importance" for coupled fields. Analytic example problems are solved to illustrate properties of the adjoint functions.

Perturbation theory is used to define sensitivity coefficients for burnup-dependent responses. Specific sensitivity coefficients are written for different types of nuclear data and for the initial condition of the nuclide field. Equations are presented for uncertainty analysis of burnup calculations.

Uncoupled depletion sensitivity theory is applied to the analysis of an irradiation experiment being used to evaluate new actinide cross-section data. The computed sensitivity coefficients are used to determine the sensitivity of various nuclide concentrations in the irradiated sample to actinide cross sections. Uncertainty analysis is used to calculate the standard deviation in the computed values for the plutonium isotopics.

Coupled depletion sensitivity theory is used to analyze a 3000 MW<sub>th</sub> denatured LMFBR model (2 region, sphere). The changes in the final inventories of <sup>232</sup>U, <sup>233</sup>U, and <sup>239</sup>Pu due to changes in concentrations of several nuclides at the beginning of cycle are predicted using depletion perturbation theory and are compared with direct calculation. In all cases the perturbation results show excellent agreement with the direct changes.

## CHAPTER I

### INTRODUCTION AND BACKGROUND

The area of nuclear engineering known as burnup analysis is concerned with predicting the long-term isotopic changes in the material composition of a reactor. Analysis of this type is essential in order to determine optimum fissile loading, efficient refueling schedules, and a variety of operational characteristics that must be known to ensure safe and economic reactor performance. Burnup physics is unique in that it is concerned not only with computing values for the neutron flux field within a reactor region, but also with computing the time-dependent behavior of the nuclide-density field. In general the flux and nuclide fields are coupled nonlinearly, and solving the so-called burnup equations is quite a formidable task which must be approached with approximate techniques.

It is the goal of this study to develop a perturbation theory for application to burnup analysis. Based on such a technique, a sensitivity methodology will be established which seeks to estimate the change in various computed quantities when the input parameters to the burnup calculation are varied. A method of this type can be a useful analysis tool, applicable to several areas of practical interest. Two of the important areas are (a) in assessing the sensitivity of computed parameters to data uncertainties, and (b) in determining the effect of design changes at beginning-of-life on a parameter evaluated at some time in the future.

Sensitivity analysis at Oak Ridge National Laboratory (ORNL) (1, 2, 3) and elsewhere (4, 5, 6) has flourished both theoretically and computationally during the last several years, culminating in recent uncertainty estimates (7) for performance parameters of large LMFBR reactors, including both differential and integral information. Current work, however, has been focused largely on the time-independent problem for functionals of the neutron flux. Much of the advance in this area can be attributed to the development of "generalized perturbation theory" (GPT) for eigenvalue equations put forth by Usachev (8), Gandini (9), Pomraning (10) and others during the 1960's, although groundwork for the theory was actually developed by Lewins (11) in the late 1950's. Essentially GPT extended the application of "normal perturbation theory" (for  $k_{\text{eff}}$ ) to include analysis of any arbitrary ratio of functionals linear or bilinear in the flux and/or adjoint flux.

It is interesting to note that even though nearly all the applied perturbation theory work of the last decade has focused on the time-independent neutron transport equation, much of the early work in adjoint theory was concerned with the time-*dependent* case. For example, the classic book by Weinberg and Wigner (12) talks about the effect on future generations of introducing a neutron into a critical reactor, although ultimately the effect is related back to a static eigenvalue. The important work by Lewins in 1960 is the first that really dwells in detail on adjoint equations for the time-dependent reactor kinetics equations (13). In that work the concept "time-dependent neutron

importance" is clearly quantified and pointed the way for future developments based on the importance principle. At about this same time (early 1960's) Lewins published another important paper which is related to work presented in this thesis. In that work he derived adjoint equations for a nonlinear system (14). However, his work was somewhat academic in that it did not address any specific equations encountered in reactor physics, but merely provided some of the necessary theoretical development for arbitrary nonlinear equations. Details were sketchy, and the potential value of this early work was never realized.

Such was the state of the art when this thesis was begun, with the idea in mind of extending sensitivity analysis based on GPT for the time-independent neutron field to include burnup-related parameters, which depend not only on the time-dependent neutron field but also on the time-dependent nuclide field. In addition the governing equations are nonlinear, and thus further work in the nonlinear perturbation theory was required. The original goals of this work have nearly all been realized, but since the study was begun independent work has been published by other sources in some of the planned areas of endeavor. This recent work includes derivation of an adjoint equation for the *linear* transmutation equation by Gandini (15), with a modification to couple with static GPT results by Kallfelz (16), and some interesting work on nonlinear adjoint equations for fuel cycle costs published by Harris as part of his doctoral thesis (17). For the most part, these works represent special cases of the more general developments discussed

herein; however, the quality of this early work merits acknowledgement, and it is felt that the present work will provide useful and needed extensions to their work, as discussed below.

From a theoretical viewpoint it is convenient to categorize burnup perturbation analysis into two types. In this text these types are called the uncoupled and the coupled formalisms. The distinction lies in how the interaction between the nuclide and neutron fields is treated.

In the uncoupled perturbation method, it is assumed that a perturbation in the nuclide-field equation does not affect the flux field, and vice versa. In effect, the nonlinear coupling between the two field equations is ignored for the perturbed state; or alternatively, one might say that for the depletion perturbation analysis, the flux field is treated as an *input* quantity, and not as a dependent variable. With this assumption, it is legitimate to consider the flux field as data, which can be varied independently along with the other data parameters. This is the formulation originally addressed by Gandini and is only valid under limited circumstances. Kallfelz partially circumvented this problem by linking perturbation theory for the nuclide field with static GPT; however, his technique has the serious disadvantage of requiring a separate GPT calculation for each cross section in the nuclide field equation (16).

In the coupled formalism, the nuclide and neutron fields cannot vary independently. Any data perturbation which changes one will also change the other, because the two fields are constrained to "move"

only in a fashion consistent with their coupled field equations. In developing a workable sensitivity theory for the case of coupled neutron/nuclide fields, one must immediately contend with the specific type of formulation assumed in obtaining solutions to the burnup equations – the perturbation expressions themselves should be based on the approximate equations rather than the actual burnup equations, since the only solutions that exist for practical purposes are the approximate solutions. Harris' study of perturbation theory for generic nonlinear equations is not directly applicable to the approximation employed by most depletion codes, hence his "nonlinear adjoint equations" cannot be implemented into a code such as VENTURE. Furthermore, the adjoint burnup equations which were presented are limited to a simple model; e.g., they do not explicitly treat space dependence, nor arbitrary energy and angle dependence for the neutron flux field, and are applicable only to a specific type of response.

At present there exists a need for a unifying theory which starts from the general burnup equations and derives perturbation expressions applicable to problems of arbitrary complexity. In particular, the physical and mathematical consequences of approximate treatments for the time-dependent coupling interaction between the nuclide and flux fields should be examined, and the role of perturbation theory in defining sensitivity coefficients for generic "responses" of the flux and nuclide fields should be clarified. This study attempts to provide a general theoretical framework for burnup sensitivity theory *that is compatible with existing methods for treating the time dependence of the neutron field.*

In summary, the specific purposes of the present work are stated as follows:

1. To further investigate perturbation theory for nonlinear equations and contrast the technique to that for linear equations. Attention is given to the order of approximation inherent in "nonlinear adjoint equations," and the concept of a "first-order adjoint equation" is introduced.
2. To review various formulations of the burnup equations and to examine how perturbations affect the equations (e.g., "coupled" vs. "uncoupled" perturbations).
3. To derive appropriate adjoint equations for each of the formulations.
4. To present a calculational algorithm for numerically solving the adjoint burnup equations, and to summarize work completed at Oak Ridge in implementing the procedure.
5. To examine the physical meaning of the burnup adjoint functions and to illustrate their properties with analytic calculations.
6. To derive sensitivity coefficients for generic responses encountered in burnup analysis, both for variations in nuclear data and in initial conditions, and to establish the relation between coupled and uncoupled perturbation theory.
7. To present equations for uncertainty analysis in burnup calculations.
8. To give results of application of uncoupled, depletion perturbation theory to analysis of an irradiation experiment.

9. To give results of application of coupled, depletion perturbation theory to analysis of a denatured LMFBR.

## CHAPTER II

### ADJOINT EQUATIONS FOR NONLINEAR SYSTEMS

In this chapter we will examine in general terms the roles played by adjoint functions in analyzing effects of (a) perturbations in initial conditions and (b) in other input parameters on the solution to linear and nonlinear initial value problems. This discussion will serve as a prelude to following chapters in which perturbation theory will be developed for the specific case of the nonlinear burnup equations. Here we introduce the concepts of an "exact adjoint function" and a "first-order adjoint function," and contrast perturbation theory for linear and nonlinear systems. More details of the mathematics involved can be found in Appendix B.

First consider the reference state-vector  $y(x,t)$  described by the *linear* initial value problem

$$L(x,t) \cdot y(x,t) = \frac{\partial}{\partial t} y(x,t) \quad \text{II-1}$$

with a specified initial value  $y(x,0) \equiv y_0(x)$ . In this equation,  $x$  stands for all variables other than time (such as space, momentum, etc.), and  $L$  is a linear operator, assumed to contain no time derivative operators (however,  $\partial/\partial x$  operators are allowed). We will assume that it is desired to know some output scalar quantity from this system which depends on an integral over  $x^\dagger$  of the reference state vector evaluated at

---

$^\dagger [ ]_{x,y,\dots}$  indicates integration over  $x, y, \dots$ .

specified time  $T_f$ :

$$O_{T_f} = [h(x) \cdot y(x, T_f)]_x \quad \text{II-2}$$

The question often arises, How will the output  $O_{T_f}$  computed with the reference solution change if the initial condition or the operator  $L$  is perturbed?

To answer this, consider the following adjoint equation,<sup>†</sup> which is a final-value problem,

$$L^*y^*(x,t) = -\frac{\partial}{\partial t} y^*(x,t) \quad \text{II-3}$$

$$y^*(x, T_f) = h(x)$$

At this point there are two properties of the above equation which should be stressed. The first is that  $y^*$  is an integrating factor for Eq. II-1, since

$$[y^*Ly]_x - [yL^*y^*]_x = [y^* \frac{\partial}{\partial t} y]_x + [y \frac{\partial}{\partial t} y^*]_x,$$

which implies that

$$\frac{d}{dt} [y \cdot y^*]_x = 0 \quad \text{II-4}$$

Furthermore, integrating II-4 from  $t$  to  $T_f$  gives

---

<sup>†</sup> $L^*$  indicates the adjoint operator to  $L$ , defined by the commutative property  $[f \cdot Lg]_x = [gL^*f]_x$ .

$$[y(x,t) \cdot y^*(x,t)]_x = [y(x,T_f) \cdot y^*(x,T_f)] = 0_{T_f} \quad \text{II-5}$$

for all values of  $t$ .

Thus  $y^*$  is an integrating factor which transforms Eq. II-1 into an exact differential in time. It is interesting to note that Eq. II-4 expresses a conservation law for the term  $[y \cdot y^*]_x$ , which has led to the designation of this quantity as the "contributon density" in neutron transport theory (18, 19).

Evaluating Eq. II-5 at  $t = 0$  gives the fundamental relation

$$[y^*(x,0) \cdot y_0(x)]_x = 0_{T_f},$$

which shows that the desired output parameter can be evaluated simply by folding the initial condition of  $y$  with the adjoint function evaluated at  $t = 0$ , without ever even solving Eq. II-1! This relation is *exact*, and is a consequence of the fact that  $y^*$  is a Green's kernel for the output. An adjoint equation that provides solutions with the property in Eq. II-5 will be called an "exact adjoint equation."

The second important property of the adjoint function for a linear system arises from the fact that  $L^*$  is *independent of the forward solution*. Since  $L$  is linear, it does not depend on  $y$  and hence neither does  $L^*$ ; i.e., a perturbation in the reference value of  $y$  will not perturb  $y^*$ . This observation leads to the "predictor property" for a linear-equation adjoint function,

$$0_{T_f} = [y^* \cdot y_0]_x \quad \text{II-6}$$

for all values of  $y'(0)$ . Furthermore, subtracting II-5 from II-6 allows the change in  $\theta$  at  $T_f$  to be computed *exactly*, for arbitrary perturbations in initial conditions,

$$\Delta\theta_{T_f} = [y^*(0)\Delta y_0]_x \quad \text{II-7}$$

where  $\Delta$  implies a deviation from the reference state value found from Eq. II-1. Note that for a linear system, an exact adjoint equation will always have the property in Eq. II-7.

Now let us consider a *nonlinear* initial value problem, specified by the same initial condition  $y(x,0) = y_0(x)$ ,

$$M(y) \cdot y = \frac{\partial}{\partial t} y, \quad \text{II-8}$$

where  $M(y)$  is a nonlinear operator which now depends on the solution  $y$ . (See Appendix B.) If we proceed formally as before, the following adjoint equation is obtained:

$$M^*(y) \cdot y^* = - \frac{\partial}{\partial t} y^* \quad \text{II-9}$$

$$y^*(x, T_f) = h(x)$$

This "nonlinear adjoint equation" is actually linear in  $y^*$ , a property which has been noted by other authors (20) but it depends on the reference solution to the forward equation. As before, Eq. II-9 still provides an integrating factor for Eq. II-8, since it implies that

$$\frac{d}{dt} [y^*y]_x = 0$$

In this sense, Eq. II-9 is the "exact adjoint equation" for the reference system in Eq. II-8.

However, the predictor property of the adjoint system is no longer valid for arbitrary initial conditions, because in this case if the initial value of  $y$  is perturbed, Eq. II-8 becomes

$$M'(y') \cdot y' = - \frac{\partial}{\partial t} y' , \quad \text{II-10}$$

so that the adjoint equation for the perturbed system is

$$M'^*(y') \cdot y_p^* = - \frac{\partial}{\partial t} y_p^* . \quad \text{II-11}$$

The change in  $y_0$  has perturbed the adjoint operator, and hence it is impossible to express the adjoint system independent of the state of forward system, as could be done for a linear equation.

This problem can be illustrated in the following manner. First, express  $y'$  as the reference solution plus a time-dependent deviation from the reference state:

$$y'(t) = y(t) + \Delta y(t) \quad \text{II-12}$$

The left-hand side of II-10 is now expanded in a Taylor series about the reference solution (see Appendix B):

$$\left( M(y) \cdot y \right)' = \sum_{i=0}^{\infty} \frac{1}{i!} \delta^i (M \cdot y) , \quad \text{II-13}$$

where  $\delta^i$  is the perturbation operator defined in Appendix B.

When these values are substituted back into Eq. II-10, an equation for the time-dependent deviation is obtained:

$$\sum_{i=1}^{\infty} \frac{1}{i!} \delta^i(M \cdot y) = \frac{\partial}{\partial t} \Delta y \quad \text{II-14}$$

As shown in Appendix B,  $\delta^i$  is a nonlinear operator in  $\Delta y$  for all terms  $i > 1$ :

$$\delta^i(M \cdot y) = \delta^i(\Delta y) ,$$

so the left-hand side of Eq. II-14 is also a nonlinear operator in  $\Delta y$ . As discussed in Appendix B, an "exact adjoint operator" to this perturbed operator is given by

$$\sum_i \frac{1}{i!} \delta^{i*}(\Delta y) \cdot y^* \quad \text{II-15}$$

where  $\delta^{i*}(\Delta y)$  is any operator (in general depending on  $\Delta y$ ) which satisfies the relation

$$[y^* \delta^i(\Delta y)]_{x,t} = [\Delta y \delta^{i*}(\Delta y) \cdot y^*]_{x,t} \quad \text{II-16}$$

We thus have the "exact adjoint equation" for the perturbed equation in II-14:

$$\sum_i \frac{1}{i!} \delta^{i*}(\Delta y) \cdot y^* = - \frac{\partial}{\partial t} \Delta y \quad \text{II-17}$$

Note that  $\delta^{i*}$  is a linear operator in  $y^*$ .

Also, Equation II-17 explicitly shows how the "exact adjoint equation" depends on the perturbation in the forward solution. Defining the final condition in II-17 to again be  $y^*(T_f) = h$ , the predictor property is again exactly

$$\Delta O_{T_f} = y^*(0)\Delta y_0 ,$$

which is obtained by employing the relation in Eq. II-16. However, in this case the above equation is of academic interest only, since the perturbation  $\Delta y(t)$  must be known in order to compute  $y^*$ ! We can partially circumvent the problem by truncating the infinite series on the left-hand side of II-17 after the first term to obtain a "first-order adjoint equation"

$$\delta^{1*} \cdot y_1^* = - \frac{\partial}{\partial t} y_1^* \tag{II-18}$$

Using the relations in Appendix B,  $\delta^{1*}$  is found to be

$$\delta^{1*} = M^*(y) + \left( \frac{dM}{dy} \cdot y \right)^* \tag{II-19}$$

Substituting the above expression into Eq. II-18 gives

$$M^* y_1^* + \left( \frac{dM}{dy} \cdot y \right)^* y_1^* \tag{II-20}$$

The perturbed forward equation II-14 can be written

$$\delta^1(M \cdot y) + \sum_{i=2}^{\infty} \frac{1}{i!} \delta^i(\Delta y) = \frac{\partial}{\partial t} \Delta y ,$$

or

$$\left( M + \frac{dM}{dy} \cdot y \right) \Delta y + \sum_{i=2}^{\infty} \frac{1}{i!} \delta^i(\Delta y) = \frac{\partial}{\partial t} \Delta y \quad \text{II-21}$$

Using Eq. II-21 and the first-order adjoint equation in II-20, the predictor property for the perturbed nonlinear equation is

$$\Delta O_{T_f} = [y_1^*(x,0) \cdot \Delta y_0(x)]_x + \left[ y_1^* \sum_{i=2}^{\infty} \frac{1}{i!} \delta^i(\Delta y) \right]_{x,t}$$

where  $\delta^i(\Delta y) = \theta(\Delta y^i)$  (Note:  $\theta$  means "on the order of").

The above equation for the perturbed output is exact, however, it contains expressions which depend on  $\Delta y(x,t)$  in the higher order terms.

If terms higher than first order are neglected, we again obtain the linear relation between the change in the final condition and the change in the initial condition

$$\Delta y(T_f) \approx \left[ y_1^*(0) \cdot \Delta y_0 \right]_x , \quad \text{II-22}$$

but the relation is now only an approximation, in contrast to the exact relation for the linear case. Equation II-18 could also have been derived by first linearizing the forward equation (II-14), and then taking the appropriate adjoint operators; i.e., Eq. II-18 is the "exact"

adjoint equation for the *linearized* system, but is only a "first-order" adjoint, for the true nonlinear system.

Because of the extreme desirability of having an adjoint equation which is independent of changes in the forward solution, first-order adjoint functions are usually employed for perturbation analysis of nonlinear systems. The price which must be paid for this property is the introduction of second-order errors that do not appear in linear systems. Since the burnup of fuel in a reactor core is a nonlinear process, depletion sensitivity analysis is faced with this limitation and can be expected to break down for large perturbations in initial conditions.

For perturbations in parameters other than initial conditions, such as in some data appearing in the operator L on the left-hand side of II-1, even linear systems cannot be analyzed exactly with perturbation theory. For these cases, it is well known that (21)

$$\Delta O_{T_f} = \int_0^{T_f} [y^*(t)\Delta Ly(t)]_x dt + \theta[\Delta L\Delta y]_x . \quad \text{II-23}$$

For perturbation analysis of nonlinear systems using a first-order adjoint function, additional second-order terms are obtained, such as  $\Delta y^2$  as well as higher order terms. In general it is not obvious how much additional error (above the error normally encountered in linear systems) these terms will introduce, since the relative magnitudes and the possibility of cancelling errors must be considered. The accuracy

of the depletion perturbation method, which will be developed in the following sections, can only be determined by applying the technique to many real-world problems until some feel for its range of validity is established.

A simple extension of the preceding discussion is to allow the output observable  $0$  to be an integral over time of any arbitrary function of  $y(t)$  (differentiable in  $y$ ):

$$0 = [f(y)]_{x,t} \quad \text{II-24}$$

The first observable discussed is a special case of the above equation with

$$f(y) = h(x)y(x,t)\delta(t - t_f) , \quad \text{II-25}$$

where  $\delta$  is a Dirac delta function. The appropriate first-order adjoint equation for this general output is (using notation as in II-18) a fixed source problem,

$$\delta^{1*}y_1^* = - \frac{\partial}{\partial t} y_1^* - \frac{\partial f}{\partial y} \quad \text{II-26}$$

$$y_1^*(T_f) = 0 \quad \text{II-27}$$

Again note that Eq. II-26 reduces to Eq. II-18 when  $f$  is given by Eq. II-25, since in that case

$$\frac{\partial f}{\partial y} = h(x)\delta(t - t_f) \quad \text{II-28}$$

This delta-function source is equivalent to a fixed final condition of  $y^*(T_f) = \partial f / \partial y$  (21) and therefore Eq. II-26 is equivalent to Eq. II-18.

For the more general expression for 0, consider the result of a perturbation in the initial condition of Eq. II-8. The output is perturbed to

$$\begin{aligned} 0' &= [f(y')]_{x,t} = [f(y) + \frac{\partial f}{\partial y} \cdot \Delta y + \frac{\partial^2}{\partial y^2} f \Delta y + \dots]_{x,t}, \\ \Delta 0 &= \left[ \frac{\partial f}{\partial y} \Delta y + \dots \right]_{x,t} \end{aligned} \quad \text{II-29}$$

and the perturbed forward equation is again given by Eq. II-13, with the time-dependent change in  $y$  obeying Eq. II-21. Now multiply the first order adjoint equation (II-26) by  $\Delta y$ , and Eq. II-21 by  $y_1^*$ ; integrate over  $x$  and from  $t = 0$  to  $t = T_f$ ; and then subtract:

$$\int_0^T dt \frac{\partial}{\partial t} [y_1^* \Delta y]_x + \int_0^T dt \left[ \frac{\partial f}{\partial y} \Delta y \right]_x = \sum_{i=2}^{\infty} \left[ \frac{1}{i!} y_1^* \delta^i(M \cdot y) \right]_{x,t} \quad \text{II-30}$$

Substituting the value for  $\Delta 0$  from Eq. II-29 into II-30, and evaluating the first term on the left-hand side [recall,  $y_1^*(T) \equiv 0$ ] gives

$$[y_1^*(0) \cdot \Delta y_0]_x = \Delta 0 - \left[ \sum_{i=2}^{\infty} \frac{\partial^i}{\partial y^i} f \cdot \frac{\Delta y^i}{i!} + \sum_{i=2}^{\infty} \frac{1}{i!} y_1^* \delta^i(M \cdot y) \right]_{x,t} \quad \text{II-31}$$

Equation II-31 is still exact, and explicitly shows the terms involving powers of  $\Delta y$  higher than first order contained both in the perturbed response and in the  $\delta^i$  operator. If these terms are neglected, Eq. II-31 reduces to

$$\Delta 0 = [y_1^*(0) \cdot \Delta y_0]_x$$

Again we see that the first-order adjoint function allows one to estimate the change in the output to first-order accuracy, when the initial state is perturbed.

We will end this introductory development by summarizing the following important points concerning perturbation theory for linear and nonlinear initial value problems:

1. In a *linear* system, the change in the output due to an arbitrary change in initial condition can be computed exactly using perturbation theory (Eq. II-7)
2. In a *linear* system, the change in the output due to an arbitrary change in the system operator can be estimated only to *first-order accuracy* using perturbation theory (Eq. II-23)
3. For a *nonlinear* system, there exists an associated "first-order adjoint system" corresponding to the "exact adjoint system" for the linearized forward equation (Eq. II-26). This system depends on the reference forward solution, but is independent of variations about the reference state.

4. In a *nonlinear* system, the change in the output due to an arbitrary change in initial condition can be computed accurate only to first order with perturbation theory using a first-order adjoint function (Eq. II-22)

5. In a *nonlinear* system, the change in output due to an arbitrary change in the system operator can be estimated to first-order accuracy using perturbation theory based on the first-order adjoint function. Note that this is the same order of accuracy as in item 2 for a linear system, although usually the perturbation expressions for the nonlinear system will have more second order terms.

Having completed a general overview of nonlinear perturbation theory, we can now proceed with developing a perturbation technique for burnup analysis. Nearly all derivations of adjoint equations in the text are actually specializations of the general theory discussed in this chapter. It is an interesting exercise to determine the point in each derivation at which the assumption "neglect 2nd order terms" is made. Sometimes the assumption is obvious and sometimes it is more subtle, but the reader must be aware that this approximation is being made in each case, since we are dealing exclusively with first-order adjoint equations.

## CHAPTER III

### FORMULATIONS OF THE BURNUP EQUATIONS

In analyzing the time-dependent behavior of a power reactor, one finds that most problems that are encountered fall in one of three generic time scales. In this development, these will be labeled the short-range, intermediate-range, and long-range time periods.

The short-range time period is on the order of milliseconds to seconds, and is concerned with the power transients due to the rapid increase or decrease in the population of neutrons when a reactor is perturbed from critical. The study of these phenomena of course constitutes the field of reactor kinetics. Except possibly for extreme accident conditions, the material composition of the reactor will not change during these short time intervals.

The intermediate range involves time periods of hours to days. Problems arising on this time scale include computing the effect of xenon oscillations in an LWR, calculating efficient poison management programs, etc. Unlike the kinetics problem, the overall population of neutrons does not change significantly during intermediate-range problems, but the distribution of the neutrons within the reactor may change. Furthermore, the time-dependent behavior in the concentrations of some nuclides with short half-lives and/or high absorption cross sections (i.e., fission products) may now become important. When the space-dependent distribution of these nuclides significantly affects the space-dependent distribution of the flux, nonlinearities appear, and feedback with time constants on the order of hours must be considered.

The last time scale of interest is the long-range period, which may span months or even years. Analysis at this level is concerned with predicting long term isotopic changes within the reactor (fuel depletion, plutonium and fission product buildup, etc.), especially how these changes affect reactor performance and economics. Analysis in this time range must consider changes both in the magnitude and distribution of the neutron field, although the changes occur very much more slowly than for the kinetics case. But the most distinguishing feature of this type of analysis is the importance of time-dependent variables in the nuclide field. On this time scale the time-dependent behavior of a relatively large number of nuclides must be considered, and these changes will be fed back as changes in the neutron field; the nonlinearity appears with a much longer time constant than in the intermediate range case, however.

In reality, of course, processes in all three time ranges occur simultaneously in a power reactor, and their effects are superimposed. It is possible to write a single set of mathematical equations which fully describe the time variations in both the neutron and nuclide fields (22); however, in practice the equations cannot be solved efficiently due to the nonlinearities and the extremely widely spaced time eigenvalues. Therefore reactor physicists must assume separability for the three time scales. Specific solution techniques have evolved for each time range and are designed to exploit some property of the time scale of interest (e.g., slowly varying flux, etc.). In this work we will deal exclusively with the two longest time scales, with the major focus

being on calculations for the long-range scale; such calculations comprise the area called burnup or depletion analysis.

The purpose of this section is to review the burnup equations, expressing them in the operator form which will be followed throughout the text. We are interested in the interaction between the neutron flux field and the nuclide density field, both of which change with time and both of which influence one another.

A material reactor region is completely described by its nuclide density vector, which is defined by

$$\underline{N}(\hat{r}, t) = (N_1(\hat{r}, t), N_2(\hat{r}, t), \dots, N_n(\hat{r}, t)) \quad \text{III-1}$$

where  $N_i(\hat{r}, t)$  = atom density of nuclide  $i$  at position  $\hat{r}$  and time  $t$ .

While in operation, the reactor volume will also contain a population of neutrons whose distribution is described by the neutron flux field  $\phi(\hat{\rho})$ , where

$\hat{\rho}$  = vector in the 7-dimensional vector space of  $(\hat{r}, t, \hat{\Omega}, E)$ .

Note that the space over which  $\underline{N}$  is defined is a subdomain of  $\rho$ -space.

Given an initial reactor configuration that is described by  $\underline{N}_0(\hat{r})$  at  $t = 0$ , and that is exposed to the neutron flux field for  $t \geq 0$ , all future reactor configurations, described by the nuclide field  $\underline{N}(\hat{r}, t)$ , will obey the nuclide transmutation equation (Bateman equation)\*

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\*[ ]<sub>x,y,...</sub> indicates integration over x,y,... .

$$\frac{\partial}{\partial t} \underline{N}(\hat{r}, t) = [\phi(\hat{p}) \underline{R}(\sigma)]_{E, \Omega} \underline{N}(\hat{r}, t) + \underline{D}(\Lambda) \underline{N}(\hat{r}, t) + \underline{C}(\hat{r}, t) \quad \text{III-2}$$

where

$\underline{R}$  is a cross section matrix whose elements are

$\sigma_{ij}(\hat{r}, E)$  = microscopic cross section and yield data for production of nuclide  $i$  by nuclide  $j$ , and

$\sigma_{ii} = -\sigma_{ai}$  = absorption cross section for nuclide  $i$

$\underline{D}$  is a decay matrix whose elements are

$\Lambda_{ij}$  = decay constant for decay of nuclide  $j$  to nuclide  $i$ , and

$\Lambda_{ii} = -\Lambda_i$  = total decay constant for nuclide  $i$

$\underline{C}(\hat{r}, t)$  is an external source of nuclides, accounting for refueling, control rod motion, etc.

We will find it convenient to define a transmutation operator by

$$\underline{M} = \underline{M}(\phi(\hat{p}), \sigma(\hat{r}, E), \Lambda) = [\phi(\hat{p}) \underline{R}(\sigma)]_{E, \Omega} + D(\Lambda) \quad \text{III-3}$$

Then the equation for the nuclide field vector becomes

$$\frac{\partial}{\partial t} \underline{N}(\hat{r}, t) = \underline{M}(\phi, \sigma, \Lambda) \underline{N}(\hat{r}, t) + \underline{C}(\hat{r}, t) \quad \text{III-4}$$

The neutron-flux field obeys the time-dependent transport equation expressed by

$$\begin{aligned}
& 1/v \frac{\partial}{\partial t} \phi(\hat{p}) + \hat{\Omega} \cdot \nabla \phi(\hat{p}) + \underline{N}(\hat{r}, t) \cdot \underline{\sigma}_t(\hat{r}, E) \phi(\hat{p}) \\
& = \underline{N}(\hat{r}, t) \cdot \left[ \underline{\sigma}_s(\hat{r}, E', \Omega' \rightarrow E, \Omega) + (1 - \beta) \frac{\chi(E)}{4\pi} \underline{v\sigma}_f(E') \phi(\hat{p}) \right]_{E', \Omega'} \\
& + \sum_i \chi_{Di}(E) \lambda_i d_i(\underline{N}) \qquad \qquad \qquad \text{III-5}
\end{aligned}$$

where

$\underline{\sigma}_t$  is the total cross-section vector, whose components are the total microscopic cross sections corresponding to the components of  $\underline{N}$ ,

and similarly defined are

$\underline{\sigma}_s$ , as the differential-scatter cross-section vector

$\underline{v\sigma}_f$ , as the fission-production cross-section vector,

and

$\chi(E)$  = prompt neutron fission spectrum

$\chi_{Di}(E)$  = delayed neutron fission spectrum for precursor group  $i$

$\lambda_i$  = decay constant for precursor group  $i$

$d_i(\underline{N})$  =  $i$ th group-precursor concentration, which is an effective average over various components of  $\underline{N}$ .

$\beta$  = yield of all precursors, per fission neutron.

Defining the Boltzman operator in the indicated manner,  $B = B[\underline{N}(r, t), \underline{\sigma}(r, E)]$ , Eq. III-5 becomes

$$1/v \frac{\partial}{\partial t} \phi(\hat{\rho}) = B(\underline{N}, \sigma) \phi(\hat{\rho}) + \sum_i \chi_{D_i}(E) \lambda_i d_i(\underline{N}) \quad \text{III-7}$$

In the work that follows, the above equation will be called the "initial value" form of the neutron-field equation. (Note: The usual equations for describing delayed-neutron precursors are actually embedded in the nuclide-field equation.)

Equations III-4 and III-7 are the desired field equations for the nuclide and neutron fields within the reactor. In addition to these conditions, there may also be external constraints placed on the system, such as minimum power peaking, or some specified power output from the reactor. In general these constraints are met by adjusting the nuclide source  $\underline{C}$  in Eq. III-4, for example by moving a control rod. For this development we will consider only the constraint of constant power production:

$$[\underline{N}(\hat{r}, t) \cdot \underline{\sigma}_f(\hat{r}, E) \phi(\rho)]_\rho = P \quad \text{III-8}$$

In this study the system of coupled, nonlinear equations given by Eqs. III-4, 7, and 8 are referred to as the burnup equations. The unknowns are the nuclide and neutron fields, and the nuclide control source which must be adjusted to maintain criticality. These equations are obviously quite difficult to solve; in reality some suitable approximation must be used. One common approximation assumes that the Boltzman operator can be replaced by the diffusion operator, thus reducing the dimension of  $\rho$ -space from 7 to 5. Even with the diffusion

approximation, however, the system is still coupled nonlinearly. In the next section we will examine assumptions which will decouple Eqs. III-4 and III-7 at a given instant in time, but first let us consider an alternate formulation for the flux-field equation which is useful in numerical calculations for the long-range time scale.

Suppose that  $\phi(\rho)$  is slowly varying in time. Then at a given instant the term  $1/v \partial/\partial t \phi$  can be neglected. We will also assume that for the long exposure times encountered in burnup analysis, the fluctuations about critical arising from delayed-neutron transients are unimportant (i.e., on the average the reactor is critical so that the precursors are at steady state). With these assumptions Eq. III-7 can be approximated by

$$B(\underline{N})\phi(\hat{\rho}) = 0 , \quad \text{III-9}$$

if the prompt fission spectrum in Eq. III-5 is modified to  $(1 - \beta)\chi(E) + \sum \beta_i \chi_{D_i}(E)$ .

Equation III-9 is homogeneous and thus at any given time will have nontrivial solutions only for particular values (an infinite number) of  $\underline{N}$ . To simulate the effect of control-rod motion, we will single out one of the components of  $\underline{N}$  which will be designated the control nuclide  $N_c$ . Also we will express the B operator as the sum of a fission operator and a loss-plus-inscatter operator:

$$B = L - \lambda F , \quad \text{III-10}$$

so that Eq. III-9 becomes

$$\left[ L(\underline{N}, N_c) - \lambda F(\underline{N}, N_c) \right] \phi(\rho) = 0, \quad \text{III-11}$$

where

$$\lambda = \frac{1}{k_{\text{eff}}} = \text{instantaneous fundamental lambda mode eigenvalue.}$$

The value for  $N_c$  is usually found indirectly by adjusting its magnitude until  $\lambda = 1$ . The concentration of the control nuclide is thus fixed by the eigenvalue equation and does not need to be considered as an unknown in the transmutation equation.

An alternate method of solving Eq. III-9 is to directly solve the lambda mode eigenvalue equation (given  $\underline{N}$ ,  $\lambda$  is sought from Eq. III-11). In this case  $\lambda$  may or may not equal one. For both of these techniques, only the flux shape can be found from Eq. III-11. The normalization is fixed by the power constraint in Eq. III-8.

It is important to realize that both of these methods are approximations, and that in general they will yield different values for the flux shape. The former case is usually closer to "reality" (i.e., to the true physical process) while the latter is usually faster to solve numerically. For many problems concerned only with nuclide densities, results are not extremely sensitive to the approximation used (23, 24).

We will next write  $\phi(\rho)$  as a product of time-dependent normalization factor,  $\Phi$ , and a slowly varying shape function  $\psi$ , which is a solution to Eq. III-11 normalized to unity; i.e.,

$$\phi(\rho) = \Phi(t)\psi(\rho) \quad \text{III-12}$$

with

$$[\psi(\rho)]_{E,\Omega,V} = 1 \quad \text{III-13}$$

The normalization factor is fixed by the power constraint

$$H(\underline{N}, \sigma_f, \psi) \cdot \Phi = P, \quad \text{III-14}$$

where

$$H \equiv [\underline{N} \cdot \sigma_f \psi(\rho)]_{E,\Omega,V} \quad \text{III-15}$$

In this form, the burnup equations can be expressed concisely in matrix notation as

$$\begin{bmatrix} L(\underline{N}) - \lambda F(\underline{N}) & 0 & 0 \\ 0 & H(\underline{N}, \psi, \alpha) & 0 \\ 0 & 0 & \underline{M}(\Phi, \psi, \alpha) \end{bmatrix} \begin{bmatrix} \psi \\ \Phi \\ \underline{N} \end{bmatrix} = \begin{bmatrix} 0 \\ P \\ \frac{\partial}{\partial t} \underline{N} \end{bmatrix} \quad \text{III-16}$$

For future reference, Eq. III-16 will be called the time-continuous, eigenvalue form of the burnup equations, since both the nuclide and neutron fields (as well as the eigenvalue  $\lambda$ ) occur as continuous functions in time. The only approximations which have been made so far are to neglect the time derivative of the flux and the transients in delayed-neutron precursors. However, this time-continuous form of the burnup equations is not practical for most applications, since at any

instant in time they contain products of the unknowns  $\underline{N}$ ,  $\psi$ , and  $\Phi$ ; i.e., the equations are still nonlinear. For numerical calculations we must make further assumptions which will approximate the nonlinear equations with a cost-efficient algorithm. Specifically, it is necessary to minimize the number of times which the neutron transport equation must be solved, since calculating the neutron field requires much more computing time than calculating the nuclide field.

The approximation made in most present-day depletion codes is based on decoupling the calculations for the neutron and nuclide fields at a given instant in time by exploiting the slowly varying nature of the flux. The simplest decoupling method is to treat the flux as totally separable in time and the other phase-space variables over the entire time domain  $(t_0, t_f)$ . In this case the shape function is time-independent, and thus

$$\phi(\hat{p}) = \Phi(t)\psi_0(\hat{r}, E, \hat{\Omega}) \quad \text{for } 0 < t < t_f, \quad \text{III-17}$$

The shape function  $\psi_0$  can be determined from a time-independent calculation at  $t = 0$  using one of the eigenvalue equations discussed in the previous section. As before it is normalized such that

$$[\psi_0(\hat{r}, E, \hat{\Omega})]_{E, \Omega, V} = 1 \quad \text{III-18}$$

Substituting Eq. III-17 into Eq. III-2,

$$\frac{\partial}{\partial t} \underline{N}(\hat{r}, t) = \Phi(t) [\psi_0 \underline{R}(\sigma)]_{E, \Omega} \underline{N} + \underline{D} \underline{N} + \underline{C} \quad \text{III-19}$$

Equation III-19 can be simplified by writing the first term on the RHS as

$$\Phi(t) \underline{R}_0(\sigma_0) \underline{N}(\hat{r}, t), \quad \text{III-20}$$

where  $\underline{R}_0$  is a one-group cross-section matrix whose components have the form

$$\sigma_0(\hat{r}) = [\psi_0(\hat{r}, E, \Omega) \sigma(\hat{r}, E)]_{E, \Omega} \quad \text{III-21}$$

The cross-section matrix is rigorously composed of space-dependent, one-group microscopic data which can be evaluated once and for all at  $t = 0$ . In reality, detailed space-dependent depletion calculations are rarely performed due to prohibitive computing cost. Usually the reaction matrix is averaged over some limited number of spatial zones (for example, a core zone, a blanket zone, etc.); in this case of "block depletion" the solution to the transmutation equation approximates the average nuclide field over each spatial region (25). The cross-section elements of  $\underline{R}$  for region  $z$  are given by

$$\sigma_0(z) = [\psi_0(z, E, \Omega) \sigma(z, E)]_{E, \Omega} \quad \text{III-22}$$

where  $\psi_0(z, E, \Omega) \equiv [\psi_0(\hat{r}, E, \Omega)]_{V_z}$

which has a normalization

$$\sum_z [\psi_0(z, E, \Omega)]_{E, \Omega} = 1 \quad \text{III-23}$$

Throughout the remainder of this study we will not explicitly refer to this region-averaging procedure for the nuclide-field equation. This should cause no confusion since the spatial variable "r" in Eq. III-21 can refer to either the region or spatial interval, depending on the case of interest. There is no coupling between the various r-points in the transmutation equation except through the flux-shape function, and therefore the equation for the region-averaged nuclide field appears the same as for the point-dependent field; only the cross-section averaging is different.

The value for the flux normalization in Eq. III-19 is computed from the power constraint in Eq. III-8:

$$\Phi(t) = P / [\sigma_f(\hat{r}, E) \underline{N}(\hat{r}, t) \psi_0(\hat{r}, E, \Omega)]_{E, V, \Omega} \quad \text{III-24}$$

For numerical calculations this normalization calculation is only done at discrete time intervals in the time domain,

$$\Phi_i = \frac{P}{[\sigma_f(r, E) \underline{N}_i(r) \psi_0]_{E, V, \Omega}}, \text{ where } \underline{N}_i \equiv \underline{N}(\hat{r}, t_i) \quad \text{III-25}$$

and is then held constant over some "broad time interval" ( $t_i, t_{i+1}$ ).

One should realize that the broad time intervals at which the flux normalization is performed do not usually correspond to the finer time intervals over which the nuclide field is computed. To avoid confusion on this point, we will continue to represent  $\underline{N}$  as an explicit function of time, rather than in its finite-difference form.

Note the discontinuity in  $\Phi_i$  at each of the time intervals: at  $t = t_i^-$ ,  $\Phi = \Phi_{i-1}$ , while at  $t = t_i^+$ ,  $\Phi = \Phi_i$ . There is no corresponding discontinuity in the nuclide field; i.e.,

$$\underline{N}(\hat{r}, t_i^+) = \underline{N}(\hat{r}, t_i^-) ,$$

but there is discontinuity in the derivative of  $\underline{N}$  at  $t_i$ .

Because of the discontinuities in the flux field and the eigenvalue, this formulation (and the one which follows) is called the "time-discontinuous eigenvalue" approximation.

With all the preceding assumptions, the nuclide-field equation becomes

$$\frac{\partial}{\partial t} \underline{N}(\hat{r}, t) = \Phi_i R_0 \underline{N}(\hat{r}, t) + \underline{D} \underline{N}(\hat{r}, t) + \underline{C}(\hat{r}, t) , \quad \text{III-26}$$

for  $t_i < t < t_{i+1}$  with

$$\underline{N}(\hat{r}, t_i^+) = \underline{N}(\hat{r}, t_i^-) \quad \text{III-27}$$

as the initial condition of the broad time interval.

At a given value of  $r$  (either a region or a point), Eq. III-26 depends only on the time coordinate; i.e., it is an ordinary differential equation in which  $r$  appears as a parameter. The assumption of total separability in the time variable of the flux field has completely eliminated the need for solving the transport equation, except for the initial eigenvalue calculation at  $t = 0$  which was required to collapse

the cross-section data. Some computer codes, such as ORIGEN (26), store standard cross-section libraries containing few-group cross sections ( $\sim 3$  groups) that have been collapsed using flux spectra for various types of reactors (e.g., a PWR library, an LMFBR library, etc.). It is then only necessary to input the ratios (usually estimated) of the epithermal and fast fluxes to the thermal flux in order to obtain the one-group reaction matrix.

In summary, the calculation usually proceeds as follows:

- (i) solve Eq. III-11 at  $t = 0$  for flux shape
- (ii) integrate cross-section data using Eqs. III-21 or III-22
- (iii) solve Eq. III-25 for flux normalization at  $t = t_i$
- (iv) solve Eq. III-26 for  $\underline{N}(\underline{r}, t)$  over the broad time interval
 
$$t_i < t < t_{i+1}$$
- (v) go to iii

This rather simplistic approximation is employed mainly when emphasis is on computing the nuclide rather than the neutron field, and when the flux shape is known (or assumed) over the time scale of interest. Example applications include calculation of saturating fission products (27), analysis of irradiated experiment samples (28), and determination of actinide waste burnout in an LMFBR (29).

When the time variation of the flux shape becomes important, or when accurate values for flux-dependent parameters such as reactivity are required (as in analysis of a power reactor), a more sophisticated technique must be used. The most commonly employed calculational method for this analysis is based on a "quasi-static" approximation, a mathematical method sometimes referred to as "quasilinearation" (30).

The quasi-static depletion approximation, as used in this investigation,\* essentially consists of a series of the above type calculations (31). Instead of assuming that the flux shape is totally separable in time over the domain of interest, it is only required that  $\psi$  be constant over some finite interval  $(t_i, t_{i+1})$ . The flux-shape function for each broad time interval is obtained from an eigenvalue calculation at the "initial" state  $t_i$ ,

$$\left[ L(\underline{N}_i) - \lambda F(\underline{N}_i) \right] \psi_i(\hat{r}, E, \hat{\Omega}) = 0 \quad \text{III-28}$$

for  $t = t_i, \dots$ , ( $i = 1$ , through number of time intervals) and the flux normalization is obtained from the power constraint at  $t = t_i$ ,

$$\Phi_i [\psi_i(\hat{\rho}) \underline{N}_i \sigma_f]_{E, V, \Omega} = P_i, \quad \text{III-29}$$

for  $t = t_i, \dots$ . Thus the time-dependent flux is approximated by the stepwise continuous function

$$\phi(\rho) \sim \Phi_i \psi_i(\hat{r}, E, \hat{\Omega}), \quad t_i^+ < t < t_{i+1}^- \quad \text{III-30}$$

After each eigenvalue calculation, a new set of one-group cross sections can be generated using the new value of  $\psi_i$ , resulting in a new cross-section matrix

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\*Beware of difference in terminology from kinetics studies.

$$\underline{R}_i(\sigma_i) \equiv [\psi_i \underline{R}]_{E, \Omega}, \quad \text{III-31}$$

with components

$$\sigma_i(\hat{r}) = [\sigma(\hat{r}, E) \psi_i(\hat{r}, E, \Omega)]_{E, \Omega} \quad \text{III-32}$$

The transmutation equation is then solved over the next time interval using the "constant" matrix  $\underline{R}_i$ ,

$$\frac{\partial}{\partial t} \underline{N}(\hat{r}, t) = \phi_i \underline{R}_i \underline{N}(\hat{r}, t) + \underline{DN}(\hat{r}, t) + \underline{C}(\hat{r}, t), \quad \text{III-33}$$

$$t_i^+ < t < t_{i+1}^-$$

Note that the time-dependent flux given in Eq. III-30 is again discontinuous (this time, both the shape and the magnitude) at the boundaries of the broad time intervals, while the nuclide field is continuous (its derivative is discontinuous). The basic procedure for the quasi-static approximation is as follows:

- (i) solve flux eigenvalue equation for  $\psi_i$  at  $t_i$
- (ii) integrate cross-section data using Eq. III-32
- (iii) solve Eq. III-29 for normalization at  $t_i$
- (iv) solve Eq. III-33 between  $t_i$  and  $t_{i+1}$
- (v) go to (i)

Variations of this basic procedure are presently in use. For example, some computer programs (32) iterate on the initial and final conditions of a broad time interval until the *average* power production over the interval (as opposed to the end-point values) meets some

specified value; however, these refinements will not be considered in this study.

In Eqs. III-28, 29, and 33, we have developed the quasi-static burnup equations. The approximations that were made have reduced the original coupled nonlinear equations to a series of equations which appear linear at any given instant. In reality, of course, the equations still approximate a nonlinear process, since a change in the value of  $\psi_i$  is ultimately fed back as a perturbation in the Boltzman operator for the calculation of  $\psi_{i+1}$ . It is this nonlinearity which will make the adjoint burnup equations derived shortly quite interesting.

Let us now review the assumptions leading to the various approximations for the burnup equations. Recall that the basic assumption made for the long-term time scale was that the flux field is slowly changing with time, which allowed us to transform the original initial-value problem into an instantaneous  $\lambda$  mode eigenvalue equation (the "time-continuous eigenvalue" approximation). We were then able to make further simplifications by writing the time-dependent flux as a product of a normalization and a slowly varying shape function. For numerical calculations the shape function is approximated by a Heaviside-function time behavior; i.e., it is assumed to remain constant over relatively broad time intervals, the most extreme case being a single broad interval spanning the entire time domain (total-time separability). This assumption resulted in the quasi-static or time-discontinuous eigenvalue formulation. Note that the assumptions leading to the

quasi-static depletion method are related to similar assumptions made in deriving the adiabatic and quasi-static kinetics approximations for the short-range time scale, although neglecting delayed neutrons and introducing a time-varying nuclide field makes the relation somewhat blurred.

This last formulation is well suited for the long-term time scale in which the flux shape does not change significantly over several days, or perhaps weeks. However there are some problems which arise in the intermediate time scale which require the initial-value formulation, such as analysis of Xe oscillations. The usual procedure for this type of analysis is to linearize the initial-value burnup equations in III-2 and III-7 and to neglect the effect of delayed neutrons (33). Since in the intermediate range fuel depletion can be neglected, the flux normalization is constant in time. Furthermore, the nuclide-field vector has a limited number of components (usually the only nuclides of interest for the Xe problem are  $^{139}\text{I}$  and  $^{139}\text{Xe}$ ) whose time-dependent behavior must be explicitly treated.

The appropriate equations describing the deviations in the flux and nuclide fields about steady-state values are thus:

$$\underline{B}(\underline{N}) \cdot \Delta\phi + \frac{\partial \underline{B}}{\partial \underline{N}} \phi \Delta \underline{N} = \frac{1}{v} \frac{\partial}{\partial t} \Delta\phi \quad \text{III-34}$$

$$\underline{M}(\phi) \cdot \Delta \underline{N} + \frac{\partial \underline{M}}{\partial \phi} \underline{N} \Delta\phi = \frac{\partial}{\partial t} \Delta \underline{N} , \quad \text{III-35}$$

where for Xe analysis  $\Delta \underline{N}$  is zero except for the Xe and I isotopes. In matrix notation we have

$$\begin{bmatrix} B(\underline{N}) & \frac{\partial B}{\partial \underline{N}} \phi \\ \frac{\partial \underline{MN}}{\partial \phi} & \underline{M} \end{bmatrix} \begin{bmatrix} \Delta \phi \\ \Delta \underline{N} \end{bmatrix} = \frac{\partial}{\partial t} \begin{bmatrix} \frac{1}{v} \Delta \phi \\ \Delta \underline{N} \end{bmatrix}$$

III-36

Although most of the work in this thesis will be concerned with obtaining a perturbation methodology for the eigenvalue formulation of the burnup equations (i.e., for the long-time scale analysis), we will also examine a perturbation technique for the initial-value formulation that can be employed to analyze the above type of problem which occurs in the intermediate time range.

## CHAPTER IV

### DERIVATION OF ADJOINT EQUATIONS FOR BURNUP ANALYSIS

The desired end result of virtually all design calculations is an estimated value for some set of reactor performance parameters. Each such parameter will be called a "response" in this study. For the case of burnup analysis, the generic response will be an integral of the flux and nuclide fields; i.e., it is mathematically a functional of *both* fields, which in turn are coupled through the respective field equations. As an example, the desired response may be the final  $^{239}\text{Pu}$  mass at shutdown (a nuclide response); it may be the time-integrated damage to some nondepleting structural component (a flux response); or it may be some macroscopic reaction rate (a nuclide and flux functional). These functionals all take the general form of

$$R = R(\phi(\hat{p}), \underline{N}(\hat{r}, t), \underline{h}) , \quad \text{IV-1}$$

For future reference, we also note that the quasi-static formulation of Eq. IV-1 is

$$R_{QS} = R(\phi_i, \psi_i, \underline{N}, \underline{h}) . \quad \text{IV-2}$$

In these expressions  $\underline{h}$  is a "realization vector" which can have the form of a cross section or of some constant vector which determines the response of interest. There may actually be several realization vectors appearing in the response, in which case  $\underline{h}$  will symbolically represent all realization vectors.

Let us consider several types of specific responses. First, recall from Chapter II that the system output (for the perturbation development, "output" is synonymous to "response") is of two generic types: one is evaluated at an instant in time, while the other is an integral over a time interval; the relation between the two has been previously illustrated. The former type response will be called a final-time response, and the latter a time-integrated response.

One important class of responses depends only on the nuclide field — a "nuclide-field response,"

$$R = R(\underline{h}, \underline{N}) \quad \text{IV-3}$$

In this case,  $\underline{h}$  will be a vector with constant components. For example suppose that  $R$  corresponds to the number of atoms of Pu-239 at 100 days after startup. Then

$$R = [\underline{h} \cdot \underline{N}(r, t = 100)]_V, \quad \text{IV-4}$$

where all components of  $\underline{h}$  are 0 except the component for Pu-239 which is 1. For the spatial average Pu-239 concentration, simply change the 1 to  $1/V$ , where  $V$  is the volume. If  $R$  corresponds to fissile inventory (kg.) after 100 days, then  $\underline{h}$  has nonzero components for all fissile nuclides, and the values are equal to the respective mass per atom values.

These examples were all final-time responses, but similar definitions will hold for time-integrated responses

$$R = [\underline{h} \cdot \underline{N}(r, t)]_{V, t}, \quad \text{IV-5}$$

such as for a time-average nuclide density. We may also be interested in nuclide ratios

$$R = \frac{[\underline{h}_1 \underline{N}]}{[\underline{h}_2 \underline{N}]}, \quad \text{IV-6}$$

as for an enrichment parameter.

Another class of responses of interest in burnup analysis depends on reaction rates. For example, if one wished to know the capture rate in U-238 after 100 days,

$$R = \left[ \sigma_c^{U8}(r, E) \phi(r, E, \Omega, t = 100), N^{U8}(r, t = 100) \right]_{V, E, \Omega}$$

We see in this case that  $\underline{h}$  has all zero components except for U-238, where its value is equal to the U-238 capture cross section; i.e., for this example the component of  $\underline{h}$  is function of space and energy. A very important response belonging in this class is  $k_{\text{eff}}$ , which is a ratio of reaction rates:

$$k_{\text{eff}}(t = 100) = \frac{[\underline{h}_1(r, E) \underline{N}(r, t = 100) \phi(r, E, \Omega, t = 100)]_{V, E, \Omega}}{[\underline{h}_2(r, E) \underline{N}(r, t = 100) \phi(r, E, \Omega, t = 100)]_{V, E, \Omega}}$$

where

$$\underline{h}_1 \underline{N} = F(\underline{N})$$

$$\underline{h_2N} = L(\underline{N})$$

IV-7

with  $F$ ,  $L$  being the fission and loss operators previously defined in Eq. III-10.

It can be seen that a very wide variety of reactor parameters can be addressed using the notation discussed. Rather than limit the following development to any one particular type of response, we will continue to use  $R$  to stand for any arbitrary response depending on either or both the nuclide and neutron fields.

It is the goal of perturbation and sensitivity analysis to find the effect that varying some nuclear data parameter (e.g., a cross section, a decay constant, a branching ratio, etc.) or the initial nuclide field will have on the response  $R$ . This will be accomplished by defining a "sensitivity coefficient" for the data in question, which will relate the percent change in  $R$  to the percent change in the data.

For example, let  $\alpha$  be a nuclear data parameter contained in either or both the  $B$  and the  $\underline{M}$  operators. Then the sensitivity of  $R$  to  $\alpha$  is given by

$$\delta R/R = \left[ S(\hat{p}) \frac{\delta \alpha}{\alpha} (\hat{p}) \right]_{\rho} + \text{second-order terms} \quad \text{IV-8}$$

For small  $\delta \alpha$ , we obtain the familiar linear relation between  $\delta R/R$  and  $\delta \alpha/\alpha$ , with  $S(\hat{p})$  serving as the sensitivity coefficient at position  $\hat{p}$  in phase space. A change in the value of  $\alpha$  in general will perturb both the nuclide and flux fields in some complex manner, depending on the specific  $\delta \alpha(\hat{p})$ .

Treating the response as an implicit function of  $\alpha$ ,  $\underline{N}$ , and  $\phi$ , we can expand  $R$  in a first-order Taylor series about the unperturbed state

$$R' \cong R + \left[ \left( \frac{\partial R}{\partial \alpha} \right) \delta \alpha(\hat{\rho}) + \left( \frac{\partial R}{\partial \underline{N}} \right) \frac{d\underline{N}}{d\alpha} \delta \alpha(\hat{\rho}) + \left( \frac{\partial R}{\partial \phi} \right) \frac{d\phi}{d\alpha} \delta \alpha(\hat{\rho}) \right]_{\rho} \quad \text{IV-9}$$

$$\delta R/R \cong \left[ \frac{\alpha}{R} \left( \frac{\partial R}{\partial \alpha} + \frac{\partial R}{\partial \underline{N}} \frac{d\underline{N}}{d\alpha} + \frac{\partial R}{\partial \phi} \frac{d\phi}{d\alpha} \right) \frac{\delta \alpha}{\alpha}(\hat{\rho}) \right]_{\rho} \quad \text{IV-10}$$

From this expression it is evident that

$$S(\hat{\rho}) = \alpha/R \left( \frac{\partial R}{\partial \alpha} + \frac{\partial R}{\partial \underline{N}} \frac{d\underline{N}}{d\alpha} + \frac{\partial R}{\partial \phi} \frac{d\phi}{d\alpha} \right) . \quad \text{IV-11}$$

It is important to realize that the derivatives  $d\underline{N}/d\alpha$  and  $d\phi/d\alpha$  are *not independent*, since they must be computed from the constraint conditions (i.e., the field equations) which are coupled in  $\underline{N}$  and  $\phi$  (34).

In order to clarify this statement, consider the coupled burnup equations in Eq. III-16. The time-continuous eigenvalue form of the flux equation will be used in the illustration, and so we must first write Eq. IV-10 in terms of the magnitude and shape functions:

$$\Delta R = \left[ \frac{\partial R}{\partial \alpha} \Delta \alpha + \frac{\partial R}{\partial \psi} \Delta \psi + \frac{\partial R}{\partial \Phi} \Delta \Phi + \frac{\partial R}{\partial \underline{N}} \cdot \Delta \underline{N} \right]_{\rho} . \quad \text{IV-12}$$

We wish to show that the variations (and hence the derivatives in Eq. IV-11) in  $\alpha$ ,  $\psi$ ,  $\Phi$  and  $\underline{N}$  are dependent. This can be seen by considering variations about some reference state described by Eq. III-16. After linearization, the perturbed equations become

$$\begin{bmatrix} B & 0 & \frac{\partial B}{\partial N} \psi \\ \frac{\partial H}{\partial \psi} \phi & H & \frac{\partial H}{\partial N} \phi \\ \frac{\partial M}{\partial \psi} N & \frac{\partial M}{\partial \phi} N & M \end{bmatrix} \begin{bmatrix} \Delta \psi \\ \Delta \phi \\ \Delta N \end{bmatrix} = \frac{\partial}{\partial t} \begin{bmatrix} 0 \\ 0 \\ \Delta N \end{bmatrix} - \begin{bmatrix} \frac{\partial B}{\partial \alpha} \psi \\ \frac{\partial H}{\partial \alpha} \phi \\ \frac{\partial M}{\partial \alpha} N \end{bmatrix} \Delta \alpha \quad \text{IV-13}$$

The coupling between the field variations is apparent in this equation. In theory the above system of equations could be solved and  $\Delta R$  estimated using Eq. IV-12. In reality this is not practical since the "source" on the right-hand side of the equation depends on  $\Delta \alpha$ . Instead, it is much more efficient to use the adjoint system to define sensitivity coefficients independent of the particular data being perturbed.

We will now obtain appropriate adjoint equations for the various formulations of the burnup equations discussed in the previous chapter.

#### A. Time-Continuous Eigenvalue Approximation

From the discussion in Chapter II we already know that the adjoint system appropriate for the nonlinear equations in III-16 is actually a first order adjoint; and furthermore we know that the first order adjoint equations can be obtained in a straightforward manner from the linearized equations in IV-13. Therefore, let us consider the following inhomogenous system of equations, adjoint to Eq. IV-13.

$$\begin{bmatrix} B^* & \left(\frac{\partial H}{\partial \psi} \phi\right)^* & \left(\frac{\partial \underline{M} \underline{N}}{\partial \psi}\right)^* \\ 0 & H^* & \left(\frac{\partial \underline{M} \underline{N}}{\partial \phi}\right)^* \\ \left(\frac{\partial B}{\partial \underline{N}} \psi\right)^* & \left(\frac{\partial H}{\partial \underline{N}} \phi\right)^* & \underline{M}^* \end{bmatrix} \begin{bmatrix} \underline{\Gamma}^* \\ \underline{P}^* \\ \underline{N}^* \end{bmatrix} = -\frac{\partial}{\partial t} \begin{bmatrix} 0 \\ 0 \\ \underline{N}^* \end{bmatrix} - \begin{bmatrix} \frac{\partial R}{\partial \psi} \\ \frac{\partial R}{\partial \phi} \\ \frac{\partial R}{\partial \underline{N}} \end{bmatrix} \quad \text{IV-14}$$

Note that the "adjoint source" depends only on the response of interest. This specific form for the source was chosen for the following reason: multiply Eq. IV-13 by the vector  $(\underline{\Gamma}^*, \underline{P}^*, \underline{N}^*)$  and Eq. IV-14 by  $(\Delta\psi, \Delta\phi, \Delta\underline{N})$ ; integrate over  $\Omega, E,$  and  $V$ ; and subtract,

$$\begin{aligned} & \frac{\partial}{\partial t} [\Delta\underline{N} \cdot \underline{N}^*]_V - \left[ \left( \underline{\Gamma}^* \frac{\partial B \psi}{\partial \alpha} + \underline{P}^* \frac{\partial H}{\partial \alpha} \phi + \underline{N}^* \frac{\partial \underline{M} \underline{N}}{\partial \alpha} \right) \Delta\alpha \right]_{\Omega, E, V} \\ & + \left[ \frac{\partial R}{\partial \psi} \Delta\psi + \frac{\partial R}{\partial \phi} \Delta\phi + \frac{\partial R}{\partial \underline{N}} \Delta\underline{N} \right]_{\Omega, E, V} = 0 \quad \text{IV-15} \end{aligned}$$

Defining  $\underline{N}^*(t=T_f) = 0$ , we can now integrate Eq. IV-15 over time to give

$$\begin{aligned} \Delta R/R = & \left[ 1/R \Delta\underline{N}_0 \cdot \underline{N}^* \right]_V + \int \left[ \frac{\alpha}{R} \left( \frac{\partial R}{\partial \alpha} - \frac{\underline{\Gamma}^* \partial B \psi}{\partial \alpha} - \frac{\underline{P}^* \partial H}{\partial \alpha} \phi \right. \right. \\ & \left. \left. + \underline{N}^* \frac{\partial \underline{M} \underline{N}}{\partial \alpha} \right) \frac{\Delta\alpha}{\alpha} \right]_{\Omega, E, V} dt \quad \text{IV-16} \end{aligned}$$

and thus

$$S_\alpha(\rho) = \frac{\alpha}{R} \left( \frac{\partial R}{\partial \alpha} - \frac{\underline{\Gamma}^* \partial B \psi}{\partial \alpha} - \frac{\underline{P}^* \partial H}{\partial \alpha} \phi + \underline{N}^* \frac{\partial \underline{M} \underline{N}}{\partial \alpha} \right) \quad \text{IV-17}$$

This last expression represents the sensitivity coefficient to changes in data in the time-continuous, eigenvalue form of the burnup equations. It is independent of the data perturbation. From the first term on the right-hand side of IV-16, one can also see that the sensitivity coefficient for a change in the initial condition  $N_0$  is simply

$$S_{N_0}(r) = \underline{N}^*(r, t = 0) \cdot \frac{1}{R} . \quad \text{IV-18}$$

The adjoint equation in IV-14 is quite interesting in its physical interpretation. More time will be given to examining the "importance" property of the adjoint functions in a later chapter. For now simply note that the adjoint equation is linear in the adjoint variables and contains the reference values for the forward variables (a general property of first-order adjoint equations, as discussed in Chapter II). Also notice that there is coupling between the various adjoint equations, suggesting that the adjoint functions must somehow interact with each other.

It was previously pointed out that the time-continuous form of the burnup equation is not efficient to solve numerically. Such is also the case for the adjoint system. In the forward case, this problem was overcome by using a quasi-static approximation for the equations, and an adjoint system for this formulation will be developed shortly. But first we should examine a simpler approximation based on Eq. IV-14 which has been shown to give good results for some types of problems.

### B. Uncoupled Perturbation Approximation

Let us suppose that we have computed or have been given a reference solution to the burnup equations for some case of interest; i.e., we have available  $\underline{N}(r,t)$ ,  $\Phi(t)$ ,  $\psi(r,E,\Omega,t)$  and their accuracy is indisputable. When a perturbation is made in some input data, the perturbation in the fields will obey Eq. IV-13 to first order. Now if the neutron and nuclide fields are only loosely coupled, then the perturbed fields can vary essentially independently about the reference state; i.e., the *perturbations* in the neutron and nuclide fields will be uncoupled (this does not exclude a coupled, nonlinear calculation to determine the reference state). Mathematically, this approximation amounts to neglecting the off-diagonal terms in Eq. IV-13 containing derivatives of one field with respect to the other, so that the adjoint system is

$$\begin{bmatrix} B^* & \left(\frac{\partial H}{\partial \psi} \Phi\right)^* & 0 \\ 0 & H^* & 0 \\ 0 & 0 & \underline{M}^* \end{bmatrix} \begin{bmatrix} \Gamma^* \\ P^* \\ \underline{N}^* \end{bmatrix} = -\frac{\partial}{\partial t} \begin{bmatrix} 0 \\ 0 \\ \underline{N}^* \end{bmatrix} - \begin{bmatrix} \frac{\partial R}{\partial \psi} \\ \frac{\partial R}{\partial \Phi} \\ \frac{\partial R}{\partial \underline{N}} \end{bmatrix} \quad \text{IV-19}$$

Note that the 2nd term in row 1 relates coupling between magnitude and shape of the neutron field (not between neutron and nuclide fields) and hence must be retained. There is now no coupling between the nuclide and neutron adjoint functions. There are several cases of practical interest which we will examine.

First, suppose that the response is a time-independent ratio of microscopic reaction rates. This response depends only on the flux shape and is equivalent to a static response of

$$R = \frac{[h_1\psi]_{r,E,\Omega}}{[h_2\psi]_{r,E,\Omega}} \quad \text{IV-20}$$

so that

$$\frac{\partial R}{\partial N} = 0, \quad \frac{\partial R}{\partial \Phi} = 0.$$

In this case, we simply obtain the familiar generalized adjoint equation for the static case:

$$(L^* - \lambda F^*)\Gamma^* = \frac{h_1 \cdot [h_2\phi]_{r,E,\Omega} - h_2 [h_1\phi]_{r,E,\Omega}}{[h_2\phi]_{r,E,\Omega}^2}$$

Now suppose that R is a linear, time-independent functional of the form

$$R = \Phi[h \cdot \psi]_{r,E,\Omega} \quad \text{IV-22}$$

This response depends not only on the flux shape but also its magnitude, which is fixed by the power constraint (actually some other normalization constraint could be used just as well),

$$H \cdot \Phi = P = [\Sigma_f \psi] \cdot \Phi$$

Thus we have

$$\frac{\partial R}{\partial \psi} = \Phi \cdot h \quad \text{IV-23}$$

$$\frac{\partial R}{\partial \Phi} = [h \cdot \psi]_{r,E,\Omega} ,$$

$$\frac{\partial R}{\partial N} = 0$$

$$\left( \frac{\partial H}{\partial \psi} \Phi \right)^* = \Phi \Sigma_f$$

The problem is again a static one. The appropriate adjoint equations are now

$$\begin{bmatrix} (L^* - \lambda F^*) & \Phi \Sigma_f \\ 0 & [\Sigma_f \psi]_{r,E,\Omega} \end{bmatrix} \begin{bmatrix} \Gamma^* \\ P^* \end{bmatrix} = - \begin{bmatrix} \Phi h \\ [h \psi]_{r,E,\Omega} \end{bmatrix} \quad \text{IV-24}$$

$$\therefore P^* = - \frac{[h \psi]_{r,E,\Omega}}{[\Sigma_f \psi]_{r,E,\Omega}} \quad \text{IV-25}$$

and substituting the expression for  $P^*$  into the adjoint shape equation gives

$$\begin{aligned} (L^* - \lambda F^*) \Gamma^* &= \Sigma_f(r,E) \frac{\Phi [h \cdot \psi]_{r,E,\Omega}}{[\Sigma_f \psi]_{r,E,\Omega}} - \Phi \cdot h \\ (L^* - \lambda F^*) \Gamma^* &= R \left\{ \frac{\Sigma_f(r,E)}{[\Sigma_f \psi]_{r,E,\Omega}} - \frac{h(r,E)}{[h(r,E) \psi]_{r,E,\Omega}} \right\} \quad \text{IV-26} \end{aligned}$$

The above adjoint equation for a linear response functional is applicable to a static eigenvalue problem in which the normalization of

the flux is fixed, a case which has not been addressed with the previous static generalized perturbation method! Thus we see that the preceding developments have not only extended GPT to include time-dependent, neutron and nuclide fields, but have also enlarged the class of responses which can be addressed with the static theory, as a special case.

As a third example, consider the case when the response is a nuclide field response for which the neutron field is fixed. We then have

$$R = [\underline{h} \cdot \underline{N}]_{r,t} \quad \text{IV-27}$$

$$\frac{\partial R}{\partial \psi} = \frac{\partial R}{\partial \Phi} = 0, \text{ and}$$

$$\frac{\partial R}{\partial \underline{N}} = \underline{h}(r,t) \quad \text{IV-28}$$

The adjoint equation is

$$\underline{M}^* \underline{N}^* = - \frac{\partial}{\partial t} \underline{N}^* - \underline{h}(r,t) \quad \text{IV-29}$$

$$\underline{N}^*(r, t_f) = 0$$

and the corresponding sensitivity coefficient is

$$S_{\alpha}(\rho) = \frac{\alpha}{R} \underline{N}^* \frac{\partial}{\partial \alpha(\rho)} \underline{M} \underline{N} \quad \text{IV-30}$$

The above equation for a nuclide field not coupled to a neutron field has been derived previously by Williams and Weisbin using a variational principle (35). If R is further restricted to be a final-time functional (recall from Chapter II that a final-time response gives rise to a final condition rather than a fixed source), then,

$$\underline{M^*N^*}(r,t) = - \frac{\partial}{\partial t} \underline{N^*}(r,t) \quad \text{IV-31}$$

$$\underline{N^*}(r,t_f) = \underline{h}(r) , \quad \text{IV-32}$$

These equations were originally published by Gandini (15), and can be seen to be a special case of a more general development.

One can easily think of even more general time-dependent examples in which all three adjoint functions are involved simultaneously, though with no coupling between the flux and nuclide adjoints. For instance in the second example if the response were evaluated in the future ( $t_f \neq t_0$ ) and  $h$  were a function of  $\underline{N}$  (as a macro cross section), then a perturbation in the transmutation operator at  $t = t_0$  could affect the nuclide field in a manner that would perturb the response even without perturbing the flux, since  $h$  could change. In this case  $\underline{N^*}$  is not zero, nor are  $\Gamma^*$  and  $P^*$ . However for now we will be mostly interested in the case of a nuclide-field response, Eq. IV-27. This response is very common and appears to be the type to which the uncoupled formalism is most applicable.

Notice that Eq. IV-29 is simply the adjoint equation (not the first-order adjoint equation) to the reference state transmutation equation; i.e., if not for the nonlinearity introduced by the flux, Eq. IV-29 would be the exact adjoint equation to Eq. III-4. This observation suggests an alternate interpretation of the uncoupled nuclide adjoint equation — if we consider the transmutation equation as a *linear equation*, in which the flux field appears as *input data* (just as a cross section is input), then we would obtain Eq. IV-29 as the appropriate

adjoint equation. In other words the flux is treated as an independent rather than a dependent variable. When will such an approximation be valid? Surprisingly, there are quite a few practical examples when just this assumption is made. For example, in design scoping studies sometimes a detailed reference depletion calculation will be done in which the flux values are computed and saved. These values can then be input into other calculations that only compute the nuclide field (for example, using the ORIGEN code) to examine the effects of perturbations to the reference state. Another case of interest is in analyzing an irradiation experiment. If a small sample of some nuclide is irradiated in a reactor for some period of time, then chemical analysis of the products built up can be used to draw conclusions about cross sections appearing in the buildup chains. Because of the small sample size, the flux field will not be greatly perturbed by the nuclide field of the sample. Usually the value for the flux is either measured or provided from an independent calculation. In this case the uncoupled approximation is very good, and sensitivity coefficients computed with Eq. IV-30 can provide very useful information. Details of such a study will be given in a later chapter.

Thus we can see that there are indeed cases in which the uncoupled approximation is expected to give good results. However, in the more general case, as in analyzing a power reactor, the uncoupled approximation is not adequate. We will next focus on obtaining adjoint equations for the quasi-static formulation of the burnup equations.

## C. Quasi-Static Depletion Approximation

For the derivation, we will use a variational technique described by Pomraning (10) and Stacy (36). With this method the quasi-static burnup equations in III-28, III-29, III-33, and III-13 are treated as constraints on the response defined in Eq. IV-2, and as such are appended to the response functional using Lagrange multipliers. We will specifically examine the case in which the shape function is obtained by solving the lambda-mode eigenvalue equation, rather than the case in which  $\psi$  is obtained from a control variable (" $N_c$ ") search. The two cases are quite similar, the only difference being a "k-reset." (Eq. IV-48 illustrates the mathematical consequence of the reset.) Let us consider the following functional

$$\begin{aligned}
 K[\underline{N}, \psi_i, \Phi_i, \alpha, \lambda, h] &= R[\underline{N}, \psi_i, \Phi_i, h] \\
 &+ \sum_{i=1}^T \int_{t_i^+}^{t_i^-} dt \underline{N}^*(\hat{r}, t) \left( [\psi_i]_{E, \Omega} \Phi_i + \underline{D} - \frac{\partial}{\partial t} \underline{N}(\hat{r}, t) + \underline{C} \right) dt \\
 &- \sum_{i=1}^T \left[ \Gamma_i^*(\hat{\rho}) \left( L(\underline{N}_i) - \lambda_i F(\underline{N}_i) \right) \psi_i(\hat{\rho}) \right]_{\Omega, E, V} \\
 &- \sum_{i=1}^T P_i^* \left( [\psi_i]_{\Omega, E, V} \Phi_i - P_i \right) - \sum_{i=1}^T a_i \left( [\psi_i]_{\Omega, E, V} - 1 \right) \quad \text{IV-33}
 \end{aligned}$$

where

$T$  = number of broad time intervals in the quasi-static calculation,

$$\underline{N}_i = \underline{N}(\hat{r}, t_i), \text{ and}$$

$\underline{N}^*(\hat{r}, t)$ ,  $\Gamma_i^*(\rho)$ ,  $P_i^*$  and  $a_i$  are the Lagrange multipliers.

If  $P_i^*$  and  $\Gamma_i^*$  are set to zero and space dependence ignored, then the functional in Eq. IV-33 reduces to the same one discussed in ref. 33, which was used to derive the uncoupled, nuclide adjoint equation in Eq. IV-29.

Note that if  $\underline{N}$ ,  $\psi_i$ , and  $\phi_i$  are exact solutions to the quasi-static burnup equations, then

$$K = R \tag{IV-34}$$

In general, an alteration in some data parameter  $\alpha$  will result in

$$K \rightarrow K'[\underline{N}', \psi_i', \phi_i', \alpha', \lambda', h'] , \tag{IV-35}$$

where the prime variables refer to their perturbed values. Again, if  $\underline{N}'$ ,  $\psi_i'$ ,  $\phi_i'$  are exact solutions to the *perturbed* quasi-static equations,

$$K' = R' . \tag{IV-36}$$

Expanding  $K'$  about the unperturbed state, and neglecting second-order terms,

$$K' = K + \left[ \frac{\partial K}{\partial \alpha} \delta \alpha + \frac{\partial K}{\partial \underline{N}} \delta \underline{N} + \frac{\partial K}{\partial h} \delta h + \sum_i \left( \frac{\partial K}{\partial \psi_i} \delta \psi_i + \frac{\partial K}{\partial \phi_i} \delta \phi_i + \frac{\partial K}{\partial \lambda_i} \delta \lambda_i \right) \right] \tag{IV-37}$$

If we can force the quantities  $\partial K/\partial N$ ,  $\partial K/\partial \psi_i$ ,  $\partial K/\partial \phi_i$ ,  $\partial K/\partial \lambda_i$  to vanish, then using Eqs. IV-34, 36, and 37,

$$\delta R = \left[ \frac{\partial K}{\partial \alpha} \delta \alpha + \frac{\partial K}{\partial \underline{h}} \delta \underline{h} \right]_{\rho}, \quad \text{IV-38}$$

or

$$\delta R/R = \left[ \frac{\alpha}{R} \left( \frac{\partial K}{\partial \alpha} + \frac{\partial R}{\partial \underline{h}} \frac{\partial \underline{h}}{\partial \alpha} \right) \frac{\delta \alpha}{\alpha} \right]_{\rho}. \quad \text{IV-39}$$

From Eq. IV-39, it is obvious that the sensitivity coefficient for  $\alpha$  is simply

$$S(\hat{\rho}) = \frac{\alpha}{R} \left( \frac{\partial K}{\partial \alpha} + \frac{\partial R}{\partial \underline{h}} \frac{\partial \underline{h}}{\partial \alpha} \right). \quad \text{IV-40}$$

The partial derivatives in Eq. IV-40 are trivial to evaluate, and therefore the problem of sensitivity analysis for the quasi-static burnup equations reduces to finding the appropriate stationary conditions on the K-functional. We will now set upon determining the required Euler equations, which will correspond to the adjoint field equations.

Consider first the functional derivative with respect to  $\phi_i$

$$\frac{\partial K}{\partial \phi_i} = \frac{\partial R}{\partial \phi_i} + \int_{t_i}^{t_{i+1}^-} \left[ \underline{N}^* [\psi_i \underline{R}]_{E,\Omega} \underline{N} \right]_V dt - P_i^* [\psi_i \underline{\sigma}_f \underline{N}_i]_{E,\Omega,V} \quad \text{IV-41}$$

In order for this expression to vanish, we should choose

$$P_i^* = \frac{\int_{t_i^+}^{t_{i+1}^-} \left[ \underline{N}^* [\underline{\psi}_i \underline{R}]_{\Omega, E} \underline{N} \right]_V dt + \frac{\partial R}{\partial \phi_i}}{[\underline{\psi}_i \underline{\sigma}_f \underline{N}_i]_{\Omega, E, V}} \quad \text{IV-42}$$

Now examine the term  $\partial K / \partial \psi_i$ , employing the commutative property of adjoint operators,

$$\begin{aligned} \frac{\partial K}{\partial \psi_i} &= \frac{\partial R}{\partial \psi_i} - \left( L^*(\underline{N}_i) - \lambda_i F^*(\underline{N}_i) \right) \Gamma_i^* - P_i^* \phi_i \underline{\sigma}_f \underline{N}_i + \\ &\quad \phi_i \int_{t_i^+}^{t_{i+1}^-} \underline{N}^* \underline{R} \underline{N} dt - a_i \end{aligned} \quad \text{IV-43}$$

with  $L^*$ ,  $F^*$   $\equiv$  adjoint operators to  $L$  and  $F$ , respectively. The vanishing of this term implies that (assuming the "standard" adjoint boundary conditions)

$$\left\{ L^*(\underline{N}_i) - \lambda_i F^*(\underline{N}_i) \right\} \Gamma_i^*(\hat{\rho}) = Q_i^*, \quad \text{IV-44}$$

where

$$Q_i^*(\hat{\rho}) = \frac{\partial R}{\partial \psi_i} + \phi_i \int_{t_i^+}^{t_{i+1}^-} \underline{N}^*(\hat{r}, t) \underline{R}(\sigma) \underline{N}(\hat{r}, t) dt - \phi_i P_i^* \underline{\sigma}_f \underline{N}_i - a \quad \text{IV-45}$$

At this point it should be noted that Eqs. IV-44 and III-28 demand that the flux shape function be orthogonal to the adjoint source; i.e.,

$$[\psi_i Q_i^*]_{\Omega, E, V} = 0, \text{ at all } t_i.$$

From Eqs. IV-45 and IV-42 it is easily shown that this condition requires

$$\left[ \psi_i \frac{\partial R}{\partial \psi_i} \right]_{E, \Omega, V} - \phi_i \left[ \frac{\partial R}{\partial \phi_i} \right]_{E, \Omega, V} = a_i \quad \text{IV-46}$$

which fixes the value of "a." For most cases of practical interest, this term is zero. For example if R is bilinear in  $\psi_i$  and  $\phi_i$ , or is a bilinear ratio, then "a" will vanish.

The term  $\partial K / \partial \lambda_i$  is evaluated to be

$$\frac{\partial K}{\partial \lambda_i} = [\Gamma_i^*(\hat{\rho}) F(N_i) \psi_i]_{\Omega, E, V} = 0, \quad \text{IV-47}$$

which forces  $\Gamma_i^*(\hat{\rho})$  to be orthogonal to the fission source at  $t = t_i$ . This condition requires that  $\Gamma_i^*$  contain no fundamental mode from the homogeneous solution. More specifically, if  $\Gamma_p^*$  is a solution to Eq. IV-44 and  $\Gamma_p^* \perp \phi_H^*$ , where  $\phi_H^*$  is the fundamental solution to the homogeneous equation, then  $\Gamma_p^* + b\phi_H^*$  is also a solution for all b. However, Eq. IV-47 fixes the value of "b" to be zero, so that  $\Gamma_i^* = \Gamma_p^*$ .

This is true only for the case in which there is no k-reset (i.e.,  $\lambda$  is allowed to change with data perturbations). For the case in which  $\lambda$  is made invariant by adjusting a control variable  $N_c$ , it is easily shown that the proper orthogonality condition is

$$\left[ \Gamma_i^*(\hat{\rho}), \frac{\partial}{\partial N_c} (L(N_i) - \lambda_i F(N_i)) \psi_i \right]_{\Omega, E, V} = \frac{\partial K}{\partial N_c} = 0 \quad \text{IV-48}$$

Now the value of "b" is *not* zero, but is given by

$$\frac{\left[ \Gamma_p^* \frac{\partial}{\partial N_c} (L - \lambda P) \psi \right]_{\Omega, E, V}}{\left[ \phi^* \frac{\partial}{\partial N_c} (L - \lambda P) \psi \right]_{\Omega, E, V}} \quad \text{IV-49}$$

Thus the effect of adjusting a control variable is to "rotate"  $\Gamma_i^*$  so that it will have some fundamental component. The specific projection along  $\phi^*$  depends on the specific control variable.

The Euler condition corresponding to a variation in  $\underline{N}(r, t)$  is slightly more complex than for the other variables. Rather than simply taking the partial functional derivative, it will be more instructive to consider the differential (variation) of  $K$  with respect to  $\delta \underline{N}$

$$\begin{aligned} \delta K[\delta \underline{N}] &= \left[ \frac{\partial R}{\partial \underline{N}}, \delta \underline{N} \right]_{\rho} \\ &+ \sum_{i=1}^T \int_{t_i^+}^{t_{i+1}^-} dt \left[ \delta \underline{N}(\hat{r}, t) \left( [\psi_i R^*]_{\Omega, E} \phi_i + \underline{D}^* + \frac{\partial}{\partial t} \right) \underline{N}^* \right]_V \\ &- \sum_{i=1}^T \left[ (\underline{N}_{i+1}^* \delta \underline{N}_{i+1}^- - \underline{N}_i^* \delta \underline{N}_i^+) \right]_V \\ &- \sum_{i=1}^T \left[ \delta \underline{N}_i \left[ \Gamma_i^* \frac{\partial}{\partial \underline{N}_i} (L(N_i) - \lambda F(N_i)) \psi_i \right]_{\Omega, E} \right]_V \\ &- \sum_{i=1}^T P_i^* \phi_i \left[ \delta \underline{N}_i [\psi_i \sigma_f]_{\Omega, E} \right]_V, \end{aligned} \quad \text{IV-50}$$

where  $\underline{N}_{i+1}^{*-} = \underline{N}^*(\hat{r}, t_{i+1}^-)$ , etc.; and  $\underline{R}^* \equiv$  transpose  $\underline{R}$ ,  $\underline{D}^* \equiv$  transpose  $\underline{D}$  (i.e.,  $\underline{R}^*$  and  $\underline{D}^*$  are the adjoint operators to  $\underline{R}$  and  $\underline{D}$ ).

This variation will be stationary if the following conditions are met. The first two expressions on the right-hand side of Eq. IV-50 will vanish if

$$\Phi_i[\psi_i R^*]_{\Omega, E} \underline{N}^* + \underline{D}^* \underline{N}^* = - \frac{\partial}{\partial t} \underline{N}^* - \left[ \frac{\partial R}{\partial \underline{N}} \right]_{\Omega, E}, \text{ for } t_i^+ < t < t_{i+1}^-, \quad \text{IV-51}$$

which can be written

$$\underline{M}^* \underline{N}^* + \underline{C}^* = - \frac{\partial}{\partial t} \underline{N}^*, \quad \text{IV-52}$$

where

$$\underline{C}^* = \left[ \frac{\partial R}{\partial \underline{N}} \right]_{\Omega, E} \quad \text{IV-53}$$

This equation is valid for the *open* interval  $(t_i, t_{i+1})$ . But the question of the behavior of  $\underline{N}^*(\hat{r}, t)$  at the time boundaries  $t_i$  has not yet been answered. The remaining terms in Eq. IV-50 will provide the necessary boundary conditions for each broad time interval. These terms may be written as

$$\sum_{i=1}^T \left[ \delta \underline{N}_i \left\{ \underline{N}_{i+1}^{*+} - \left[ \Gamma_i^* \frac{\partial}{\partial \underline{N}_i} (L - \lambda F) \psi_i \right]_{\Omega, E} - \Phi_i P_i^* [\psi_i \sigma_f]_{\Omega, E} \right\} - \delta \underline{N}_{i+1} \underline{N}_{i+1}^{*-} \right]_V \quad \text{IV-54}$$

where we have employed the continuity condition on the nuclide field,

$$\underline{N}_i = \underline{N}_i^- = \underline{N}_i^+ .$$

Expanding the summation, we get

$$\begin{aligned} & \left[ \delta \underline{N}_0 \left\{ \underline{N}_0^* - \left[ \Gamma_0^* \frac{\partial}{\partial \underline{N}_0} (L - \lambda F) \psi_0 + P_0^* \Phi_0 \psi_0 \underline{\sigma}_f \right]_{\Omega, E} \right\} \right. \\ & + \delta \underline{N}_1 \left\{ (\underline{N}_1^+ - \underline{N}_1^-) - \left[ \Gamma_1^* \frac{\partial}{\partial \underline{N}_1} (L - \lambda F) \psi_1 + P_1^* \Phi_1 \psi_1 \underline{\sigma}_f \right]_{\Omega, E} \right\} \\ & + \dots - \delta \underline{N}_f \underline{N}_f^* \left. \right]_V \end{aligned} \quad \text{IV-55}$$

By allowing a discontinuity in the nuclide adjoint field we can make all the terms containing  $\delta \underline{N}_i$  vanish, except at the end points  $t = 0$  and  $t = t_f$ . Therefore we assert the following property of  $\underline{N}^*(r, t)$  at the time boundaries,

$$\begin{aligned} \underline{N}^*(\hat{r}, t_i^-) &= \underline{N}^*(\hat{r}, t_i^+) - \left[ \Gamma_i^* \frac{\partial}{\partial \underline{N}_i} (L - \lambda F) \psi_i + \Phi_i P_i^* \underline{\sigma}_f \psi_i \right]_{\Omega, E} \\ &= \underline{N}^*(\hat{r}, t_i^+) - [\Gamma_i^* \underline{\beta}_i + P_i^* \underline{\Pi}_i]_{\Omega, E} , \end{aligned} \quad \text{IV-56}$$

where

$$\begin{aligned} \underline{\beta}_i &= \frac{\partial}{\partial \underline{N}_i} (L(\underline{N}_i) - \lambda_i F(\underline{N}_i)) \psi_i \\ \underline{\Pi}_i &= \Phi_i \underline{\sigma}_f \psi_i \end{aligned} \quad \text{IV-57}$$

The second term on the right-hand side of Eq. IV-56 represents a "jump condition" on  $\underline{N}^*$  at  $t = t_i^-$ ; its value depends on the magnitude of the other adjoint variables  $\Gamma_i^*$  and  $P_i^*$ . Essentially,  $\Gamma_i^* \beta_i$  and  $P_i^* \Pi_i$  are sensitivity coefficients to changes in  $\underline{N}_i$ .

The term in Eq. IV-55 containing  $\delta N_f$  will vanish if we fix the final condition of  $\underline{N}^*$  to be

$$\underline{N}^*(\hat{r}, t_f) = 0. \quad \text{IV-58}$$

(For responses which are delta functions in time, the final condition will be inhomogeneous — see next section.)

With all these restrictions placed on  $\underline{N}^*$ , the summation in Eq. IV-55 reduces to a single expression,

$$\left[ \delta N_0 \left\{ N_0^* - [\Gamma_0^* \beta_0 + P_0^* \Pi_0]_{\Omega, E} \right\} \right]_V = \left[ \delta N_0 N^*(t_0^-) \right]_V \quad \text{IV-59}$$

From this equation we can define a sensitivity coefficient for the initial condition of nuclide  $m$  to be

$$S_0^m = N_0^m \left\{ N_0^{m*} - [\Gamma_0^* \beta_0^m + P_0^* \Pi_0^m]_{\Omega, E} \right\} = N_0^m N^{m*}(t_0^-) \quad \text{IV-60}$$

For no change in the initial condition of the nuclide field, Eq. IV-59 will also vanish. To be general, however, we will not make this assumption, and will retain the expression in Eq. IV-60 as part of the sensitivity coefficient.

This rather involved development has provided the adjoint-field equations for the quasi-static approximation. We have found that there exist adjoint equations corresponding to the nuclide transmutation equation, to the flux-shape equation (transport equation), and to the power-constraint equation. In addition, we have found that it is convenient to ascribe additional restrictions on the adjoint fields — namely, that  $\Gamma_i^*$  be orthogonal to the fission source and that  $\underline{N}^*$  be discontinuous at each time boundary. The adjoint field equations are coupled, linear equations which contain the unperturbed forward values for  $\underline{N}$ ,  $\psi_i$ , and  $\phi_i$ . These equations are repeated below:

Adjoint flux-shape equation

$$\left\{ L^*(\underline{N}_i) - \lambda_i F^*(\underline{N}_i) \right\} \Gamma_i^* = Q_i^* \quad \text{IV-61}$$

at  $t = t_i$ .

Adjoint flux-normalization equation:

$$P_i^* = \frac{\int_{t_i^+}^{t_{i+1}^-} \left[ \underline{N}^* [\psi_i R]_{\Omega, E} \underline{N} \right]_V dt + \frac{\partial R}{\partial \phi_i}}{[\psi_i \sigma_f \underline{N}_i]_{\Omega, E, V}}, \quad \text{at } t = t_i \quad \text{IV-62}$$

Adjoint transmutation equation:

$$-\frac{\partial}{\partial t} \underline{N}^*(\hat{r}, t) = \underline{M}^*(\phi_i, \psi_i) \underline{N}^*(\hat{r}, t) + \underline{C}^*(\hat{r}, t), \quad t \in (t_i, t_{i+1}) \quad \text{IV-63}$$

$$\underline{N}^*(\hat{r}, t_i^-) = \underline{N}^*(\hat{r}, t_i^+) - [\Gamma_i^* \beta_i + P_i^* \Pi_i]_{\Omega, E}, \text{ at } t = t_i, i \neq f \quad \text{IV-64}$$

$$\underline{N}^*(\hat{r}, t_f) \equiv \underline{N}_f(\hat{r}) = 0, \text{ at } t = t_f \quad \text{IV-65}$$

In the limit, as the length of the broad time-step goes to zero, the flux becomes a continuous function of time and there is no jump condition on the nuclide adjoint. For this special case, if the fundamental mode approximation is made for the spatial shape of the flux, the energy dependence expressed in few-group formalism, and the components of  $\underline{N}$  limited to a few isotopes important to thermal reactor analysis, then the equations reduce to a form similar to those derived by Harris (17). Harris' equations are in fact simply an approximation to the time-continuous adjoint system to Eq. IV-14.

The adjoint field equations previously derived were for an arbitrary response. A specific type of response which is often of interest is the type originally considered by Gandini in his derivation of the uncoupled, nuclide adjoint equation, discussed earlier,

$$R = R[\underline{N}_f, h] = R[\underline{N}(\hat{r}, t) \delta(t - t_f), h]. \quad \text{IV-66}$$

i.e., the response is a delta function in time at  $t = t_f$ . In this case, the adjoint source is equivalent to a fixed final condition, and the adjoint field equations will simplify by

$$\underline{C}^*(\hat{r}, t) = 0 \quad \text{for } t < t_f \quad \text{IV-67}$$

$$\underline{N}_f^* = \frac{\partial R}{\partial \underline{N}_f} \quad \text{at } t = t_f \quad \text{IV-68}$$

$$\frac{\partial R}{\partial \Phi_i} = \frac{\partial R}{\partial \psi_i} = 0 \quad \text{at } t = t_i \quad \text{IV-69}$$

If the values for the variables  $P_i^*$  and  $\Gamma_i^*$  are also small (i.e., the effect of flux perturbation is negligible), then the discontinuity in  $\underline{N}^*$  at  $t_i$  will be small, and the nuclide adjoint equation reduces to the uncoupled form in Eqs. IV-31 and 32.

#### D. Initial-Value Approximation

The previous developments were aimed at deriving adjoint and perturbation equations for application to the long-range time scale. We will now present briefly an adjoint equation for the intermediate-range problem discussed in Chapter III. The derivation is very straightforward — since Eq. III-36 is the linearized form of the equation of interest — which is the initial-value form for the burnup equation, the first order adjoint system is

$$\begin{bmatrix} B^*(\underline{N}) & \left(\frac{\partial MN}{\partial \phi}\right)^* \\ \left(\frac{\partial B}{\partial \underline{N}} \phi\right)^* & \underline{M}(\phi)^* \end{bmatrix} \begin{bmatrix} \Gamma^* \\ \underline{N}^* \end{bmatrix} = -\frac{\partial}{\partial t} \begin{bmatrix} \frac{1}{v} \Gamma^* \\ \underline{N}^* \end{bmatrix} - \begin{bmatrix} \frac{\partial R}{\partial \phi} \\ \frac{\partial R}{\partial \underline{N}} \end{bmatrix} \quad \text{IV-70}$$

with the final conditions

$$\Gamma^*(T_f) = 0 \quad \text{IV-71}$$

$$\underline{N}^*(T_f) = 0 \quad \text{IV-72}$$

(Note: the term  $(\partial B / \partial \underline{N} \phi)^* \Gamma^*$  in the  $\underline{N}^*$  equation is actually integrated over  $E, \Omega$ , though not explicitly shown).

Using the property that the adjoint of a product of operators is the inverse product of the adjoint operators (and also recall that functions are self-adjoint), we can write

$$\left( \frac{\partial \underline{M} \underline{N}}{\partial \phi} \right)^* \underline{N}^* = \underline{N} \left( \frac{\partial \underline{M}}{\partial \phi} \right)^* \underline{N}^*$$

and

$$\left( \frac{\partial B}{\partial \underline{N}} \phi \right)^* \Gamma^* = \phi \left( \frac{\partial B}{\partial \underline{N}} \right)^* \Gamma^*$$

so that Eq. IV-70 can be expressed

$$\begin{bmatrix} B^* & \left( \underline{N} \frac{\partial M^*}{\partial \phi} \right) \\ \left( \phi \frac{\partial B^*}{\partial \underline{N}} \right) & M^* \end{bmatrix} \begin{bmatrix} \Gamma^* \\ \underline{N}^* \end{bmatrix} = - \frac{\partial}{\partial t} \begin{bmatrix} \frac{1}{v} \Gamma^* \\ \underline{N}^* \end{bmatrix} - \begin{bmatrix} \frac{\partial R}{\partial \phi} \\ \frac{\partial R}{\partial \underline{N}} \end{bmatrix} \quad \text{IV-73}$$

Again, one should realize that the term  $\phi \partial B^* / \partial \underline{N} \Gamma^*$  is actually an integral over  $E$  and  $\Omega$ . As would be expected, the adjoint equations to a system of initial-value equations is a system of final-value equations. As usual, the source term can be transformed to an inhomogeneous final condition if  $R$  is a delta function in time. An example application of this equation would be to analyze a "flux tilt" response, defined as the ratio of the flux at one location to the flux at another at some specified time:

$$R = \frac{[\phi(r_1, E, \Omega, T_f)]_{E, \Omega}}{[\phi(r_2, E, \Omega, T_f)]_{E, \Omega}} = \frac{[\phi(\rho)\delta(r - r_1)\delta(t - T_f)]_\rho}{[\phi(\rho)\delta(r - r_2)\delta(t - T_f)]_\rho} \quad \text{IV-74}$$

It is usually desirable to minimize a response of this type. In this case,

$$\frac{\partial R}{\partial N} = 0 ,$$

and the final condition on the neutron field is

$$\frac{\partial R}{\partial \phi} = R \cdot \left\{ \frac{\phi(r_1, E, \Omega, T_f)\delta(r - r_1)}{[\phi(r_1, E, \Omega, T_f)]_{E, \Omega}} - \frac{\phi(r_2, E, \Omega, T_f)\delta(r - r_2)}{[\phi(r_2, E, \Omega, T_f)]_{E, \Omega}} \right\} \quad \text{IV-75}$$

which corresponds to point sources located at positions  $r_1$  and  $r_2$ , respectively. The sensitivity coefficient for the flux tilt to some data  $\alpha$  is

$$S_\alpha(\rho) = \frac{\alpha}{R} \left\{ \Gamma^*(\rho) \frac{\partial B}{\partial \alpha} \phi + \underline{N}^* \frac{\partial}{\partial \alpha} \underline{MN} \right\} . \quad \text{IV-76}$$

## CHAPTER V

### SOLUTION METHODS FOR THE ADJOINT BURNUP EQUATIONS

In this chapter we will discuss techniques developed for solving the adjoint burnup equations for the uncoupled and coupled quasi-static cases.

#### A. Uncoupled, Nuclide Adjoint Solution

In the uncoupled case, one is only concerned with solving the nuclide adjoint equation (not the neutron-field equation) which is simply a system of simultaneous, linear, first-order equations. Capability for solving the forward equations was already available at ORNL in the ORIGEN computer code, and therefore it was necessary only to make modifications to this basic code to allow for adjoint solutions. An overview of the basic calculational method is given below.

The burnup equation is a statement of mass balance for a radioactive nuclide field subjected to a neutron flux. The equation for nuclide species  $i$  can be written:

$$\frac{dN_i}{dt} = -(\sigma_{ai}\phi + \lambda_i)N_i + \sum_{j \neq i} (\sigma_{j \rightarrow i}\phi + \lambda_{j \rightarrow i})N_j \quad V-1$$

In matrix notation, the above equation is:

$$\underline{M}(\alpha_{j \rightarrow i})\underline{N} = \frac{\partial}{\partial t} \underline{N} ,$$

$\alpha_{j \rightarrow i}$  = probability per unit time that isotope  $i$  will be produced from isotope  $j$ , and  $\alpha_{ii} = -\sum_j \alpha_{i \rightarrow j}$ .

In Eq. V-1, the value for  $\underline{N}$  can be found with the matrix exponential technique as

$$\underline{N}(t) = \exp(\underline{M}t) \underline{N}_0, \quad \text{V-2}$$

where  $\exp(\underline{M}t)$  is the time dependent matrix given by the infinite series

$$\underline{I} + \underline{M}t + \frac{\underline{M}^2 t^2}{2!} \dots \equiv \underline{B}(t). \quad \text{V-3}$$

Of course in reality the series is truncated at some finite number of terms dictated by the tolerance placed on  $\underline{N}(t)$ . The computer code ORIGEN solves the burnup equations using this method, and a discussion of the numerical procedures involved in its implementation can be found in reference (26).

Note that the matrix  $\underline{B}(t)$  is independent of the initial conditions  $\underline{N}_0$ ; therefore, in theory it is possible to obtain a solution for a given  $\underline{M}(\phi)$  that does not depend on the initial reactor configuration. Then the time-dependent nuclide field is

$$\underline{N}(t) = \underline{B}(t)\underline{N}_0 \quad \text{for any } \underline{N}_0, \quad \text{V-4}$$

Unfortunately the nuclear data matrix  $\underline{B}$  is problem dependent (through the flux) and is too large ( $\sim 800$  by  $800$  words for each time step in ORIGEN) to be used efficiently. It is more advantageous to recalculate  $\underline{N}(t)$  for each  $\underline{N}_0$ .

As previously discussed the adjoint burnup equation is

$$-\frac{d}{dt} \underline{N}^* = \underline{M}^T \underline{N}^* . \quad \text{V-5}$$

Equation V-5 can be expressed in a form compatible with the present ORIGEN computational technique (i.e., a *positive* time derivative) by making a change of variable:

$$t' = t_f - t$$

$$-\frac{d}{dt} = \frac{d}{dt'} \quad \text{V-6}$$

$$\underline{N}^*(t_f) = \underline{N}^*(t' = 0) \quad \text{V-7}$$

Then the adjoint solution is merely

$$\underline{N}^*(t') = e^{\underline{M}^T t'} \underline{N}^*(t' = 0), \quad 0 < t < t_f \quad \text{V-8}$$

$$\underline{N}^*(t) = \underline{N}^*(t_f - t') , \quad \text{V-10}$$

$$\underline{N}^*(t_f) = \underline{N}^*(t' = 0) \equiv \underline{N}^*_F$$

Equation V-8 is the same solution obtained by the forward ORIGEN code, except the data matrix is transposed.

Equation V-8 can be written as

$$\underline{N}^*(t) = \exp [\underline{M}^T (t_f - t)] \underline{N}^*_F . \quad \text{V-11}$$

It is easy to show that

$$\exp(\underline{A}^T) = (\exp \underline{A})^T,$$

and therefore

$$\underline{N}^*(t) = \underline{B}^T(t_f - t)\underline{N}_f^* \quad \text{V-12}$$

It is interesting to note that

$$\begin{aligned} \underline{N}^T(t)\underline{N}^*(t) &= [\underline{e}^{\underline{M}t}\underline{N}_0]^T [\underline{e}^{\underline{M}^T(t_f - t)}\underline{N}_f^*] \\ &= \underline{N}_0^T [\underline{e}^{\underline{M}^T(t - t + t_f)}] \underline{N}_f^* \\ &= \underline{N}_f^{*T} e^{\underline{M}t_f} \underline{N}_0 = \underline{N}_f^{*T} \underline{N}_f = R \quad \text{V-13} \end{aligned}$$

This result was derived in Chapter II as a conservation law.

One of the more puzzling difficulties encountered in providing adjoint capability for the ORIGEN code arose in the treatment of nearly stable (both in decay and in reaction) product nuclides such as  $\text{He}^4$ ,  $\text{H}^2$ , etc. When the parent-daughter relation among nuclides is reversed by transposing  $\underline{M}$ , it is possible for nuclides which previously had no daughters to have transmutation products, since their parents are then identified as daughters. The presence of a zero (or very small) transition probability for a nuclide with daughter products causes a series of numerical problems in ORIGEN, the final result being a "divide check."

The solution to this problem is discussed below, for a hypothetical decay chain of three nuclides — A, B, C — the last of which is stable.

We assume the appropriate burnup equations are the following:

$$\begin{bmatrix} -\lambda_A & 0 & 0 \\ \lambda_{AB} & -\lambda_B & 0 \\ 0 & \lambda_{BC} & 0 \end{bmatrix} \begin{bmatrix} N_A \\ N_B \\ N_C \end{bmatrix} = \frac{d}{dt} \begin{bmatrix} N_A \\ N_B \\ N_C \end{bmatrix} \quad \text{V-14}$$

The adjoint system is

$$\begin{bmatrix} -\lambda_A & \lambda_{AB} & 0 \\ 0 & -\lambda_B & \lambda_{BC} \\ 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} N_A^* \\ N_B^* \\ N_C^* \end{bmatrix} = \frac{-d}{dt} \begin{bmatrix} N_A^* \\ N_B^* \\ N_C^* \end{bmatrix} \quad \text{V-15}$$

The equation for  $N_C^*$  is

$$\frac{d}{dt} N_C^* = 0 \quad N_C^* = \text{constant} . \quad \text{V-16}$$

Therefore  $N_C^* = (\underline{h})_C$ , where  $\underline{h}$  is the input realization vector. Since this value is fixed by the specified final condition, the calculation of stable-nuclide adjoints is omitted from ORIGEN-A.

Considering Eq. V-15 again, and omitting the equation for  $N_C^*$ ,

$$\begin{bmatrix} -\lambda_A & \lambda_{AB} \\ 0 & -\lambda_B \end{bmatrix} \begin{bmatrix} N_A \\ N_B \end{bmatrix} = \frac{-d}{dt} \begin{bmatrix} N_A \\ N_B \end{bmatrix} - \begin{bmatrix} 0 \\ (\underline{h})_C \lambda_{BC} \end{bmatrix} \quad \text{V-17}$$

Thus we see that a stable nuclide can give rise to a fixed source term in the adjoint-burnup equation, depending on the value of  $h$ .

In summary, Eqs. V-8, V-9 and V-10 can be incorporated into the ORIGEN to allow uncoupled, nuclide adjoint solutions, with four modifications:

- (a) enter "initial" charge as  $N_f^*$ , the response realization vector,
- (b) reverse the parent-daughter relationship among nuclides,
- (c) reverse flux and time arrays,
- (d) interpret all results backwards in the time variable.

With these modifications, as well as several changes in the numerical methods, the ORIGEN code is called ORIGEN-A, which is presently in use at ORNL. The input description for this code appears in reference (35).

#### B. Quasi-Static Solution

Solving the adjoint quasi-static equations requires not only computing the nuclide adjoint field, but also computing a special type of "generalized adjoint" function for the neutron field. The latter calculation can be quite difficult, but fortunately much work has already gone into this area as part of the ORNL static sensitivity program. After much deliberation it was decided to use the VENTURE/BURNER code system (37, 32) as a starting point for the quasi-static adjoint solution. This decision was based on the following considerations:

- (a) VENTURE/BURNER were the most up-to-date depletion codes available at ORNL and will be widely used for burnup calculations not only at ORNL but also at other installations.

(b) BURNER had an option of solving the nuclide-field equation by the matrix exponential technique, which (as previously shown) is easily adaptable to the nuclide adjoint solution.

(c) VENTURE had the capability of solving the diffusion-theory, generalized adjoint-flux equation.

(d) Modular code structure allowed independent calculational modules to be integrated into the system.

The major drawback to the VENTURE system, as far as implementing adjoint capability is concerned, was the necessity of dealing with a multitude of interface files which many times were not well formatted for an efficient adjoint solution algorithm. We will now examine a general overview of the method used to solve the adjoint quasi-static burnup equations. But before outlining a computational flow chart, it may be helpful to make some preliminary observations.

First, it is shown in Eq. IV-45 that the flux adjoint source  $Q_i^*$  at  $t_i$  depends on an integral of  $\underline{N}^*$  over the future time interval  $(t_i, t_{i+1})$  — this fact is strong incentive for solving the adjoint equations backwards in time. We will not dwell on the difficulties encountered in solving the adjoint-flux equation, other than to point out that the operator on the left-hand side of Eq. IV-44 is singular (hence the requirement that the fixed source be orthogonal to the fundamental forward eigenfunction). A discussion of the numerical methods required to solve these "generalized adjoint" equations can be found in ref. (38).

Second, notice that over any given time interval  $(t_i, t_{i+1})$ , Eq. IV-52 for the coupled nuclide adjoint is identical to Eq. IV-29 for the uncoupled case; i.e., it is a final-value equation with constant coefficients. A method for solving this equation was described earlier.

Finally, we see from Eq. IV-56 that the final value of  $\underline{N}^*$  at the end of each time interval is fixed by the "jump" condition. Its magnitude depends not only on the future behavior of  $\underline{N}^*$ , but also on  $\Gamma^*$  and  $p^*$  at the final time of the interval.

In summary, the adjoint quasi-static equations are coupled in the following manner:

- (a) the variables  $\underline{N}^*$  and  $p^*$  appear in the source term of the equation for  $\Gamma^*$ ,
- (b) the variable  $\underline{N}^*$  appears in the defining equation for  $p^*$ ,
- (c) the variables  $\Gamma^*$  and  $p^*$  appear in the "jump condition" for  $\underline{N}^*$ .

With these conditions in mind, we will now attempt to establish a suitable computational algorithm for numerical solution of the adjoint quasi-static equations. Toward this end, consider the following flow chart:

- (i) starting with the  $T$ th time interval (i.e., the last interval), solve Eq. IV-63 for the value of  $\underline{N}^*$  between  $(t_{T-1}^+, t_f^-)$ . The final value  $\underline{N}_f^*$  is fixed by Eq. IV-68.
- (ii) compute the value for  $p_{T-1}^*$  at  $t_{T-1}$  from Eq. IV-62
- (iii) compute  $Q_{T-1}^*$  using Eq. IV-45
- (iv) solve Eq. IV-61 for  $\Gamma_{T-1}^*$  at  $t_{T-1}$

- (v) with the known values for  $p^*$ ,  $\Gamma^*$ , and  $\underline{N}^*$  at  $t_{T-1}^+$ , compute the value for  $\underline{N}^*$  at  $t_{T-1}^-$  from Eq. IV-64
- (vi) using this new value for the final condition of  $\underline{N}^*$ , again solve Eq. IV-63 for the behavior of  $\underline{N}^*$  between  $(t_{T-2}^+, t_{T-1}^-)$
- (vii) etc.

This marching procedure is followed backward through all the time intervals until the values at  $t = 0$  are obtained, at which time the adjoint calculation is complete. When all the adjoint values have been obtained, the sensitivity coefficient for data variations is computed with Eq. IV-40, and for initial-value variations with Eq. IV-60.

Much progress has been made in implementing the above algorithm into the VENTURE system. The works cited below have greatly expedited the development:

(a) The VENTURE/BURNER code system developed by Vondy, Fowler, and Cunningham would already perform the forward quasi-static calculation as well as the generalized adjoint flux calculation when the current study was begun. These computations are the most numerically complex ones encountered in the adjoint algorithm, and hence the most difficult coding was essentially already done. The majority of the required programming involved interfacing between various VENTURE/BURNER calculations and combining results in the necessary manner. However, this was no trivial task and much work has been put into the effort by J. R. White (39).

(b) At the request of the author, G. W. Cunningham modified the BURNER code to allow calculation of the nuclide adjoint vector (40) (analogous to work done for ORIGEN-A).

(c) J. R. White, as part of his Master's thesis, has programmed into the VENTURE system a module called DEPTH (Depletion Perturbation Theory) (39) for applying the methodology established in this dissertation to design calculations. This module performs the  $p^*$  integration, computes the generalized adjoint source for the VENTURE  $I^*$  calculation, and accounts for the jump condition in the nuclide field.

There are still many programming details in the adjoint codes which should be resolved before the system is efficient; however, the ability does currently exist at ORNL for performing coupled depletion-perturbation calculations for final-time nuclide responses. Some results obtained with the codes are discussed in Chapter IX. Work is ongoing in this area to improve the adjoint calculational efficiency as well as to extend the capability to more general responses and to automate the computation of sensitivity coefficients. Further developments will be reported in White's thesis and in future ORNL reports.

## CHAPTER VI

### SENSITIVITY COEFFICIENTS AND UNCERTAINTY ANALYSIS FOR BURNUP CALCULATIONS

In earlier chapters, general expressions in operator notation were presented for sensitivity coefficients. This chapter will focus on deriving specific sensitivity coefficients for multi-group calculations in uncoupled and coupled burnup sensitivity analysis. In the uncoupled case, sensitivity coefficients are presented for the following types of data appearing in the transmutation operator: (a) capture, fission, and (n, 2n) multi-group cross sections; (b) decay constants (half-lives); (c) yield data; (d) initial condition of the nuclide field. For the coupled case, we will assume that the neutron-field equation corresponds to the diffusion equation, as usually done in burnup calculations. These same types of data are also considered for the coupled, quasi-static case, as well as the following data which appears only in the diffusion operator: (a) multigroup scattering cross sections, (b) multigroup transport cross section, (c) neutron yield per fission.

The notation below will be employed:

$N_{A,M}(z,t)$  = atom density in reactor zone  $z$ , at time  $t$  for a nuclide  
with  $A$  protons and  $M - A$  neutrons

$\Phi_i$  = flux normalization factor at time step  $i$

$\psi_i(z,g)$  = zone average flux in zone  $z$ , group  $g$  at broad time  
step  $i$

$\psi_i(r,g)$  = point flux at position  $r$  ( $= x,y,z$ ), group  $g$ , broad time step  $i$

$V_r$  = volume of interval  $r$

$V_z$  = volume of zone  $z$

Similar notation holds for the adjoint variables

$N_{A,M}^*(z,t), \Gamma_i^*(z,g), \Gamma_i^*(r,g)$

We assume that the required forward and adjoint values have already been computed, using one of the methods described earlier. The expressions for calculating the sensitivity coefficients using these values for a response  $R$  are summarized below.

#### A. Sensitivity Coefficients for Uncoupled Approximation

1. Multigroup Capture Cross Section,  $\sigma_{A,M}^C(z,g)$

$$S_1(z,g) = \frac{\sigma_{A,M}^C(z,g) \cdot V_z}{R} \sum_i \left\{ \Phi_i \psi_i(z,g) \int_{t_i^+}^{t_{i+1}^-} N_{A,M}(z,t) (N_{A,M+1}^*(z,t) - N_{A,M}^*(z,t)) dt \right\}$$

2. Multigroup Fission Cross Section,  $\sigma_{A,M}^f(z,g)$

$$S_2(z,g) = \frac{\sigma_{A,M}^f(z,g) \cdot V_z}{R} \sum_i \left\{ \Phi_i \psi_i(z,g) \int_{t_i^+}^{t_{i+1}^-} N_{A,M}(z,t) \right\}$$

$$\left( \sum_{K,L} \gamma_{A,M \rightarrow K,L} N_{K,L}^*(z,t) - N_{A,M}^*(z,t) \right) dt \left. \right\}$$

where  $\gamma_{A,M \rightarrow K,L} \equiv$  yield of  $N_{K,L}$  from fission of  $N_{A,M}$

3. Multigroup (n,2n) Cross Section,  $\sigma_{A,M}^{2n}(z,g)$

$$S_3(z,g) = \frac{\sigma_{A,M}^{2n}(z,g) \cdot V_Z}{R} \sum_i \left\{ \Phi_i \psi_i(z,g) \int_{t_i^+}^{t_{i+1}^-} N_{A,M}(z,t) \right. \\ \left. (N_{A,M-1}^*(z,t) - N_{A,M}^*(z,t)) dt \right\}$$

4. Decay Constant,  $\lambda_{A,M \rightarrow K,L}$

$$S_4(z) = \frac{\lambda_{A,M \rightarrow K,L} \cdot V_Z}{R} \sum_i \int_{t_i^+}^{t_{i+1}^-} N_{A,M}(z,t) (N_{K,L}^*(z,t) - N_{A,M}^*(z,t)) dt$$

5. Fission-Product Yield,  $\gamma_{A,M \rightarrow K,L}$

$$S_5(z) = \frac{\gamma_{A,M \rightarrow K,L} V_Z}{R} \sum_i \left\{ \Phi_i \left( \sum_g \psi_i(z,g) \sigma_{A,M}^f(z,g) \right) \right. \\ \left. \int_{t_i^+}^{t_{i+1}^-} N_{A,M}(z,t) N_{K,L}^*(z,t) dt \right\}$$

6. Initial Condition,  $N_{A,M}(z)$

$$S_6(z) = \frac{N_{A,M}(z, t_0^+) \cdot N_{A,M}^*(z, t_0^+) V_Z}{R}$$

B. Sensitivity Coefficients for Coupled,  
Quasi-Static Approximations

7. Multigroup Capture Cross Section,  $\sigma_{A,M}^C(z,g)$

$$S_7(z,g) = S_1(z,g) + \frac{\sigma_{A,M}^C(z,g)}{R} \sum_i \left\{ N_{A,M}(z,t_i) \sum_{r \in Z} \Gamma_i^*(r,g) \right. \\ \left. \left( 1 - 3D^2(z,g,t_i) \nabla^2 \right) \psi_i(r,g) V_r \right\}$$

8. Multigroup Fission Cross Section,  $\sigma_{A,M}^f(z,g)$

$$S_8(z,g) = S_2(z,g) + \frac{\nu \sigma_{A,M}^f(z,g)}{R} \sum_i N_{A,M}(z,t_i) \left\{ \sum_{r \in Z} \left( \sum_{g'} \frac{\chi_{g'} \Gamma_i^*(r,g')}{k_{eff,i}} \right. \right. \\ \left. \left. - 3D^2(z,g,t_i) \nabla^2 \right) \psi_i(r,g) V_r + \Phi_i \psi_i(z,g) P_i^* V_Z \right\}$$

9. Decay Constant, same as  $S_4$  for uncoupled case.

10. Fission-Product Yield, same as  $S_5$  for uncoupled case.

11. Multigroup Transport Cross Section,  $\sigma_{A,M}^{tr}(z,g) = \sigma_{A,M}^t(z,g) - \mu \sigma_{A,M}^S(z,g)$

$$S_{11}(z,g) = -3 \frac{\sigma_{A,M}^{tr}(z,g)}{R} \sum_i N_{A,M}(z,t_i) D^2(z,g,t_i) \sum_{r \in Z} \\ \Gamma_i^*(r,g) \nabla^2 \psi_i(r,g) V_r$$

12. Multigroup Scatter Cross Section,  $\sigma_{A,M}^S(z,g' \rightarrow g)$

$$S_{12}(z,g) = \frac{1}{R} \sum_i \left\{ N_{A,M}(z,t_i) \sum_{r \in Z} \Gamma_i^*(r,g) \cdot \sum_{g'} \psi_i(r,g') \sigma_S(z,g' \rightarrow g) \right\}$$

13. Neutron Yield,  $\nu_{A,M}(z,g)$ 

$$S_{13}(z,g) = \frac{\nu_{A,M}(z,g)}{R} \sum_i \left\{ N_{A,M}(z,t_i) \sum_{r \in Z} \psi_i(r,g) \sigma_{A,M}^f(z,g) \sum_{g'} \chi_{g' \rightarrow i} \Gamma_i^*(r,g') \right\}$$

14. Initial Condition of Nuclide Field,  $N_{A,M}(z,t_0)$ 

$$S_{14}(z) = S_6(z) - \frac{N_{A,M}(z,t_0)}{R} \left\{ \sum_g \left[ \sum_{r \in Z} \right. \right. \\ - 3 \sigma_{A,M}^{tr}(z,g) D^2(z,g,t_0) \Gamma_0^*(r,g) \nabla^2 \psi_0(r,g) + \Gamma_0^*(r,g) \psi_0^*(r,g) \sigma_{A,M}^R(z,g) \\ - \left. \Gamma_0^*(r,g) \left( \sum_{g'} \left( \frac{\chi_{g' \rightarrow 0} \nu_{A,M}^f(g')}{k_{eff}} + \sigma_{g' \rightarrow g} \right) \psi_0(r,g') \right) \right] v_r \\ \left. \left. + \Phi_0 \psi_0(z,g) \sigma_{A,M}^f(z,g) P_0^* \right\}$$

## C. Time-Dependent Uncertainty Analysis

Time-dependent uncertainty analysis for burnup calculations is similar to the static uncertainty theory previously developed (41). The established approach is to use the sensitivity coefficients previously presented in conjunction with covariance files for basic nuclear data to develop uncertainties in responses of interest.

The existing evaluations of nuclear data can be thought of as representing the mean value (albeit weighted) derived from a distribution of microscopic measurements. With the issue of ENDF/B-IV — and greatly extending into ENDF-V — the second moments of the distribution of measurements (i.e., the variances and covariances) representing correlated

uncertainties are specified to provide the analyst with a measure of the quality of the data (42, 43).

For the derivations which follow, all required nuclear data such as the various multigroup  $\sigma$ 's and  $\lambda$ 's used in the burnup calculation are assembled in a set that will be called the "reference data vector,"  $\underline{S}(\alpha)$ . For our purposes, the  $i^{th}$  component of the data vector,  $S_i$ , corresponds to the data  $\alpha_i$  that appears at some location (possibly at multiple locations) in the burnup matrix or transport operator, and thus the number of components of  $\underline{S}$  is equal to the number of different data parameters required for the burnup calculation. (Note: Each multi-group constant counts as a separate data parameter.) With this collection of data, the expected value of the response is calculated to be  $R(\underline{S})$ .

If some other data vector  $\underline{S}_n$  were used in the calculation, then another value for the response would be obtained,  $R_n(\underline{S}_n)$ . The distribution of all such possible calculated responses, due to the distribution of nuclear data, is described by the response variance, given by

$$V = \frac{1}{N} \sum_{n=1}^N (R_n - R)^2, \quad \text{VI-1}$$

with  $N$  = number of data vectors used in computing the mean set  $\underline{S}$ ; i.e.,  $N$  is related to the number of measurements for the  $\alpha$ 's in  $\underline{S}$ .

Expanding  $R_n$  in a first-order Taylor series about the expectation value gives

$$R_n = R + \frac{\partial R(\underline{S})^T}{\partial \underline{S}} (\underline{S}_n - \underline{S}) , \quad \text{VI-2}$$

Substituting Eq. VI-2 into Eq. VI-1 results in

$$V = \frac{1}{N} \sum_n \left( \frac{\partial R^T}{\partial \underline{S}} \Delta \underline{S}_n \right)^2 \quad \text{VI-3}$$

Now defining a diagonal matrix of the form

$$D = \begin{bmatrix} \alpha_1 & 0 & 0 & \dots \\ 0 & \alpha_2 & 0 & \dots \\ \vdots & & & \\ 0 & 0 & \dots & \alpha_m \end{bmatrix} \quad \begin{array}{l} \text{where } \alpha_1 = \text{first component of } \underline{S} , \\ \alpha_2 = \text{second component of } \underline{S} , \\ \vdots \\ \alpha_i = i^{\text{th}} \text{ component of } \underline{S} \end{array}$$

Equation VI-3 can be written

$$\begin{aligned} V &= \frac{1}{N} \sum_n \left\{ \frac{\partial R^T}{\partial \underline{S}} \left( \underline{D} \underline{D}^{-1} \right) \Delta \underline{S}_n \right\}^2 \\ &= R^2 \frac{1}{N} \sum_n \left( \frac{\underline{D}}{R} \frac{\partial R}{\partial \underline{S}} \right)^T \left( \underline{D}^{-1} \Delta \underline{S}_n \Delta \underline{S}_n^T \underline{D}^{-1} \right) \left( \frac{\underline{D}}{R} \frac{\partial R}{\partial \underline{S}} \right) . \end{aligned} \quad \text{VI-4}$$

Noting that  $\partial R / \partial \underline{S}$  is independent of the summation index, Eq. VI-4 is finally expressed as

$$\frac{V}{R^2} = \underline{P}^T \underline{C} \underline{P} = \text{relative response variance} , \quad \text{VI-5}$$

where

$$\underline{P} = \frac{\underline{D}}{R} \frac{\partial R}{\partial \underline{S}} \quad \text{VI-6}$$

$$\underline{C} = \frac{1}{N} \sum_n \left( \underline{D}^{-1} \Delta \underline{S}_n \Delta \underline{S}_n^T \underline{D}^{-1} \right) \quad \text{VI-7}$$

The matrix  $\underline{C}$  formed by the dyadic square of  $\Delta \underline{S}_n$  is called the "relative covariance matrix," and the vector  $\underline{P}$  is called the "sensitivity vector." In general the elements of  $\underline{C}$  are energy and nuclide dependent, as are the components of  $\underline{P}$ . The off-diagonal terms of  $\underline{C}$  account for correlations in data uncertainties; these cross correlations can be between data at different energies for the same nuclide or between data of different nuclides. For example, most fission cross sections are measured relative to U-235 fission, and hence there is an indirect uncertainty in the fission cross section of most nuclides due to the uncertainty in the U-235 fission cross section. Data covariance files are generated by the data evaluators, and are independent of the sensitivity theory discussed in this text. The components of  $\underline{P}$  correspond to the sensitivity coefficients defined earlier for the various data.

The equations for uncertainty analysis of depletion calculations are of the same form as the static case, the only differences being in how the sensitivity coefficients are defined and in the types of data contained in the covariance matrix (e.g., depletion uncertainty analysis requires covariances for decay data, yield data, etc., in addition to

cross-section covariances). This fact is significant, since it implies that computer codes developed to fold sensitivity coefficients with covariance matrices for static analysis can also be used in burnup analysis.

In theory, the data vector can be "adjusted" to minimize the difference between some computed value and an experimentally measured value for a burnup related response, using the uncertainty analysis as a guide. Such "consistent" adjustment procedures have been studied for static integral experiments (44), such as measurements in the ZPPR critical assemblies; and it is possible that, using the methods discussed in this chapter, integral measurements of the isotopic composition of irradiated nuclide samples could be factored into the adjustment procedure. This type of integral data could be obtained from either analyzing spent fuel elements from power reactors or by controlled irradiation of small, pure samples placed in a reactor core. Sensitivity coefficients for the former case would have to be computed using the coupled perturbation technique, while it would probably be sufficient to use the uncoupled method for the latter case since it can be assumed that variations in the sample data do greatly affect the neutron field in the reactor. A sample uncertainty calculation for the second type of experiment is given in Chapter VIII.

## CHAPTER VII

### BURNUP ADJOINT FUNCTIONS: INTERPRETATION AND ILLUSTRATIVE CALCULATIONS

We will now present a physical interpretation of the burnup adjoint functions previously derived on strictly mathematical grounds. This will be done by examining various properties of the adjoint functions and drawing analogies with neutron transport theory, and by presenting example problems which illustrate these properties. Recall from Chapter II that the adjoint burnup equations are actually "first-order adjoint equations"; i.e., they contain the adjoint operators for the *linearized* forward equations given in Eq. III-35 for the initial value formulation and in IV-13 for the eigenvalue formulation. This fact will be used later in examining conservation laws for the "response flow."

Let us begin by considering only the linear transmutation equation for the nuclide field and temporarily neglecting the effect of the neutron field (i.e., the uncoupled approximation in which the flux can be specified independently of the nuclide field). Also, all independent variables except "time" are suppressed for notational purposes, and we will specifically consider a final-time response  $R(t_F)$ . Therefore, the nuclide field is described by the linear equation

$$\underline{M} \underline{N}(t) = \frac{d}{dt} \underline{N}(t) , \underline{N}(0) = \underline{N}_0 \quad \text{VII-1}$$

where  $\underline{M}$  is a linear matrix operator. The corresponding adjoint equation is

$$\underline{M}^* \underline{N}^*(t) = - \frac{d}{dt} \underline{N}^*(t) , \quad \underline{N}^*(t_f) = \frac{\partial R}{\partial \underline{N}} (t_f) \quad \text{VII-2}$$

Note the similarity between VII-1 and the linear neutron transport equation

$$B\phi(t) = \frac{1}{v} \frac{d}{dt} \phi(t) \quad \phi(0) = \phi_0 \quad \text{VII-3}$$

with the adjoint equation

$$B^* \phi^*(t) = - \frac{1}{v} \frac{d}{dt} \phi^* , \quad \phi^*(t_f) = \frac{\partial R}{\partial \phi} (t_f) .$$

It is well known that the solution to the adjoint time-dependent Boltzmann transport equation can be interpreted as follows (21):

$\phi^*(t)$  = "importance of a neutron at time  $t$  to the response at time  $t_f$ ." (Note — again, all phase-space variables except "time" are implicitly treated.)

By analogy we would expect the time-dependent nuclide adjoint to play a similar role for final-time functionals in burnup calculations. We assert the following axiom:

If  $N_i(t)$  =  $i^{th}$  component of the nuclide-field vector  $\underline{N}(t)$ , then

$N_i^*(t)$  = importance of nuclide  $i$  at time  $t$  to the response at time  $t_f$  = average future response contained in atoms of nuclide  $i$ .

If the nuclide adjoint is normalized properly then this definition can be stated

$N_i^*$  = fraction of atoms of nuclide  $i$  present at time  $t$ , which will be transmuted into response nuclides at time  $t_f$ .  
 = probability at time  $t$  that nuclide  $i$  will contribute to the response at time  $t_f$ .

For the burnup equation with a fixed neutron-flux field, the above definitions show that the adjoint nuclide field is independent of the forward field, and, therefore, a particular adjoint calculation is applicable to any nuclide composition exposed to the same flux field as used in the original calculation. This fact is analogous to the situation for the neutron adjoint, which is applicable to all neutron-flux fields that have a common nuclide field. In both instances the forward field is fixed by the initial conditions, and the adjoint field is fixed by the final response.

The importance property of the nuclide adjoint can be used to directly derive the adjoint transmutation equation for an uncoupled nuclide field using first principles, in a manner similar to the method used by Lewins to derive the neutron adjoint equation (21). Following Lewins, we introduce the principle for "conservation of nuclide importance," which states that a nuclide which does not perturb its

specified neutron environment is as important as its daughters (from both reactions and decays). From this axiom, it can be seen that the importance of nuclide  $i$  at time  $t$  is equal to its importance at  $t + \Delta t$  plus the importance of all daughters it produces during  $\Delta t$ . Let  $\alpha_i$  be the total transmutation probability per unit time for nuclide  $i$ ; then  $(1 - \alpha_i \Delta t)$  = probability that nuclide  $i$  does not transmute during  $\Delta t$ . Let  $\alpha_{i \rightarrow j}$  be the probability per unit time that nuclide  $i$  will transmute into nuclide  $j$ . Then applying the conservation of nuclide importance:

$$N_i^*(t) = N_i^*(t + \Delta t)(1 - \alpha_i \Delta t) + \sum_{j \neq i} \alpha_{i \rightarrow j} N_j^* \Delta t, \quad \text{VII-4}$$

rearranging terms,

$$\frac{N_i^*(t) - N_i^*(t + \Delta t)}{\Delta t} = -\alpha_i N_i^*(t + \Delta t) + \sum_{j \neq i} \alpha_{i \rightarrow j} N_j^*. \quad \text{VII-5}$$

Finally, taking the limit  $\Delta t \rightarrow 0$ ,

$$\sum_j \alpha_{i \rightarrow j} N_j^* = -\frac{d}{dt} N_i^*, \quad \text{VII-6}$$

where  $\alpha_{i i}$  is defined to  $-\alpha_i$ . This equation can be written in vector notation as

$$-\frac{d}{dt} \underline{N}^* = \underline{A} \underline{N}^*. \quad \text{VII-7}$$

Comparing the elements of  $\underline{A}$  to the elements of the burnup matrix  $\underline{M}$ , we see that  $\underline{A} = \text{transpose } \underline{M} = \underline{M}^*$ . Therefore  $\underline{M}^* \underline{N}^* = -d/dt \underline{N}^*$ .

The importance conservation property of the adjoint-nuclide field also makes possible the creation of a "nuclide contribution theory." The concept of neutron contribution theory has been introduced in earlier papers as a method to determine the mechanism by which neutrons flow from the forward source to the response detector, so as to locate spatial streaming paths (18). A similar idea can be applied to the nuclide field to find the major "nuclide paths" by which atoms are transformed from the initial isotopic concentrations into the final response concentrations.

To this end, a quantity known as the "contribution response-density" for nuclide  $i$  can be defined to be:

$c_i(t)$  = total response contribution which can be attributed to the atoms of nuclide  $i$  present at time  $t$ .

It is easy to see from the definition of the adjoint,

$$c_i(t) = N_i(t)N_i^*(t) . \quad \text{VII-8}$$

Because the final response must originate from some nuclide present in the system,

$$\sum_i c_i(t) = \text{final response} , \quad \text{VII-9}$$

for all  $t$  in the interval  $[t_0, t_f]$ . This can be written as

$$\underline{N}^T(t)\underline{N}^*(t) = \text{response} . \quad \text{VII-10}$$

Note that this is consistent with the conservation law discussed in Chapter II (see Eq. II-5).

A knowledge of  $c_i(t)$  for all nuclides allows one to determine which isotopes at time  $t$  contribute most heavily to the response of interest, which could possibly be beneficial to optimization studies in reactor design.

Hence we have found that for a nuclide field which is uncoupled from the flux field,  $\underline{N}^*$  corresponds to the importance of the various nuclide concentrations to the response. For coupled neutron/nuclide fields, a similar interpretation will apply; however, the principle of conservation of importance must be modified to account for coupling interactions. Before proceeding to the more difficult coupled adjoint equations, much insight can be obtained at this point by considering a detailed example addressing the properties discussed thus far for the uncoupled case.

The example problem consists of a point-depletion model provided by EPRI (Electric Power Research Institute) (45) for a homogenized PWR fuel zone. Initial concentrations are given in Table VII-1, and the time-dependent thermal flux (which was also supplied by EPRI) is given in Table VII-2. The ORIGEN-A code discussed earlier computed the forward and adjoint nuclide fields. Nuclear data came directly from the ORIGEN library (26). The response was selected to be the inventory of  $^{239}\text{Pu} + ^{240}\text{Pu} + ^{241}\text{Pu} + ^{242}\text{Pu}$  at the end of exposure ( $t_f = 25,614$  hours).

Table VII-1. Initial concentrations for homogenized fuel

Nuclide	Number density
<sup>160</sup>	4.37-02
<sup>135</sup> X	0.0
<sup>149</sup> Sm	0.0
<sup>234</sup> U	4.45-06
<sup>235</sup> U	5.67-04
<sup>236</sup> U	3.53-06
<sup>238</sup> U	2.13-02
<sup>239</sup> Pu	0.0
<sup>240</sup> Pu	0.0
<sup>241</sup> Pu	0.0
<sup>242</sup> Pu	0.0
<sup>241</sup> Am	0.0

Table VII-2. Time-dependent thermal flux

Time interval	t <sub>i</sub> (hr)	$\phi$ (x 10 <sup>13</sup> ) neutrons/cm <sup>2</sup> ·sec
1	75.34	4.52
2	376.68	4.54
3	1506.68	4.51
4	3013.42	4.43
5	4520.13	4.38
6	6026.84	4.37
7	7533.55	4.38
8	9040.26	4.41
9	10546.97	4.46
10	12053.68	4.51
11	13560.39	4.58
12	15067.10	4.65
13	16573.81	4.72
14	18080.52	4.81
15	19587.13	4.89
16	21093.94	4.98
17	22600.65	5.07
18	24107.36	5.17
19	25614.07	5.26

The values for the most important time-dependent actinide densities found in the forward ORIGEN-A calculation are shown in Figs. VII-1 and VII-2. As expected, the concentrations of uranium and plutonium isotopes dominate the results of the forward case, with  $^{238}\text{U}$  being the most predominate by far, due to its large initial concentration. Figure VII-3 shows the major chains for plutonium buildup.

Figures VII-4 - VII-8 summarize the results of the adjoint ORIGEN-A calculation. For this run the final values were zero for all nuclides except  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{242}\text{Pu}$ , which had concentrations of 1.0, since this is the realization vector corresponding to a response of "plutonium inventory at shutdown."

At first sight it may be surprising to see some of the more uncommon isotopes (such as  $^{237}\text{U}$ ,  $^{242}\text{Cm}$ , etc.) appearing among the important isotopes for producing plutonium. It may be equally surprising that the dominant nuclide in the forward calculation —  $^{238}\text{U}$  — is not among the most dominant adjoint values! The results appear more reasonable when one realizes that the "importance" of a nuclide in the uncoupled case is *independent* of its concentration. Even though nuclides such as  $^{240}\text{Np}$  have only a small number of atoms present at any given time, any atom which is present has a high probability of being transformed into a plutonium atom by shutdown. The importance of  $^{238}\text{U}$  atoms ( $\sim 10^{-3}$ ) is comparatively low due to their having a smaller capture cross section\* ( $\sim 3$  b) than more important isotopes such as  $^{237}\text{Np}$  ( $\sim 170$  b). Therefore

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\*Cross sections quoted are 2200 m/s values.

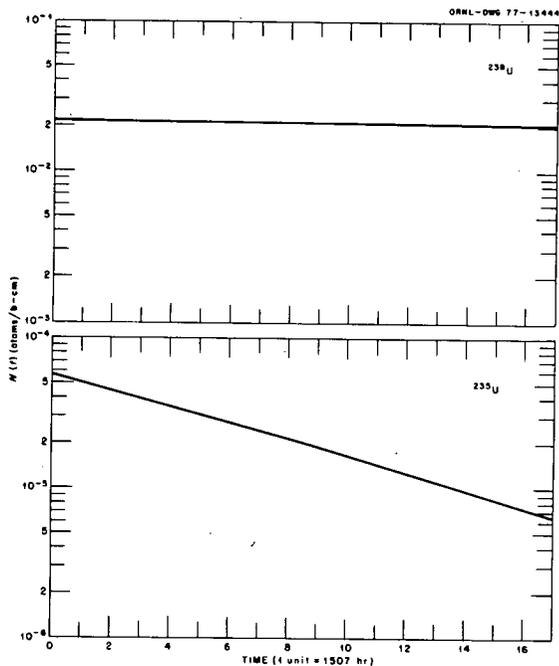


Fig. VII-1. Uranium atom densities.

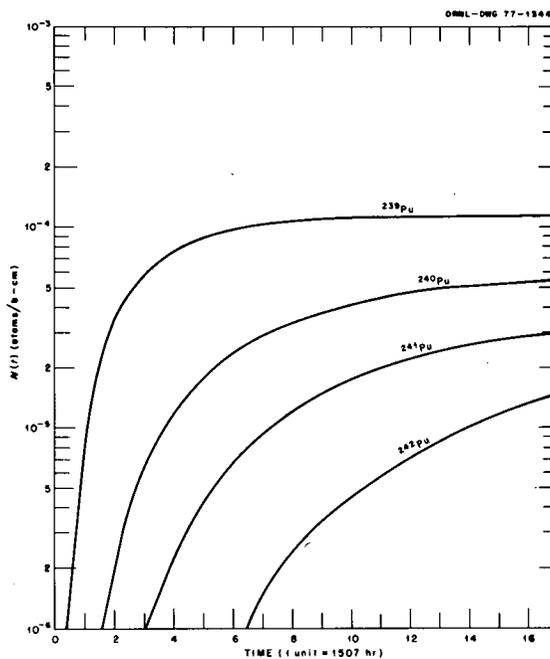


Fig. VII-2. Plutonium atom densities.

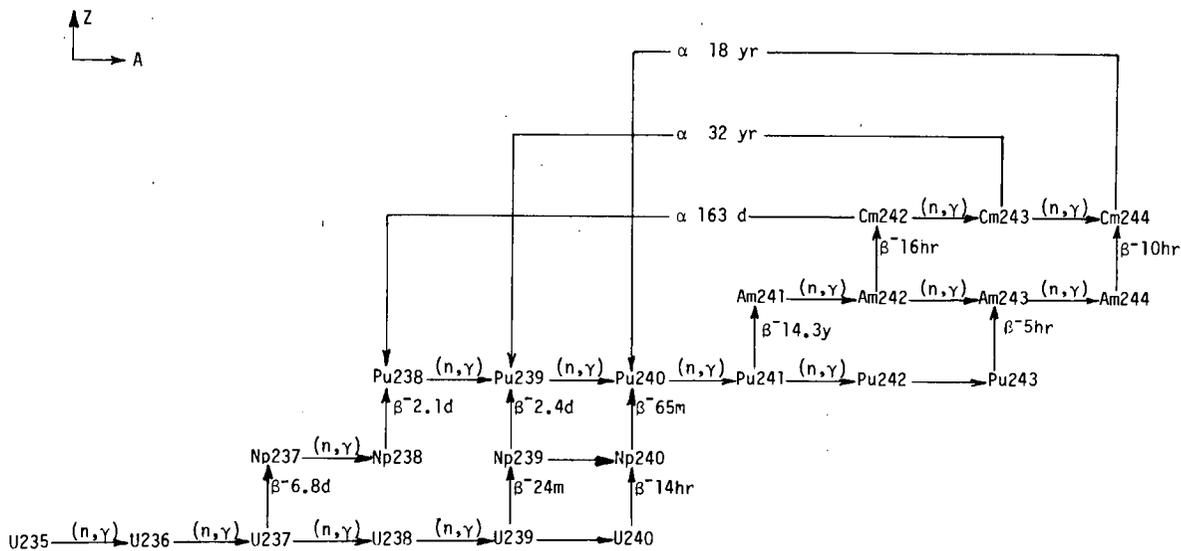


Fig. VII-3. Major chains for plutonium production.

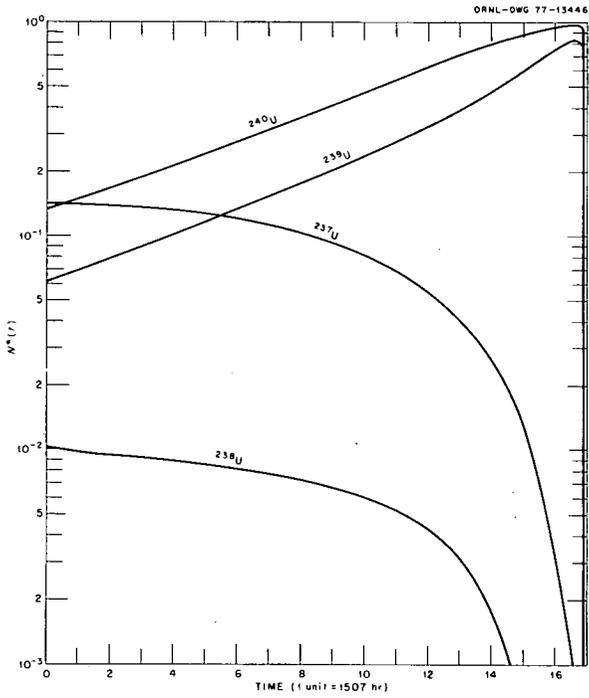


Fig. VII-4. Uranium adjoint functions.

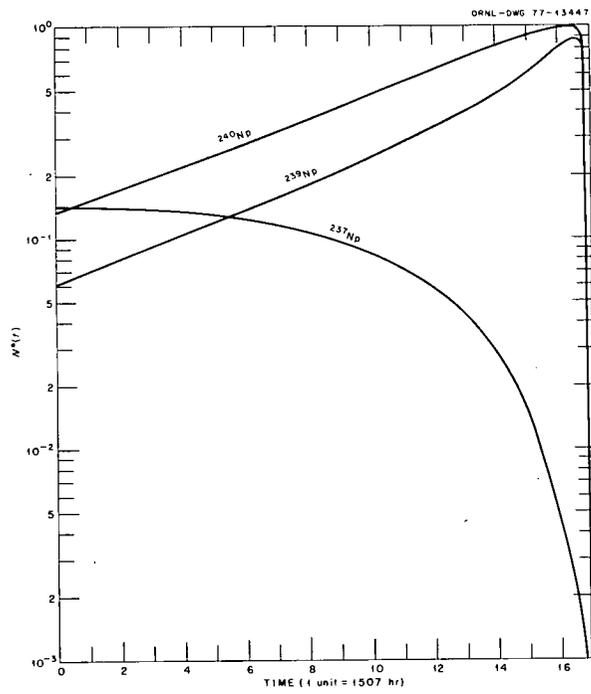


Fig. VII-5. Neptunium adjoint functions.

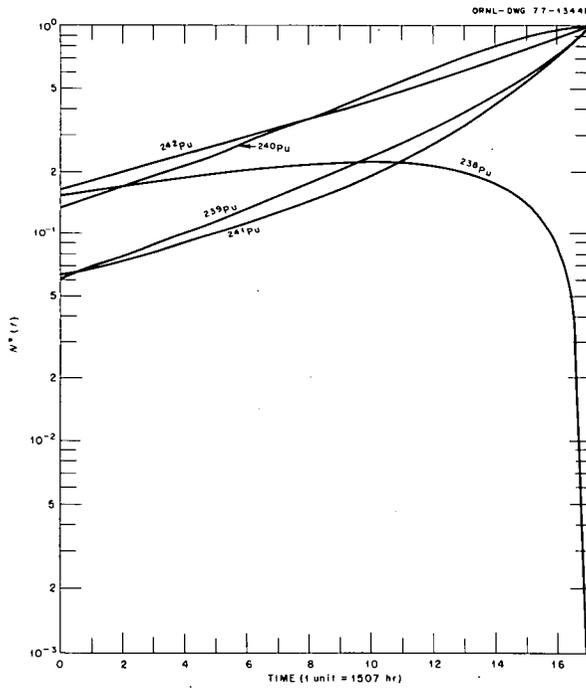


Fig. VII-6. Plutonium adjoint functions.

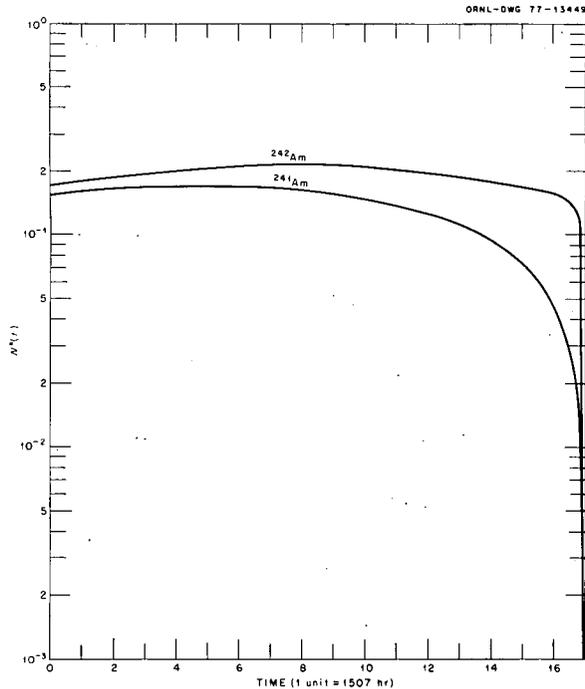


Fig. VII-7. Americium adjoint functions.

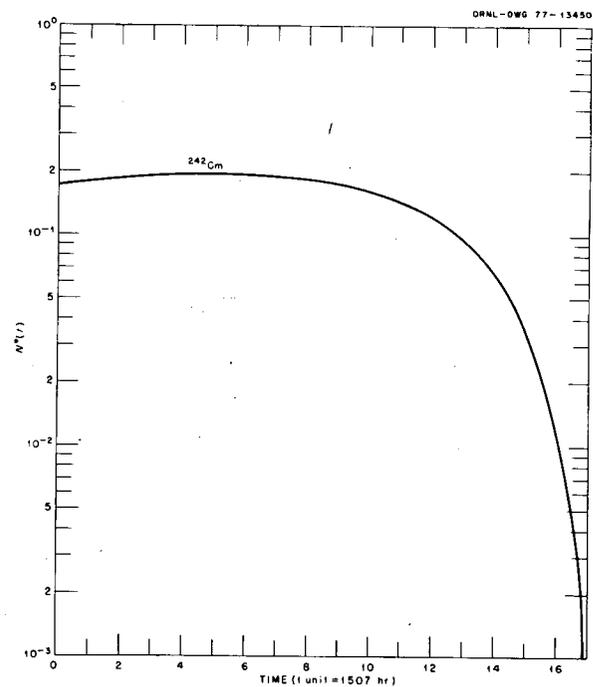


Fig. VII-8. Curium adjoint functions.

a  $^{238}\text{U}$  atom has less probability of being transformed into Pu than does a  $^{237}\text{Np}$  atom; i.e., a smaller *fraction* of  $^{238}\text{U}$  will transmute into Pu, although the absolute number of  $^{238}\text{U}$  atoms which contribute to the response is much greater than for  $^{237}\text{Np}$ , since there are far more  $^{238}\text{U}$  atoms than Np atoms present in the reactor.

An examination of several nuclide adjoints will perhaps give the reader a better physical insight. The Pu response isotopes themselves are obviously important, especially at times near  $t_f$ . At earlier times, the high fission cross section makes an atom of a fissile Pu isotope quite likely to disappear before it lives to  $t_f$ . The adjoint for  $^{238}\text{Pu}$  decreases near  $t_f$  because it was not directly contained in the response. Note that the adjoint functions for all nuclides except those contained

in the response must go to zero at the final time, a fact which accounts for the dramatic fall in some adjoints near  $t_f$ .

Actinides with an atomic number higher than 94 are usually important through their decay modes. For example,  $^{242}\text{Cm}$  has a moderate absorption cross section ( $\sim 30$  b) and a relatively short half-life (163 d); therefore it has about thirty times greater probability of decaying to  $^{238}\text{Pu}$  than of capturing a neutron to become  $^{243}\text{Cm}$  — note the similarity in the  $^{238}\text{Pu}$  adjoint curve and the  $^{242}\text{Cm}$ , adjoint curve. Furthermore, even if the  $^{242}\text{Cm}$  atom does transmute to  $^{243}\text{Cm}$ , there is still a possibility that the  $^{243}\text{Cm}$  isotope will decay to  $^{239}\text{Pu}$ .

Americium-242 is important because it decays by beta emission to  $^{242}\text{Cm}$  and by electron capture directly to  $^{242}\text{Pu}$ , and its short half-life ( $t_{1/2} = 16$  hr) makes the transition likely over a long time period. In fact, even at one time interval before shutdown its adjoint is still quite high. At early times the isotope  $^{237}\text{U}$  is an important nuclide whose mode of contribution is fairly complicated to assess. Its short half-life (7 d) and large capture cross section (480 b) provide two possible methods for the nuclide to transmute into Pu. If  $^{237}\text{U}$  captures a neutron, it becomes  $^{238}\text{U}$  and follows the familiar procedure for creating  $^{239}\text{Pu}$ . The alternate method is for  $^{237}\text{U}$  to decay by beta emission to  $^{237}\text{Np}$ . Since this nuclide has a long half-life ( $2 \times 10^6$  y), it is probable that an atom will capture a neutron ( $\sigma_c = 169$ ) and become  $^{238}\text{Np}$ , which then decays ( $t_{1/2} = 2.12$  d) into  $^{238}\text{Pu}$ . An examination of Figs. 3 and 4 reveals that over most of the cycle,  $^{237}\text{U}$  is more important than  $^{238}\text{U}$  but slightly less important than  $^{237}\text{Np}$ , a fact which leads one to believe that the second contribution mode is more important.

Table VII-3 contains the values of the contribution densities for the major nuclides. It is seen that until near the end of cycle the response stored in the  $^{238}\text{U}$  atoms overwhelms all others, due to its large initial charge. At time step 17 Pu begins to dominate, as the  $^{238}\text{U}$  atoms are "running out of time" in which they can transmute into Pu. Notice that the initial contribution density for  $^{238}\text{U}$  is  $2.15 \times 10^{-4}$ , which was found to be exactly the value of the plutonium inventory at shutdown (see last row in Table VII-3). This indicates — as expected — that initially the entire response is contained in the  $^{238}\text{U}$  atoms:

$$R(t_f) = (N_0 N_0^*)_{238\text{U}} .$$

We now proceed to examine the interpretation of adjoint functions for coupled neutron/nuclide fields. The initial value burnup equation will be studied first because it is the easiest to interpret physically. The eigenvalue formulations, although convenient for numerical solutions, are awkward to manipulate and therefore it is wise to consider the simpler initial-value formulation in order to obtain a hint of what to expect from the quasi-static solutions  $\underline{N}^*$ ,  $\Gamma^*$ , and  $p^*$ . It is also worth pointing out that for all cases we will be dealing with linearized equations that describe small deviations in the fields about some reference conditions. Only under this approximation of linearity can a physical interpretation be given for the adjoint functions, since we are dealing with the first-order adjoint equations.

Table VII-3. Major contributon densities<sup>a</sup>  
(atoms/cm<sup>3</sup> × 10<sup>-24</sup>)

Time interval	<sup>238</sup> U	<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu
1	2.15-4 <sup>b</sup>	0	0	0	0
2	2.15-4	0	0	0	0
3	2.15-4	0	0	0	0
4	2.13-4	2.36-6	0	0	0
5	2.09-4	4.63-6	1.08-6	0	0
6	2.06-4	6.80-6	2.27-6	0	0
7	2.01-4	8.94-6	3.82-6	0	0
8	1.97-4	1.12-5	5.71-6	0	0
9	1.91-4	1.36-5	7.92-6	1.04-6	0
10	1.86-4	1.63-5	1.05-5	1.54-6	0
11	1.78-4	1.91-5	1.35-5	2.13-6	1.18-6
12	1.70-4	2.24-5	1.70-5	2.89-6	1.76-6
13	1.60-4	2.63-5	2.09-5	3.82-6	2.53-6
14	1.49-4	3.11-5	2.53-5	5.02-6	3.50-6
15	1.35-4	3.71-5	3.03-5	6.57-6	4.73-6
16	1.19-4	4.43-5	3.58-5	8.65-6	6.22-6
17	9.81-5	5.39-5	4.15-5	1.15-5	8.10-6
18	7.29-5	6.70-5	4.73-5	1.55-5	1.03-5
19	4.04-5	8.58-5	5.22-5	2.15-5	1.30-5
20	0	1.14-4	5.48-5	3.02-5	1.62-5

$${}^a N_i(t) \cdot N_i^*(t).$$

<sup>b</sup>Read as  $2.15 \times 10^{-4}$ .

Therefore consider the linearized initial-value equation (III-36) and its first-order adjoint equation, IV-70. When these equations are cross-multiplied; integrated over  $E, \Omega, V$ ; and subtracted in the usual manner, the following relation is obtained:

$$-\frac{d}{dt} \left\{ \left[ \Gamma^* \frac{1}{V} \Delta\phi \right]_{E, \Omega, V} + [N^* \Delta N]_V \right\} = \left[ \frac{\partial R}{\partial N} \Delta N + \frac{\partial R}{\partial \phi} \Delta\phi \right]_{E, \Omega, V} \quad \text{VII-11}$$

The above equation is the analog to Eq. VII-10 in the uncoupled case, which expresses the conservation of response. As before, if we assume that  $R[N, \phi]$  is a final-time response, then Eq. VII-11 can be integrated from  $t$  to  $t_f$  (recall,  $\Gamma^*(t_f) = N^*(t_f) = 0$ ) to give

$$\begin{aligned} [\Gamma^* \Delta n(t)]_{E, \Omega, V} + [N^* \Delta N(t)]_V &= \left[ \frac{\partial R}{\partial N} \Delta N(t_f) + \frac{\partial R}{\partial \phi} \Delta\phi(t_f) \right]_{E, \Omega, V} \\ &= \Delta R(N, \phi) \end{aligned} \quad \text{VII-12}$$

where  $\Delta n(t) = \text{change in neutron density field} = \frac{1}{V} \Delta\phi$ , and  $t$  is any time in  $[t_0, t_f]$ .

The LHS of the above equation is again identified as the contribution response density, but now it is composed of two components — one arising from response stored in the neutron field, and the other from response stored in the nuclide field. The total response contained in both fields is conserved; however, the relative amounts contained in the individual fields may vary with time; i.e., response contained in the nuclide field may be transferred to the neutron field and vice versa!

Hence we must extend the definition of "importance" to address two simultaneous fields which interact. This can be done only with the linearized equations, for which the effects of the two fields may be superimposed. One consequence of this fact is that importance cannot be expressed independently of the reference forward solution. Within this limitation, we can state the following definition (for a final-time response):

The importance of a field at time  $t$  is the expected effect it will have on the response at  $t_f$ .

Another way of stating this definition is that the importance of a field at  $t$  is the expected change in the response if the field were perturbed slightly at time  $t$ .

This definition of importance is consistent with Eq. VII-12. For example, suppose that at time  $t$  the neutron field is perturbed by

$\frac{1}{V} \Delta\phi \delta(r_0) \delta(\Omega_0) \delta(E_0)$ . Then from Eq. VII-12,

$$\Gamma^*(r_0, E_0, \Omega_0, t_0) \frac{1}{V} \Delta\phi(r_0, E_0, \Omega_0, t) = \Delta R(t_f) .$$

Dividing the left-hand side by the number of neutrons perturbed ( $= \frac{1}{V} \Delta\phi$ ) gives the expected (average) effect at time  $t$  of a neutron with coordinates  $(r_0, E_0, \Omega_0)$ , which is  $\Gamma^*(r_0, E_0, \Omega_0, t)$ . It is important to realize that even if the response in Eq. VII-12 does not explicitly depend on the neutron field  $\left(\frac{\partial R}{\partial \phi} = 0\right)$ , a neutron may still have importance since it may alter the future behavior of the nuclide field.

We can now generalize the principle of conservation of importance originally stated by Lewins for an uncoupled neutron field and subsequently

extended to the case for an uncoupled nuclide field earlier in this chapter. The new postulate is the conservation of field importance for coupled neutron/nuclide fields:

"A field is as important as its progeny plus the importance of any transformations it induces in the other field."

Lewins' principle of conservation of neutron importance, as well as the principle of conservation of nuclide importance presented earlier in this chapter, are special cases of the principle of conservation of field importance. These special cases occur when one field does not induce transformations in the other field; i.e., when there is no coupling.

As an example application of this general principle, we will derive the nuclide adjoint equation for the initial-value formulation of the burnup equations.

Equation VII-7, which was derived for the uncoupled case, is still valid for the nuclide-progeny importance, but we must also determine the importance of transformations in the neutron field induced by nuclide  $i$ . The average loss in response contained in the neutron field at position  $\rho$  due to interactions with atoms of nuclide  $i$  at position  $r$ , time  $t$  in  $\rho$ -space is

$$\Gamma^*(\rho)\sigma_{t,i}(\rho)\phi(\rho)$$

The average gain in neutron-field response due to neutrons born from interactions with an atom of nuclide  $i$  is

$$\phi(\rho) \int \sigma_i(\rho \rightarrow \rho') \Gamma^*(\rho') d\rho'$$

Therefore the net expected change in importance of the neutron field at position  $\rho$  due to nuclide  $i$  at  $r$  and  $t$  is

$$\phi(\rho) \left\{ -\sigma_{t,i} \Gamma^*(\rho) + \int_{\rho'} \sigma_i(\rho \rightarrow \rho') \Gamma^*(\rho') d\rho' \right\} = \phi(\rho) \frac{\partial B^* \Gamma^*(\rho)}{\partial N_i}, \quad \text{VII-13}$$

where  $r$  and  $t$  are two components of  $\rho$ . The total change in neutron-field importance at position  $(r,t)$  in  $\rho$ -space due to transformations induced by nuclide  $i$  at  $(r,t)$  is

$$\left[ \phi(\rho) \frac{\partial B^* \Gamma^*(\rho)}{\partial N_i} \right]_{E,\Omega} \quad \text{VII-14}$$

Similar expressions can be written for each component of  $\underline{N}$ , and the general vector relation is

$$\left[ \phi(\rho) \frac{\partial}{\partial \underline{N}} B^* \Gamma^*(\rho) \right]_{E,\Omega} \quad \text{VII-15}$$

When this term is added to Eq. VII-7, the following adjoint equation is obtained:

$$\underline{M^* N^*} + \left[ \phi \frac{\partial B^*}{\partial \underline{N}} \Gamma^* \right]_{E,\Omega} = - \frac{\partial}{\partial t} \underline{N^*} \quad \text{VII-16}$$

which corresponds to the nuclide-adjoint equation in Eq. IV-73 (remember, that equation was implicitly integrated over  $E$  and  $\Omega$ ).

An analogous derivation can be made for the neutron-field adjoint equation. Thus we see that the burnup adjoint functions do account for the fact that the neutron and nuclide fields are coupled, at least in the initial-value formulation. However, one can no longer isolate the importance of the neutron field from the importance of the nuclide field, because the importance of one depends on the importance of the other.

Unfortunately, things become even more complicated for the quasi-static formulation, because now there are three variables ( $\underline{N}$ ,  $\Phi$ ,  $\psi$ ) and three adjoints ( $\underline{N}^*$ ,  $p^*$ ,  $\Gamma^*$ ), which are discontinuous in time. As in the initial-value formulation, the importance carried by a neutron can be transferred to the nuclide field; but now there is the additional coupling which arises from the fact that the shape of the neutron field can influence its magnitude.

As before, it is difficult to relate changes in the individual variables ( $\underline{N}$ ,  $\Phi$ ,  $\psi$ ) to a change in the response because the fields cannot be perturbed individually, i.e., a change in any one of the variables will automatically perturb the other two. The important fact to be realized is that the quasi-static adjoint functions *account* for this coupling by allowing importance to be transferred through the coupled adjoint equations. In other words, the adjoint functions not only account for the direct effect of the change to a given field, but also account for the effects of the associated transformations in the other fields caused by the initial perturbation. However, unlike the initial value formulation, in the quasi-static formulation the transfer of

importance can only occur at discrete times; for example, the "jump condition" expressed in Eq. IV-64 clearly shows how importance contained in the neutron field is transferred to the nuclide field at the boundary of each broad-time interval. We will examine the functions  $\underline{N}^*$ ,  $P^*$  and  $\Gamma^*$  one at a time. Consider first the  $\underline{N}^*$  function. In Eq. IV-59 we have shown that

$$\Delta R = \Delta \underline{N}_0 \underline{N}^*_0$$

Although this expression was derived for a perturbation in the nuclide field at  $t = 0$ , it is easy to obtain the more general relations

$$\Delta R = \Delta \underline{N}(t_i) \cdot \underline{N}^*(t_i) \quad \text{VII-17 (a)}$$

This equation shows that  $\underline{N}^*(t_i)$  in the quasi-static formulation (as in the initial value formulation) represents the importance of a change in the nuclide field at  $t_i$  to the final response. Notice that  $\underline{N}^*$  accounts for several effects — the direct effect of the change in the nuclide field, as well as the indirect effects of change in the flux shape and magnitude that accompany a perturbation in  $\underline{N}$ . All of this information is contained in  $\underline{N}^*$ . These various components of  $\underline{N}^*$  will be examined in more detail later.

Consider next the  $P^*$  term. It can be shown (for example by perturbing the power in Eq. IV-33) that

$$\Delta R = P^*_i \Delta P_i \quad \text{VII-17 (b)}$$

Since  $P_i$  fixes the flux normalization at  $t_i$ , we conclude that  $P_i^*$  represents the importance of a change in the flux magnitude at  $t_i$ . Again,  $P^*$  will account not only for the direct effect of  $\frac{\partial R}{\partial P_i}$ , but also for the indirect effects of the perturbations in  $\underline{N}$  and  $\psi$  that occur when the power is perturbed. Finally consider the function of  $\Gamma_i^*(P)$ . Suppose that the shape of the neutron flux field at  $t_i$  is perturbed by inserting a source of neutrons at position  $(r_0, E_0, \Omega_0)$ . This amounts to the addition of a delta function source of neutrons to Eq. III-28 equal to  $\Delta\psi \frac{1}{v_0} \delta(r-r_0) \delta(E-E_0) \delta(\Omega-\Omega_0)$  so that

$$B\psi_i = \frac{\Delta\psi}{v_0} \delta(r-r_0) \delta(E-E_0) \delta(\Omega-\Omega_0) .$$

If this equation is used to replace the unperturbed shape equation in IV-33, it is seen that

$$\Delta R = \Gamma_i^*(r_0, E_0, \Omega_0) \frac{1}{v_0} \Delta\psi(r_0, E_0, \Omega_0) . \quad \text{VII-17 (c)}$$

Therefore  $\Gamma_i^*(r_0, E_0, \Omega_0)$  represents the importance of a change at time  $t_i$  in the shape of the neutron field at  $(r_0, E_0, \Omega_0)$ . As in the other cases, this importance accounts not only for the direct effect of the perturbation to  $\psi$  but also its indirect effects.

It has been stated repeatedly that the various adjoint functions account for coupled perturbations arising from the interaction between  $(\underline{N}, P, \psi)$ . In fact, all three adjoint functions actually depend on the future behavior of each other! For example,

$$\underline{N}^*(t) = f[\underline{N}^*(t' > t), P_i^*(t_i > t), \Gamma_i^*(t_i > t)] .$$

It is this fact which accounts for the feedback between perturbations in the forward fields. For example  $\underline{N}^*(t)$  depends on the future value of  $P^*$  because a perturbation in the nuclide field at time  $t$  will cause a perturbation in the future value of the flux magnitude, which can be related to a change in the response by  $P^*$ . At the same time,  $P^*$  depends on the future behavior of  $\Gamma^*$  and  $\underline{N}^*$ , etc., etc. Because of the complicated interactions between the fields, it is not possible to speak of the importance of perturbations only to the nuclide field or only to the neutron field, since such perturbations are not physically realizable in general. One must deal simultaneously with perturbations to all three variables  $\underline{N}$ ,  $\phi$ , and  $\psi$ , which is exactly what the coupled adjoint functions do.

To examine how perturbations in coupled neutron/nuclide fields are accounted for by the adjoint functions, two analytic example problems will be considered for the nuclide adjoint. In the first it is shown that the value for the uncoupled nuclide adjoint, which only accounts for direct perturbations in the nuclide concentration, is modified for the coupled case to include a term accounting for the indirect effect of the change to the flux magnitude. In the second example, a similar type analysis is performed except that changes in the flux spectrum are considered.

The first example problem to be solved is the simplest possible case of an infinite, single-nuclide medium in which the energy behavior of the

flux is described by one energy group (thus technically this is a point-depletion calculation). These assumptions are sufficient to assure that the only importance of the neutron field is through its magnitude (the "shape" is constant and equal to 1). We further assume that the calculation is to be performed in a single time step. The specified purpose of the calculation is to determine the sensitivity of the nuclide concentration at time  $T_f$  to changes in the initial condition at time zero.

The burnup equations for this example are then

$$N_0 (\sigma_a - \lambda v \sigma_f) \psi_0 = 0 \quad (\text{flux shape equation}) \quad \text{VII-18}$$

$$\psi_0 N_0 \phi_0 \sigma_f = P \quad (\text{flux normalization equation}) \quad \text{VII-19}$$

$$\frac{dN}{dt} = -\sigma_a \psi_0 \phi_0 N \quad (\text{transmutation equation}) \quad \text{VII-20}$$

$$N(0) \equiv N_0 \quad (\text{initial condition}) \quad \text{VII-21}$$

Because of the simplistic nature of this problem, the lambda eigenvalue is found independently of  $N$  or  $\psi$ ,

$$\lambda = \frac{1}{k_\infty} = \frac{\sigma_a}{v \sigma_f}, \quad \text{VII-22}$$

and does not vary with time even though the flux and atom density are time-dependent.

Equation VII-18, which is to be solved for the flux-shape function, is actually satisfied by any constant; however, from the normalization constraint, the value for  $\psi_0$  is fixed to be unity.

The flux magnitude is easily computed from Eq. VII-19:

$$\phi_0 = \frac{P}{N_0 \sigma_f}, \quad \text{VII-23}$$

and the time-dependent nuclide concentration is found to be

$$N(t) = N_0 e^{-\sigma_a \phi_0 t} = N_0 e^{-\frac{\sigma_a P t}{N_0 \sigma_f}}. \quad \text{VII-24}$$

For this example the response has been defined as the concentration of nuclide N at some specified time  $T_f$  (a "final-time nuclide response"),

$$R \equiv N(T_f) = \int_0^{T_f} \delta(t - T_f) N(t) dt = N_0 e^{-\sigma_a \phi_0 T_f}. \quad \text{VII-25}$$

Now observe the consequences of perturbing the initial condition

by  $N_0 \rightarrow N_0 + \Delta N_0$

a) from Eq. VII-23

$$\phi_0 \rightarrow \frac{P}{(N_0 + \Delta N_0) \sigma}$$

b) from Eq. VII-24

$$N(t) \rightarrow N(t) + \Delta N(t) = (N_0 + \Delta N_0) e^{-\frac{\sigma_a P t}{(N_0 + \Delta N_0) \sigma_f}}$$

c) from Eq. VII-25

$$R \rightarrow R + \Delta R = (N_0 + \Delta N_0) (e^{-\sigma_a \phi_0 T_f}) (e^{-\sigma_a \Delta \phi_0 T_f}) \quad \text{VII-26}$$

The expression in (c) corresponds to the "exact" perturbed response, accurate within the limitations of the quasi-static formulation. Note that if the flux and nuclide fields were assumed to be uncoupled, a perturbation in  $N_0$  would not affect  $\phi_0$  (i.e.,  $\Delta \phi_0 = 0$ ). Equation VII-25 then reduces to

$$\Delta N(t) = \Delta N_0 e^{-\sigma_a \phi_0 t},$$

and the response would be perturbed by

$$\frac{\Delta R}{R} = \frac{\Delta N_0}{N_0}. \quad \text{VII-27}$$

Therefore the initial-condition sensitivity coefficient for the uncoupled case is 1.

The effect of the flux perturbation in Eq. VII-25 can be approximated in the following manner: using the fact that  $\Delta \phi_0 \sim -\phi_0 \Delta N_0 / N_0$  (accurate to second order), Eq. VII-26 can be written as

$$R + \Delta R \sim (N_0 + \Delta N_0) e^{-\sigma_a \Phi_0 T_f} \cdot e^{\sigma_a \Phi_0 \frac{\Delta N_0}{N_0} T_f} \quad \text{VII-28}$$

Expanding the last exponential in a Maclaurin series, and neglecting all but first-order terms,

$$\Delta R = \Delta N_0 \left\{ e^{-\sigma_a \Phi_0 T_f} + T_f \sigma_a \Phi_0 e^{-\sigma_a \Phi_0 T_f} \right\} \quad \text{VII-29}$$

This implies that

$$\frac{\Delta R}{R} = \frac{\Delta N_0}{N_0} \left\{ 1 + T_f \sigma_a \Phi_0 \right\}, \quad \text{VII-30}$$

with the term in brackets serving as the sensitivity coefficient.

Comparing the sensitivity coefficients for the coupled (Eq. VII-30) and uncoupled (Eq. VII-27) cases, we conclude that the term  $T_f \sigma_a \Phi_0$  arises from the coupling between the flux and nuclide fields.

Now consider the adjoint system for this example. The value for  $\Gamma_0^*$ , the shape function adjoint, is obviously zero from the orthogonality condition. The equation for the nuclide adjoint is

$$-\frac{dN^*}{dt} = -\sigma_a \Phi_0 N^*, \quad t < T_f \quad \text{VII-31}$$

with

$$N^*(T_f) = \frac{\partial R}{\partial N} = 1, \quad t = T_f. \quad \text{VII-32}$$

This final-value problem has the solution

$$N^*(t) = e^{-\sigma_a \Phi_0 (T_f - t)} \quad \text{VII-33}$$

The value for the normalization adjoint at  $t = 0$  is given by

Eq. IV-62, with  $\partial R / \partial \Phi_i = 0$ :

$$P^* = \frac{\int_0^{T_f} N^* (-\sigma_a) N dt}{\sigma_f N_0} = - \frac{\sigma_a N(T_f) T_f}{\sigma_f N_0}, \quad \text{VII-34}$$

and the value for  $\Pi$  in Eq. IV-57 is

$$\Pi_0 = \Phi_0 \sigma_f. \quad \text{VII-35}$$

Substituting Eqs. VII-33, 34, 35 into Eq. IV-60 for the sensitivity coefficient gives, after simplification

$$S_0 = \left\{ 1 + T_f \sigma_a \Phi_0 \right\}, \quad \text{VII-36}$$

which is the same value as in Eq. VII-30. Thus we see that the coupled adjoint equations provide a first-order estimate of the effect of the nonlinear coupling between the flux and nuclide fields, which does not appear in the uncoupled case. Of course, if the nuclide/flux coupling were ignored, then  $P^*$  would be zero and the sensitivity coefficient in Eq. VII-36 would reduce to the uncoupled value of 1.

This example has illustrated that a change in some nuclide concentration can perturb a response not only through transmutation but

also by a change in the flux magnitude, which is accounted for with depletion perturbation theory by a "P\*" effect."

For the second example we consider the indirect effect of a change in the flux shape arising from a perturbation in the nuclide field. Recall that a change in  $\psi$  can either be due to a change in the spatial distribution [the total area under  $\psi(r)$  must be one], or due to a change in the energy spectrum. As an example of this effect, we will examine the case when the flux spectrum is perturbed. This time the problem will be described by two energy groups and an infinite homogeneous medium composed of one fuel nuclide and one poison nuclide (the infinite-medium restriction can be relaxed if the flux is separable in space, and if the buckling term corresponding to the finite system is added to the flux equation). For simplicity we again only consider one time step. The response considered is the concentration of the fuel nuclide after 600 days of exposure. In this example the following notation will be employed:

$\sigma_{xj}^k$  = micro-cross-section of type x; for nuclide k, group j.

Cross-section types are indicated by r for removal, a for absorption, c for capture, f for fission, and s for scatter

$N_1(t)$  = atom density of fuel nuclide

$N_2(t)$  = atom density of poison nuclide

$\zeta(t) = N_2(t)/N_1(t)$

$\underline{N}_0 = \text{initial condition} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \times 10^{24} \text{ atoms/cm}^3$

The burnup equations describing the system are assumed to be the following:

Flux-shape equation

$$\begin{pmatrix} N_1(t) \sigma_{r1}^1 & 0 \\ -N_1(t) \sigma_{s,1-2}^1 & N_1(t) \sigma_{a2}^1 + N_2(t) \sigma_{c2}^2 \end{pmatrix} \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} - \lambda \begin{pmatrix} 0 & N_1(t) \nu \sigma_{f2}^1 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} = 0$$

VII-38

which can be written

$$\begin{pmatrix} \sigma_{r1}^1 & 0 \\ -\sigma_{s,1-2}^1 & \sigma_{a2}^1 + \zeta(t) \sigma_{c2}^2 \end{pmatrix} \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} - \lambda \begin{pmatrix} 0 & \nu \sigma_{f2}^1 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} = 0$$

VII-39

Flux-normalization equation

$$N_1 \sigma_{f2}^1 \psi_2 \Phi = P,$$

$$\Phi = \frac{P}{N_1 \sigma_{f2}^1 \psi_2}$$

VII-40

Nuclide-transmutation equation

$$\begin{pmatrix} -(\sigma_{a1}^1 \psi_1 + \sigma_{a2}^1 \psi_2) \cdot \Phi & 0 \\ \gamma \sigma_{f2}^1 \psi_2 \cdot \Phi & -(\Phi \sigma_{c2}^2 \psi_2 + \Lambda) \end{pmatrix} \begin{pmatrix} N_1 \\ N_2 \end{pmatrix} = \frac{d}{dt} \begin{pmatrix} N_1 \\ N_2 \end{pmatrix}$$

VII-41

where

$\gamma$  = yield of nuclide 2 from fission,

$\Lambda$  = decay constant of nuclide 2.

It is a straightforward, though somewhat laborious task, to obtain closed-form solutions to Eqs. VII-39, 40, 41. For the general case of

several time-steps in the quasi-static calculation, the expressions are very involved; however, if we stay with our original assumption and use only a single time-step, the resulting expressions are more manageable. The solutions are summarized below:

$$\lambda = \frac{\sigma_{r1}^1 (\sigma_{a2}^1 + \zeta(t) \sigma_{c2}^2)}{(\nu \sigma_{f2}^1) (\sigma_{s,1-2}^1)} \quad \text{VII-42}$$

$$\psi_1/\psi_2 = \frac{(\sigma_{a2}^1 + \zeta(t) \sigma_{c2}^2)}{\sigma_{s,1-2}^1} \quad \text{VII-43}$$

$$\Phi = \frac{P}{N_1 \sigma_{f2}^1 \psi^2} \quad \text{VII-44}$$

$$N_1(t) = N_1(0) e^{-a_{11}t} \quad \text{VII-45}$$

$$N_2(t) = N_2(0) e^{-a_{22}t} + \frac{N_1(0) a_{21}}{a_{22} - a_{11}} [e^{-a_{11}t} - e^{-a_{22}t}] \quad \text{VII-46}$$

where  $a_{ij}$  refers to the elements of the matrix in Eq. VII-41.

The nuclide adjoint equation is obtained by simply transposing the matrix in Eq. VII-41. The resulting nuclide adjoint solutions are

$$N_1^*(t) = N_1^*(T_f) e^{-a_{11}(T_f-t)} - \frac{a_{21}}{a_{22} - a_{11}} N_2^*(T_f) \{ e^{-a_{22}(T_f-t)} - e^{-a_{11}(T_f-t)} \}$$

$$N_2^*(t) = N_2^*(T_f) e^{-a_{22}(T_f-t)} \quad \text{VII-47}$$

where  $a_{ij}$  again refers to the matrix elements in Eq. VII-41.

The value for the flux-normalization adjoint is given by

$$p^* = \frac{\int_0^{T_f} \left\{ N_1^*(t) a_{11} N_1(t) + N_2^*(t) a_{21} N_1(t) + N_2^*(t) a_{22} N_2(t) \right\} dt}{\psi_2 \sigma_{f2}^1} \quad \text{VII-48}$$

which can be integrated analytically.

The equation for the shape adjoint function is obtained by transposing Eq. VII-39, and setting the result equal to the adjoint source defined in Eq. IV-45. For an infinite, homogeneous medium, in which  $\Gamma^*$  is orthogonal to the fission source the fission term can be ignored (see Appendix C), which makes the equation for  $\Gamma^*$  particularly simple:

$$\begin{pmatrix} \sigma_{r1}^1 & -\sigma_{s,1-2}^1 \\ 0 & \sigma_{a2}^1 + \zeta(t) \sigma_{c2}^2 \end{pmatrix} \begin{pmatrix} \Gamma_1^* \\ \Gamma_2^* \end{pmatrix} = \begin{pmatrix} Q_1^* \\ Q_2^* \end{pmatrix} \quad \text{VII-49}$$

where

$$Q_1^* = \Phi \int_0^{T_f} dt N_1^*(t) (-\sigma_{a1}^1) N_1(t)$$

$$Q_2^* = \Phi \int_0^{T_f} dt \left\{ N_1^*(t) (-\sigma_{a2}^1) N_1(t) + N_2^*(t) (\gamma \sigma_{f2}^1) N_1(t) + N_2^*(t) (\sigma_{c2}^2) N_2(t) \right\}$$

$$- \Phi P^* N_1(0) \sigma_{f2}^1 \quad \text{VII-50}$$

These expressions can be evaluated analytically using the terms in Eqs. VII-44, 45, 46. For this example the various data values were assumed to be those given in Table VII-4. These values are not particularly realistic, and were chosen arbitrarily to illustrate the technique. Using this data, the values for  $\Phi$ ,  $\psi$  and  $\underline{N}$  were computed "semi-analytically" (i.e., a computer program was written to evaluate the analytic expressions and couple the results), and are listed in Column 1 of Table VII-5.

The response considered in this particular example was the concentration of nuclide 1 after 600 days of exposure. Therefore, the appropriate final condition for the nuclide adjoint is

$$N_1^*(600) = 1$$

$$N_2^*(600) = 0$$

The results of the adjoint calculations for this response are given in Table VII-6.

Now consider the change in the final concentration of the fuel nuclide, due to varying the initial concentration of the poison nuclide. A change in the concentration of nuclide 2 does not directly affect nuclide 1, since nuclide 1 is not produced by nuclide 2 (note that  $N_2^*(t) = 0$ ). The poison nuclide was also assumed to have a zero fission cross section, and hence does not affect the flux normalization directly. Therefore the only mechanism by which a change in the concentration of nuclide 2 will affect the final concentration of nuclide 1 is through a change in the flux spectrum.

Table VII-4. Assumed values for nuclear data in  $\Gamma^*$  example

Parameter	Value
$\sigma_{r1}^1$	9 barns
$\sigma_{c1}^1$	3 b
$\sigma_{l,1-2}^1$	6 b
$\sigma_{c2}^1$	1 b
$\sigma_{a2}^1$	2b
$\sigma_{f2}^1$	.1b
$\sigma_{c2}^2$	10b
$\chi_1$	1
$\chi_2$	0
$\gamma$	.5
P	$2.0 \times 10^{14} \frac{\text{fissions}}{\text{sec-cm}^3}$
$\Lambda$	$4.0 \times 10^{-9} \text{ sec}^{-1}$

Table VII-5. Results of forward calculation in  $\Gamma^*$  example

	Reference case		Perturbed case ( $\Delta N_2 = .1$ )	
	t = 0	t = 600 days	t = 0	t = 600 days
$N_1$	$1.0 \times 10^{24}$	$.96937 \times 10^{24}$	$1.0 \times 10^{24}$	$.96436 \times 10^{24}$
$N_2$	0.0	$.17533 \times 10^{23}$	$.10 \times 10^{24}$	$.95125 \times 10^{23}$
$\Phi \cdot \psi_1$	$.6667 \times 10^{14}$	$.74992 \times 10^{14}$	$.1000 \times 10^{15}$	$.10323 \times 10^{15}$
$\Phi \cdot \psi_2$	$.2000 \times 10^{15}$	$.20632 \times 10^{15}$	$.2000 \times 10^{15}$	$.20739 \times 10^{15}$
$k_{\text{eff}}$	1.500	1.380	1.000	1.005

Table VII-6. Results of adjoint calculation<sup>a</sup> in  $\Gamma^*$  example

	t = 0	t = 600
$N_1^* (0^+)$	.96937	1.0
$N_2^* (0^+)$	0.0	0.0
$\bar{\Gamma}_1^*$	$5.0164 \times 10^{17}$	
$\Gamma_2^*$	$6.7008 \times 10^{21}$	
$p^*$	$1.5076 \times 10^8$	

<sup>a</sup>For a response of  $R = N_1$  (600 days).

Column 2 of Table VII-5 shows the results of the perturbed calculation, for a change in the initial condition of the poison nuclide equal to  $.1 \times 10^{24}$  atoms/cm<sup>3</sup>. As one would expect, the addition of the poison nuclide hardens the spectrum, which increases the rate of depletion of nuclide 1, because nuclide 1 was assumed to have a higher absorption cross section in group 1 than in group 2. Consequently, after 600 days' exposure the concentration of nuclide 1 (i.e., the response) is slightly lower for the perturbed case than for the reference case. The amount of the response perturbation is  $-.52\%$ .

We would now like to predict the response change using perturbation theory, and compare with the direct calculation. For the perturbation of

$$\underline{\Delta N} = \begin{pmatrix} 0 \\ .1 \end{pmatrix} \times 10^{24} ,$$

Equation IV-59 reduces to

$$\Delta R/R = \frac{-.1 \times 10^{24}}{.96937 \times 10^{24}} (\Gamma_2^* \sigma_{c2}^2 \psi_2) = -.52\% .$$

From this result we see that the perturbation method accurately accounts for changes in flux shape with the  $\Gamma^*$  term. This illustrates that the nuclide importance depends on the importance of the flux shape through a " $\Gamma^*$  effect."

We can summarize the results of this chapter as follows:

1. For an uncoupled nuclide field (i.e., one which does not perturb the neutron field in which it resides), it has been shown that

$\underline{N}^*$  can be interpreted as the importance of the nuclide field to the response. This is analogous to the role played by  $\phi^*$  for the uncoupled neutron field.

2. The principle of conservation of nuclide importance for an uncoupled nuclide field has been demonstrated.

3. For coupled neutron/nuclide fields, the general concept of "field-importance" has been defined for small deviations about the reference state solution to the initial-value formulation of the burnup equations. Specifically,  $\underline{N}^*$  is the importance of the nuclide field and  $\Gamma^*$  is the importance of the flux field. It was shown that the importance of one field depends on the importance of the other.

4. It has been shown that field-importance is conserved for small deviations (in which the perturbed fields obey the linearized burnup equations) about the reference state solution; however, "response" contained in one field may be transferred to the other.

5. In the quasi-static formulation it has been shown that  $\underline{N}^*$  corresponds to the importance of changes in the nuclide field,  $P^*$  to the importance of changes in the flux magnitude, and  $\Gamma^*$  to the importance of changes in the flux shape. As in the initial value formulation, the quasi-static adjoint functions are coupled in a manner that accounts for the coupled perturbations in the forward equations. This fact was illustrated by two example calculations for the nuclide adjoint function. The calculations showed that the total importance of the

nuclide field contains a " $P^*$  effect" to account for changes in flux magnitude, and a " $\Gamma^*$  effect" to account for changes in flux shape.

## CHAPTER VIII

### APPLICATION OF UNCOUPLED DEPLETION SENSITIVITY THEORY TO ANALYSIS OF AN IRRADIATION EXPERIMENT

One of the uses of static sensitivity theory which has evolved over the last five years is to aid in the design and analysis of integral experiments used in evaluating nuclear data. In particular, the sensitivity coefficients may be employed

- (a) to assess the effect of uncertainties in differential data on computed integral responses;
- (b) to determine if the measured integral parameters are sensitive to the data of interest;
- (c) to adjust differential data to minimize discrepancies between calculated and measured integral parameters; and
- (d) to assign priorities and required accuracies for differential data measurements (the "inverse problem").

In the past, the integral parameters have been limited to static responses, such as reaction-rate ratios, measured in various critical assemblies. With the development of depletion sensitivity theory, however, a much wider range of integral experiments can be addressed. For example, with this technique one may analyze "irradiation experiments"; i.e., those in which a small sample is exposed to a known flux field for a relatively long period of time. By chemically analyzing the transmutation products in the irradiated sample it is

possible to back out useful, integrated reaction rates. Figure VIII-1 is a flow chart depicting how depletion sensitivity calculations could fit into the data-evaluation stream. By iterating between sensitivity analysis and cross-section measurement, an acceptable set of differential data is eventually obtained, which allows reactor design parameters to be computed to within allowed tolerances.

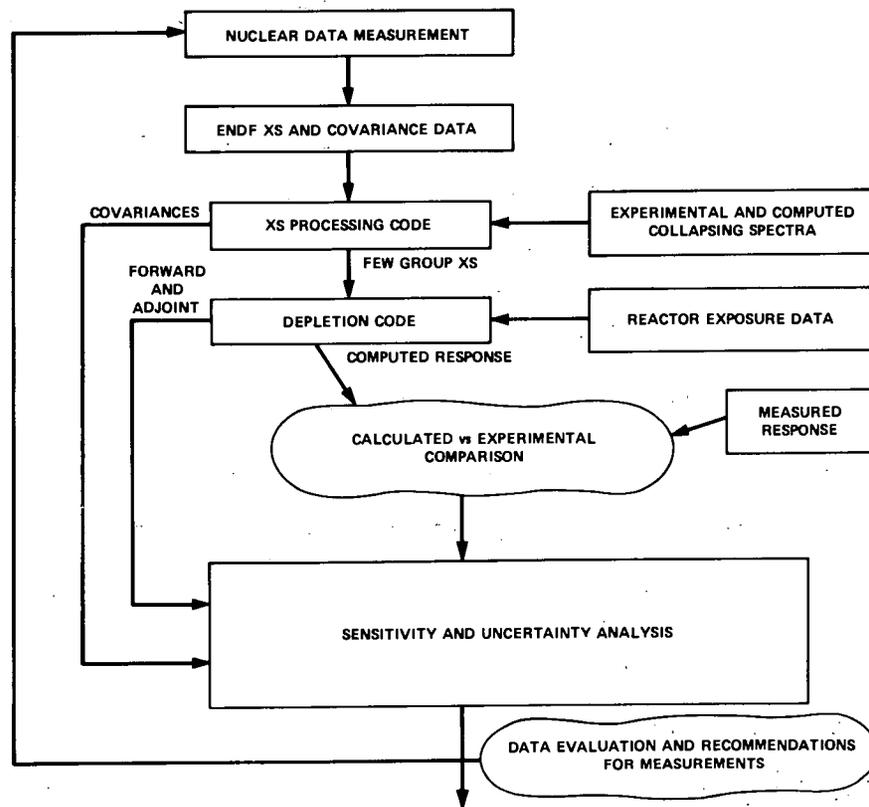


Fig. VIII-1. Flow-chart of calculations in depletion sensitivity analysis.

There is currently an ongoing project in the ORNL Physics Division to improve the higher actinide cross-section data (46). One facet of this project is the analysis of several integral irradiation experiments. The Engineering Physics Division is providing computational support for the integral experiment program, and as part of this analysis has performed depletion sensitivity and uncertainty calculations. Because of the small sample size (< 100 mg) it can be assumed that the neutron field is unperturbed by the nuclide field of the sample; therefore it was decided to use uncoupled perturbation theory for the analysis. This is the first known application of uncoupled, depletion perturbation theory to experiment analysis. Details of the experiment are given below (28).

In 1966, several actinide samples ranging from  $^{232}\text{Th}$  to  $^{241}\text{Pu}$  were irradiated for four years in the fast reactor EBR-II at Argonne National Laboratory (ANL), Idaho. The purpose of this research was to experimentally ascertain the isotopic composition of the irradiated sample. However, after one sample had been analyzed, the ANL program was halted until 1977 when interest was revived in obtaining better actinide cross sections in the higher energy range. At that time the other irradiated samples were sent to ORNL for further analysis as part of its cross-section measurement program. Oak Ridge has partially completed examination of the second sample, which was nominally 94.1 mg  $^{239}\text{PuO}_2$  with some impurities present.

The initial composition of this sample is given in Table VIII-1, and the exposure history and 14-group flux spectrum (both provided by ANL) are given in Tables VIII-2 and VIII-3, respectively.

Table VIII-1. Initial composition of  $^{239}\text{Pu}$  sample

Nuclide	Gm-atoms
$^{239}\text{Pu}$	$5.45 \times 10^{-4}$
$^{240}\text{Pu}$	$2.62 \times 10^{-5}$
$^{241}\text{Pu}$	$1.86 \times 10^{-6}$
$^{242}\text{Pu}$	$1.07 \times 10^{-7}$
$^{241}\text{Am}$	$4.61 \times 10^{-7}$

Fourteen-group cross sections were processed from preliminary ENDF/B-V data (47) using MINX (48), and were collapsed to one group using the EBR-II spectrum. The effective cross sections for important nuclides are shown in Table VIII-4, and the one-group uncertainties for some of the plutonium data are given in Table VIII-5 (49). (When this study was done, these were the only covariance files available.)

Because uncoupled sensitivity theory was deemed adequate for this study, the forward and adjoint nuclide fields could be computed with the ORIGEN-A code. Table VIII-6 gives a comparison of the computed and measured percentages of plutonium isotopes in the irradiated sample (at present, only the Pu isotopes have been experimentally analyzed). The agreement for the Pu isotopes is fairly good.

Thus far the results presented here have been obtained with "standard" analysis methods (except for possibly generating data uncertainties). But we will now begin to utilize new techniques; namely,

Table VIII-2. Exposure history of  $^{239}\text{Pu}$  sample<sup>a</sup>

Days <sup>b</sup>	Power (MW)	$\Phi$ ( $\times 10^{-15}/\text{cm}^2 \cdot \text{sec}$ )	Days	Power (MW)	$\Phi$ ( $\times 10^{-15}/\text{cm}^2 \cdot \text{sec}$ )
0-25	25.20	1.24	700-709	45.80	2.26
193-260	23.30	1.15	710-712	55.50	2.73
290-297	12.43	.611	722-742	42.60	1.82
309-349	25.73	1.27	743-749	41.50	1.77
351-372	29.48	1.45	750-752	36.00	1.53
424-451	10.48	.515	753-758	44.80	1.91
451-455	44.25	2.18	812-844	18.75	.799
457-461	15.50	.762	853-868	38.40	1.64
480-487	40.86	2.01	871-889	45.67	1.95
488-492	22.50	1.11	890-897	44.28	1.89
494-497	22.67	1.15	905-933	42.93	1.83
498-500	24.5	1.20	937-957	40.00	1.71
506-513	49.43	1.45	959-968	44.44	1.89
514-517	32.00	1.57	972-998	42.71	1.82
520-524	38.50	1.20	1004-1027	42.78	1.82
526-538	25.25	1.45	1032-1045	46.15	1.97
540-557	39.35	1.57	1106-1131	30.84	1.31
568-577	20.89	1.89	1135-1140	37.00	1.58
581-583	12.0	1.24	1140-1149	46.40	1.97
587-594	29.29	1.93	1152-1162	44.3	1.89
597-619	32.27	1.59	1162-1181	48.63	2.07
624-639	43.47	2.14	1185-1205	48.05	2.05
641-643	41.5	2.04	1207-1212	31.90	1.34
645-649	13.00	.639	1229-1259	44.80	1.91
651-655	19.75	.971	1267-1295	48.21	2.06
656-659	26.33	1.29	1298-1317	47.37	2.02
664-668	22.25	1.09	1327-1337	45.60	1.92
675-682	22.71	1.12	1342-1356	46.07	1.96
683-686	25.00	1.38	1359-1374	47.00	2.00
690-698	46.50	2.29			

<sup>a</sup>Total exposure = 27,676 MWd;  $\Phi \cdot T = 1.0661 \times 10^{-1}$  barns<sup>-1</sup>.

<sup>b</sup>Days not shown indicate shutdown.

Table VIII-3. EBR-II flux spectrum

Upper energy bound	Multigroup flux spectrum ( $\psi$ )
$.1000 \times 10^8$	.074
$.2231 \times 10^7$	.087
$.1353 \times 10^7$	.120
$.8209 \times 10^6$	.341
$.3020 \times 10^6$	.245
$.1111 \times 10^6$	.098
$.4087 \times 10^5$	.027
$.1503 \times 10^5$	.006
$.5531 \times 10^4$	.0007
$.3355 \times 10^4$	.0004
$.2035 \times 10^4$	.0003
$.4540 \times 10^3$	0
$.1013 \times 10^3$	0
$.1371 \times 10^2$	0

Table VIII-4. One-group, preliminary ENDF/B-V cross sections for EBR-II

Data	Effective value
$^{239}\text{Pu } \sigma_f$	1.66
$^{239}\text{Pu } \sigma_c$	.154
$^{240}\text{Pu } \sigma_f$	.644
$^{240}\text{Pu } \sigma_c$	.213
$^{241}\text{Pu } \sigma_f$	.175
$^{241}\text{Pu } \sigma_c$	.205
$^{242}\text{Pu } \sigma_f$	.506
$^{242}\text{Pu } \sigma_c$	.212
$^{241}\text{Am } \sigma_f$	.553
$^{241}\text{Am } \sigma_c$	.782

Table VIII-5. Uncertainties in Pu nuclear data

Data	Standard deviation (%)
$^{239}\text{Pu}$ $\sigma_c$	6.7
$^{239}\text{Pu}$ $\sigma_f$	3.0
$^{240}\text{Pu}$ $\sigma_c$	10.0
$^{241}\text{Pu}$ $\sigma_f$	12.0
$^{241}\text{Pu}$ $\sigma_c$	3.0
$^{241}\text{Pu}$ $\sigma_f$	2.7
$^{241}\text{Pu}$ decay constant	2.7

Table VIII-6. Comparison of measured and calculated Pu isotopics

Pu Isotope	Measured (%)	Calculated (%)
238	.020	.012
239	93.15	93.01
240	6.56	6.57
241	.251	.266
242	.026	.030

sensitivity coefficients and uncertainty analysis. For this particular sample four responses were considered. These corresponded to the concentrations of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{241}\text{Am}$  in the irradiated sample. Table VIII-7 gives the sensitivities of these concentrations to the indicated data used in the calculation.

The sensitivity coefficients may be interpreted as follows: If  $\alpha_{i,j}$  corresponds to the sensitivity coefficient for response  $R_i$  to data  $\sigma_j$ , then a 1% increase in the value of  $\sigma_j$  will cause an increase of  $\alpha_{i,j}$  in  $R_i$ ; i.e.,

$$\left(\frac{\Delta R_i}{R_i}\right) \% = \alpha_{i,j} \left(\frac{\Delta \sigma_j}{\sigma_j}\right) \% .$$

For example, we see that if the  $^{239}\text{Pu}$  capture cross section is increased by 1%, then the  $^{239}\text{Pu}$  concentration in the irradiated sample will decrease by about .016%, while the  $^{240}\text{Pu}$  concentration increases by about .24% and the  $^{241}\text{Pu}$  concentration increases by about .046%. The  $^{241}\text{Am}$  concentration is quite insensitive to the  $^{239}\text{Pu}$  capture cross section because it is far up the nuclide chain.

Some very important insight into the physics of transmutation can be obtained by careful examination of these sensitivity coefficients. Some of the conclusions of the sensitivity study are intuitive, while others are surprising.

For example, we can see from Table VIII-7 that  $^{239}\text{Pu}$  is most sensitive to the  $^{239}\text{Pu}$  fission cross section, and to its initial

Table VIII-7. Sensitivity coefficients for irradiated  $^{239}\text{Pu}$  sample

Data Parameter <sup>a</sup>	Specified Response <sup>b</sup>			
	R1	R2	R3	R4
Cross-Section Sensitivity Coefficients				
P9 $\sigma_C$	5.30-3	4.58-2	2.45-1	-1.64-2
P9 $\sigma_F$	-2.27-4	-2.75-3	-1.20-3	-1.77-1
P0 $\sigma_C$	5.47-2	3.06-1	-2.01-2	0
P0 $\sigma_F$	-1.19-3	-1.07-2	-6.09-2	0
P1 $\sigma_C$	-3.39-3	-1.83-2	0	0
P1 $\sigma_F$	-2.89-3	-2.56-2	0	0
P1 decay constant	3.91-1	-1.42-1	0	0
A1 $\sigma_C$	-6.96-2	0	0	0
A1 $\sigma_F$	-4.92-2	0	0	0
Initial Condition Sensitivity Coefficients				
P9	5.31-3	4.62-2	2.48-1	1.0
P0	5.01-2	2.65-1	7.54-1	0
P1	3.72-1	6.89-1	0	0
A1	5.75-1	0	0	0

<sup>a</sup>P9 indicates  $^{239}\text{Pu}$ , P0 indicates  $^{240}\text{Pu}$ , etc.

<sup>b</sup>Concentration after 1374 days irradiation: R1 =  $^{241}\text{Am}$ ,  
R2 =  $^{241}\text{Pu}$ , R3 =  $^{240}\text{Pu}$ , R4 =  $^{239}\text{Pu}$ .

condition. These conclusions are probably obvious, although one may be surprised that the sensitivity coefficient for the fission cross section is relatively small.  $^{240}\text{Pu}$  is most sensitive to its initial concentration, the initial concentration of  $^{239}\text{Pu}$ , and the capture cross section of  $^{239}\text{Pu}$ . The sensitivity coefficients for the last two parameters are essentially the same; i.e., an increase of X% in the concentration of  $^{239}\text{Pu}$  has the same effect on  $^{240}\text{Pu}$  as an increase of X% in the  $^{239}\text{Pu}$  capture cross section. The final concentration of  $^{240}\text{Pu}$  is relatively insensitive to its own absorption cross section (sensitivity coefficient  $\sim .08$ ).  $^{241}\text{Pu}$  is most sensitive to its initial concentration, its decay constant, and to the initial concentration and capture cross section of  $^{240}\text{Pu}$ .  $^{241}\text{Am}$  is most sensitive to its initial concentration, and to the initial concentration and the decay constant of  $^{241}\text{Pu}$ . Note that it is insensitive to both its fission and capture cross sections.

Recall now that this sample is supposed to be a  $^{239}\text{Pu}$  sample — the other isotopes are merely impurities. However, in many cases we can see that the response of interest is very sensitive to the concentration of impurities in the sample. A graphic example is the  $^{241}\text{Am}$  concentration. It was originally hoped that this sample could be used to provide integral data for  $^{241}\text{Am}$  cross sections, which were known to be poor in ENDF/B-IV. However, we have already seen that the  $^{241}\text{Am}$  concentration in the irradiated sample is not sensitive to these cross sections! In fact, by examining the sensitivity coefficients we conclude that most of the  $^{241}\text{Am}$  contained in the irradiated sample was either there

originally as an impurity or came from the decay of the  $^{241}\text{Pu}$  which was originally in the sample as an impurity.

Uncertainty analysis has also been performed for this sample to ascertain the effect of uncertainties in the plutonium data on the computed responses. Using the data uncertainties given in Table VIII-5, page 130, the values in Table VIII-8 were found for the standard deviations of the responses. The differences between computed and measured values for both  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are within the uncertainties due to data, while the  $^{241}\text{Pu}$  difference is within two standard deviations. The computed standard deviations do not reflect uncertainties in the initial composition of the sample.

Table VIII-8. Computed uncertainties in concentrations in irradiated sample, due to uncertainties in Pu data

Data <sup>a</sup>	$\delta R2/R2(\%)^b$	$\delta R3/R3(\%)$	$\delta R4/R4(\%)$
P9 $\sigma$	3.0-1	1.6	1.1-1
P9 $\sigma^C$	8.3-3	3.6-3	5.3-1
P0 $\sigma^F$	3.1	2.1-1	0
P1 $\sigma^C$	2.3-1	0	0
P1 $\sigma^F$	4.7-2	0	0
P1 $\lambda$	3.8-1	0	0
Totals:	3.1%	1.6%	.54%

<sup>a</sup>P9 indicates  $^{239}\text{Pu}$ , P0 indicates  $^{240}\text{Pu}$ , etc.

<sup>b</sup>R2 =  $^{241}\text{Pu}$ , R3 =  $^{240}\text{Pu}$ , R4 =  $^{239}\text{Pu}$ .

This example shows that depletion sensitivity analysis can be used not only to determine error bounds on a computed response, but also to provide insight into the physical phenomena taking place during irradiation. This method will be used in the future to analyze other samples for the same cross-section measurement program.

## CHAPTER IX

### APPLICATION OF COUPLED DEPLETION SENSITIVITY THEORY TO EVALUATE DESIGN CHANGES IN A DENATURED LMFBR

In the previous chapter depletion sensitivity theory was used to examine the effect of variations in basic nuclear data on integral parameters. Although the uncoupled formulation was employed, a similar type of analysis can be performed with coupled sensitivity theory if the problem of interest warrants the added complexity. This chapter will address another area of application for depletion sensitivity theory, which could be of significant importance in reactor design.

The problem can be simply stated as follows: Suppose that a reactor designer has determined a "reference" design for some reactor, and has performed a detailed depletion calculation to evaluate its performance over several operating cycles. A measure of the "quality" of the design is usually some set of integral parameters such as end-of-cycle (EOC) reactivity, net fissile gain (for a breeder) over a cycle, peak-to-average power ratio, etc., which the designer wishes to maximize or minimize. To optimize the set of integral parameters the designer may adjust either the beginning-of-life (BOL) reactor design or the reactor operating conditions (e.g., the burnup). Depletion sensitivity analysis is ideally suited for the former case, since it can efficiently relate changes in the initial condition of the reactor to changes in integral parameters at EOC without requiring expensive depletion calculations.

It is possible that an optimization program could be established using this method, along with a technique such as linear programming, which could make small variations about the reference design until the "best" configuration is determined. However, because linear perturbation theory is being used, only "small" variations are allowed, so that second-order effects do not become significant. This means that the reference state would have to be reasonably close to optimum. Nevertheless, it is well known that a small improvement in reactor performance (e.g., a reduction in fissile inventory or an increase in breeding gain, etc.) can mean a substantial savings in fuel-cycle costs.

It is not the purpose of this text to present a detailed plan for optimization (this is recommended for "future work"); however, we will now present an example application of coupled depletion sensitivity theory to a fairly complex LMFBR model, which illustrates that the method can accurately predict changes in EOC nuclide inventories when the concentrations of various nuclides at BOL are perturbed.

For this calculation, a one-dimensional spherical model of a 20% denatured LMFBR was employed. The model consisted of two regions (a fuel zone with outer radius of 117.6 cm and a blanket zone with outer radius of 162.1 cm) which were obtained by homogenizing a detailed six-zone RZ model (50), taken at equilibrium condition. Approximately 50 spatial intervals were used in the calculations. Control rods in the 2-D axial blanket were smeared into the blanket zone for the spherical model. The enrichment of the 1-D model was adjusted slightly to make the reactor critical over the burn cycle. Table IX-1 gives the

Table IX-1. Beginning-of-cycle atom densities  
for denatured LMFB model

Nuclide	Density (atoms/barn·cm)	
	Core Zone	Blanket Zone
$^{232}\text{Th}$	$3.08477 \times 10^{-3}$	$1.14475 \times 10^{-2}$
$^{233}\text{U}$	$7.86960 \times 10^{-4}$	$1.64215 \times 10^{-4}$
$^{235}\text{U}$	$6.25936 \times 10^{-6}$	
$^{238}\text{U}$	$3.93480 \times 10^{-3}$	
$^{239}\text{Pu}$	$1.35231 \times 10^{-4}$	
$^{240}\text{Pu}$	$8.62243 \times 10^{-6}$	
$^{241}\text{Pu}$	$3.26954 \times 10^{-7}$	
$^{242}\text{Pu}$	$1.11058 \times 10^{-8}$	
Na	$8.59359 \times 10^{-3}$	$7.00910 \times 10^{-3}$
$^{16}\text{O}$	$1.69594 \times 10^{-2}$	$2.33575 \times 10^{-2}$
Fe	$9.69531 \times 10^{-3}$	$7.68439 \times 10^{-3}$
Cr	$2.55295 \times 10^{-3}$	$2.02531 \times 10^{-3}$
Ni	$1.94792 \times 10^{-3}$	$1.54384 \times 10^{-3}$
$^{55}\text{Mn}$	$3.54168 \times 10^{-4}$	$2.80708 \times 10^{-4}$
Mo	$2.06598 \times 10^{-4}$	$1.63747 \times 10^{-4}$
Fission Products	$2.125 \times 10^{-4}$	
$^{10}\text{B}$		$7.34638 \times 10^{-5}$
$^{11}\text{B}$		$1.10186 \times 10^{-4}$
$^{12}\text{C}$		$4.58398 \times 10^{-5}$

zone-dependent atom densities. Four-group cross sections (see Table IX-2 for energy structure) were obtained by collapsing existing libraries (51), and a lumped fission product (52) was used. The depletion calculation consisted of a 300-day burn at 3000 MW<sub>th</sub>, for a core burnup of 41,000 MW-D/T. Table IX-3 summarizes the reactor operating conditions.

Table IX-2. Four-group energy structure

Group	Upper Energy (eV)
1	$1.650 \times 10^7$
2	$8.209 \times 10^5$
3	$4.090 \times 10^4$
4	$2.000 \times 10^3$

Table IX-3. Operating characteristics of model LMFBR

	BOC	EOC
Fissile inventory	3161.5 kg	3190.6
$k_{eff}$	1.0673	1.004
Breeding ratio	1.08	1.15
Specific power	.13 MW/kg	.14 MW/kg
Fuel power density	424.0 w/cm <sup>3</sup>	414.6 w/cm <sup>3</sup>

A denatured LMFBR (so called because the major fissile isotope, <sup>233</sup>U, is "denatured" with <sup>238</sup>U in order that it cannot be chemically separated for use in weapons) was chosen for the analysis because of the complexity of the transmutation process. In this type of reactor, both thorium and uranium buildup chains must be considered. Table IX-4 shows the buildup and decay processes which were assumed in the depletion calculation. Note that some of the short-lived intermediate nuclides have been neglected.

Table IX-4. Transmutation processes in denatured LMFBR model

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$^{232}\text{Th}(n,\gamma) \rightarrow ^{233}\text{Pa}(\beta^-) \rightarrow ^{233}\text{U}$
$^{232}\text{Th}(n,2n) \rightarrow ^{231}\text{Pa}(n,\gamma) \rightarrow ^{232}\text{U}$
$^{232}\text{U}(n,\gamma) \rightarrow ^{233}\text{U}(n,\gamma) \rightarrow ^{234}\text{U}(n,\gamma) \rightarrow ^{235}\text{U}(n,\gamma) \rightarrow ^{236}\text{U}$
$^{232}\text{U}(\alpha \text{ decay})$
$^{233}\text{Pa}(n,\gamma) \rightarrow ^{234}\text{U}$
$^{238}\text{U}(n,\gamma) \rightarrow ^{239}\text{Pu}(n,\gamma) \rightarrow ^{240}\text{Pu}(n,\gamma) \rightarrow ^{241}\text{Pu}$
$^{241}\text{Pu}(\beta^- \text{ decay})$
Fissionable Nuclides: $^{232}\text{Th}$ , $^{231}\text{Pa}$ , $^{233}\text{Pa}$ , $^{232}\text{U}$ , $^{233}\text{U}$ , $^{234}\text{U}$ ,
$^{235}\text{U}$ , $^{236}\text{U}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$ , $^{241}\text{Pu}$

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The forward burnup calculations were done with the VENTURE-BURNER code system (32). A new flux shape was computed every 100 days by performing a simple  $k_{\text{eff}}$  calculation (i.e., no control search was done to keep  $k = 1$ ). In addition to the reference VENTURE run, three additional runs were done in which the initial concentrations of  $^{238}\text{U}$ ,  $^{233}\text{U}$  and  $^{232}\text{Th}$  respectively were increased by 5%. The effects of these perturbations on three separate responses were considered. The observed responses were (a)  $^{232}\text{U}$  concentration, (b)  $^{233}\text{U}$  concentration, and (c)  $^{239}\text{Pu}$  concentration, all evaluated after 300 days of exposure. The results of these direct calculations are given in Table IX-5.

The adjoint burnup calculations were performed for each response with the DEPTH module (39) (see Chapter V). The final condition for each of

Table IX-5. VENTURE calculations for perturbed responses<sup>a</sup>  
 due to 5% increase in initial concentrations  
 of indicated nuclides

Initial Condition Perturbed 5%	R1		R2		R3	
	Zone 1	Zone 2	Zone 1	Zone 2	Zone 1	Zone 2
Reference (no perturbation)	1.86421-7 <sup>b</sup>	3.74582-9	6.27921-4	2.08631-4	2.31638-4	0
<sup>238</sup> U concentration	1.85042-7	3.69496-9	6.28503-4	2.08301-4	2.37646-4	0
<sup>233</sup> U concentration	1.83818-7	3.63524-9	6.59435-4	2.14904-4	2.28116-4	0
<sup>232</sup> Th concentration	1.91075-7	3.64674-9	6.33204-4	2.09615-4	2.31319-4	0

<sup>a</sup> Responses are defined as follows (total atoms  $\times 10^{-24}$ ):

- R1 = <sup>232</sup>U inventory
- R2 = <sup>233</sup>U inventory
- R3 = <sup>239</sup>Pu inventory

<sup>b</sup> Read as:  $1.86421 \times 10^{-7}$ .

the runs consisted of an "atom density" of 1.0 for the respective response nuclide, and 0.0 for all others (e.g., the adjoint calculation for the  $^{232}\text{U}$  response had a value of 1.0 for the  $^{232}\text{U}$  concentration and 0.0 for all other nuclides). Using Eq. IV-60, the forward and adjoint solutions were then combined to give the sensitivity coefficients corresponding to each of the three responses for the initial conditions of all nuclides in the system. As in the previous chapter, the initial-value sensitivity coefficient  $\alpha_{i,j}$  relates the percent change in response  $R_i$  to the percent change in the initial concentration of nuclide  $j$ :

$$\left(\frac{\Delta R_i}{R_i}\right)\% = \alpha_{i,j} \left(\frac{\Delta N_j(t=0)}{N_j(t=0)}\right)\%$$

where for this example  $R_i$  is the final concentration (300 days exposure) of either  $^{232}\text{U}$ ,  $^{233}\text{U}$ , or  $^{239}\text{Pu}$ . Table IX-6 gives the sensitivity coefficients of the three responses to the initial conditions of  $^{238}\text{U}$ ,  $^{233}\text{U}$ , and  $^{232}\text{Th}$ , computed with depletion perturbation theory. The sensitivity coefficients indicate some interesting phenomena occurring due to the coupling between the neutron and nuclide fields.

Consider first the response of  $^{232}\text{U}$ . This nuclide is produced by an (n,2n) reaction on  $^{232}\text{Th}$ , and hence we expect  $^{232}\text{Th}$  to have a large direct effect, and indeed the Th sensitivity coefficient is quite large ( $\sim .5$ ). It is more surprising to see a large negative sensitivity coefficient ( $\sim -.3$ ) for  $^{233}\text{U}$ . The reason for this is that  $^{233}\text{U}$  is the dominant fissile nuclide, and hence it is largely responsible

Table IX-6. Sensitivity coefficients computed with perturbation theory for changes in initial conditions

Response <sup>a</sup>	Sensitivity Coefficient to Indicated Initial Condition		
	<sup>238</sup> U	<sup>233</sup> U	<sup>232</sup> Th
R1	$-1.53767 \times 10^{-1}$	$-3.14563 \times 10^{-1}$	$4.68175 \times 10^{-1}$
R2	$1.105442 \times 10^{-3}$	$8.55001 \times 10^{-1}$	$1.43900 \times 10^{-1}$
R3	$5.21633 \times 10^{-1}$	$-3.13106 \times 10^{-1}$	$-2.73917 \times 10^{-2}$

<sup>a</sup>Responses are as follows:

$$R1 = {}^{232}\text{U}$$

$$R2 = {}^{233}\text{U}$$

$$R3 = {}^{239}\text{Pu}.$$

for the power output from the reactor. Since the power is constrained to stay constant, an increase in the <sup>233</sup>U concentration must be accompanied by a decrease in the flux normalization factor in order to keep the product the same; i.e., <sup>233</sup>U has a large "p\* effect." Since adding <sup>233</sup>U makes the flux magnitude decrease, the reactions which produce <sup>232</sup>U must also decrease and therefore the final <sup>232</sup>U concentration is lowered. The <sup>238</sup>U also has a negative sensitivity coefficient for this response because the addition of <sup>238</sup>U tends to soften the flux spectrum, due to inelastic scatter. Since <sup>232</sup>U is produced by a threshold reaction (n,2n), its final concentration is sensitive to a spectral shift, and the end-of-cycle response is lowered. Thus <sup>238</sup>U has a fairly important "I\* effect" because it changes the shape of the flux spectrum.

Consider now the <sup>233</sup>U response. As might be expected, this response is insensitive to the <sup>238</sup>U concentration (there is only a small I\*

effect). An increase in the Th concentration will result in an increase in  $^{233}\text{U}$  since it is contained in the Th buildup chain; however, the sensitivity coefficient is not extremely large ( $\sim .14$ ) because much of the  $^{233}\text{U}$  is in the reactor initially and is not produced from the Th. Obviously, the final  $^{233}\text{U}$  concentration will increase if its initial concentration is increased; however, notice that the sensitivity coefficient is not 1.0 as would be predicted using uncoupled perturbation theory. The coupled perturbation method predicts a sensitivity coefficient of .85, due to the negative  $p^*$  effect.

Finally, the sensitivity coefficients for  $^{239}\text{Pu}$  production contain no real surprises. This response is insensitive to the Th concentration. The  $^{238}\text{U}$  has an important direct effect (sensitivity coefficient = .5) and the  $^{233}\text{U}$  has a large negative sensitivity coefficient (-.3) due to the  $p^*$  effect.

We have thus shown how sensitivity coefficients computed with coupled depletion perturbation theory can help our understanding of the complicated interactions occurring in coupled neutron/nuclide fields. The real practical merit of the method, however, lies in its ability to predict the EOC response changes. Table IX-7 shows the changes in the three responses predicted by perturbation theory and computed exactly with VENTURE. The values in the first column were calculated using the results from Table IX-5, page 137, weighted with the proper volumes. The values in the second column were obtained by simply multiplying 5% by the appropriate sensitivity coefficient from Table IX-6. The agreement is extremely good in all cases. In other calculations not

Table IX-7. Comparison of direct-calculation and perturbation-theory results for response changes due to 5% increase in isotope concentration

Response <sup>a</sup>	$\Delta R/R\%$	
	Direct Calculation	Perturbation Theory
5% Increase in Initial <sup>238</sup> U Concentration		
R1	$-7.6 \times 10^{-1}$	$-7.7 \times 10^{-1}$
R2	$5.2 \times 10^{-3}$	$5.5 \times 10^{-3}$
R3	2.6	2.6
5% Increase in Initial <sup>233</sup> U Concentration		
R1	-1.4	-1.6
R2	4.3	4.3
R3	-1.5	-1.6
5% Increase in Initial <sup>232</sup> Th Concentration		
R1	2.3	2.3
R2	$7.1 \times 10^{-1}$	$7.2 \times 10^{-1}$
R3	$-1.4 \times 10^{-1}$	$-1.4 \times 10^{-1}$

<sup>a</sup>Responses are defined as follows:

R1 = <sup>232</sup>U

R2 = <sup>233</sup>U

R3 = <sup>239</sup>Pu

reported here, depletion sensitivity theory was used to predict changes in the EOC  $k_{\text{eff}}$  due to changes in BOC nuclide concentrations. For these cases also the perturbation theory predictions were very accurate (53).

Although the reactor model assumed for these calculations is not as complex as those used in most design calculations, it does embody most of the general features, such as space-dependent, multi-zone, multigroup fluxes, and multi-zone depletion with multiple transmutation chains. Hence there is some promise that the coupled depletion sensitivity method will be applicable to realistic design problems.

## CHAPTER X

### SUMMARY, CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE WORK

The burnup equations are a system of coupled nonlinear equations describing the time-dependent behavior of the neutron and nuclide fields within a reactor. Burnup analysis is an essential component of reactor design and fuel management studies; however, solving the burnup equations numerically is difficult and expensive for realistic problems. In this text, a technique based on first-order perturbation theory has been developed which allows one to estimate changes in reactor performance parameters arising from small changes in input data without recomputing the perturbed values for the neutron and nuclide fields. The following is a summary of the results and conclusions of the study.

The application of perturbation theory to nonlinear operators has been studied and contrasted to that for linear operators. It was concluded that in order to obtain adjoint equations which are independent of the perturbed forward state, one must deal with "first-order adjoint equations" which are in reality adjoint equations for the linearized forward system.

Various approximations for the burnup equations have been rigorously derived. These formulations included the nonlinear initial-value formulation, the time-continuous eigenvalue formulation, the uncoupled (linear) approximation for the nuclide field, and the quasi-static formulation. For each case, depletion adjoint equations have been

developed. Special attention was devoted to the quasi-static approximation, for which it was shown that there exist three adjoint functions —  $\underline{N}^*$ ,  $P^*$ , and  $\Gamma^*$  — corresponding to the nuclide-field equation, the flux-normalization equation, and the flux-shape equation.

Numerical techniques have been presented for solving the adjoint burnup equations. It was shown that currently available computer codes could be modified in a relatively straightforward manner to obtain adjoint solutions. An adjoint version of the ORIGEN depletion code has been developed. In addition, an algorithm was suggested for implementation into the VENTURE/BURNER Code system to provide quasi-static adjoint solutions. This algorithm has been programmed by J. R. White into a new BOLD VENTURE module called DEPTH.

The new technique of depletion perturbation theory (DPT) has been developed, based on the stationary property of the adjoint burnup solutions. Using DPT, generic sensitivity coefficients have been derived to relate changes in reactor performance parameters (e.g.  $k_{eff}$ , fissile loading, etc.) to changes in nuclear data (cross-sections, decay constants, yield data, etc.) and in the initial reactor loading. Multigroup, multi-zone sensitivity coefficients were written in detail for important types of data. Equations have been presented for uncertainty analysis in burnup calculations.

The relationship between "coupled" and "uncoupled" perturbation theory has been discussed. In uncoupled perturbation theory, it is assumed that the neutron and nuclide fields can be perturbed independently, while in the coupled case a change in one field will automatically perturb the other.

For uncoupled perturbation theory it was concluded that the nuclide adjoint function can be interpreted as the "importance" of a nuclide to a computed response. This led to a postulate of "conservation of nuclide importance" for an uncoupled nuclide field, which is analogous to Lewins' conservation of neutron importance for an uncoupled neutron field. For coupled neutron/nuclide fields, it was concluded that importance can be transferred between the neutron and nuclide fields. A generalization of the importance-conservation principle to the "conservation of field importance" has been suggested for interacting fields. Using this postulate, the coupled nuclide adjoint equation was derived from first principles. It has been shown that for the adjoint quasi-static burnup equations  $N^*$  represents the importance of changes in the nuclide field,  $P^*$  the importance of changes in flux normalization, and  $\Gamma^*$  the importance of changes in the shape of the neutron field. Analytic calculations were performed to illustrate these properties.

An application of uncoupled nuclide perturbation theory to analysis of an irradiation experiment has been presented. Sensitivity coefficients were used to determine the relative importance of various cross-section and decay data affecting the buildup of actinide products in an irradiated  $^{239}\text{Pu}$  sample. It was shown that this type of analysis can provide valuable insight into the physics of transmutation. Time-dependent uncertainty analysis was used to calculate standard deviations in computed actinide concentrations resulting from uncertainties in plutonium cross-section data. For most cases the measured concentrations were within the computed uncertainties of the calculated values.

Depletion perturbation theory for coupled neutron/nuclide fields has been applied to the analysis of a 3000 MW<sub>th</sub> denatured LMFBR model. The model consisted of four energy groups, a core, and a blanket zone treated with approximately 50 spatial intervals, and multiple buildup chains. This model was chosen to illustrate that DPT can be applied to complex depletion problems. Sensitivity coefficients were computed to relate changes in the initial concentrations of various nuclides to the concentrations of other nuclides after 300 days of burnup. An explanation of the physical meaning of the sensitivity coefficients was presented in the context of interactions between the neutron and nuclide fields. Finally, the perturbed, end-of-cycle nuclide concentrations due to various perturbations at beginning-of-cycle were computed with sensitivity theory and by direct re-calculation. In all cases the values predicted with DPT show excellent agreement with the exact values.

The initial results of DPT presented in this study are very encouraging, and there is reason to be optimistic about its potential uses. The basic theory (which will undoubtedly be extended as the need arises) is now well in hand; the numerical methods required to solve the adjoint burnup equations appear manageable (computational needs seem comparable to those for the forward equation); and the examples studied thus far have given excellent results. However, because the field of DPT is very new and still evolving, there are numerous interesting areas which need further study. The following is a list of recommendations for future work:

- (a) Examine the accuracy of DPT in predicting changes in flux-dependent functionals (e.g.  $k_{\text{eff}}$ ).
- (b) Modify (if necessary) adjoint equations to account for batch refueling and additional reactor constraints.
- (c) Implement and test depletion adjoint solution for two-dimensional VENTURE/BURNER calculations.
- (d) Implement and test depletion adjoint equations for LWR nodal calculations. (This would also require modifying adjoint equations to account for detailed cross-section averaging and parameterization done in LWR analysis.)
- (e) Apply methodology to realistic fast and thermal reactor analysis..
- (f) Examine the feasibility of applying DPT to reactor optimization studies.

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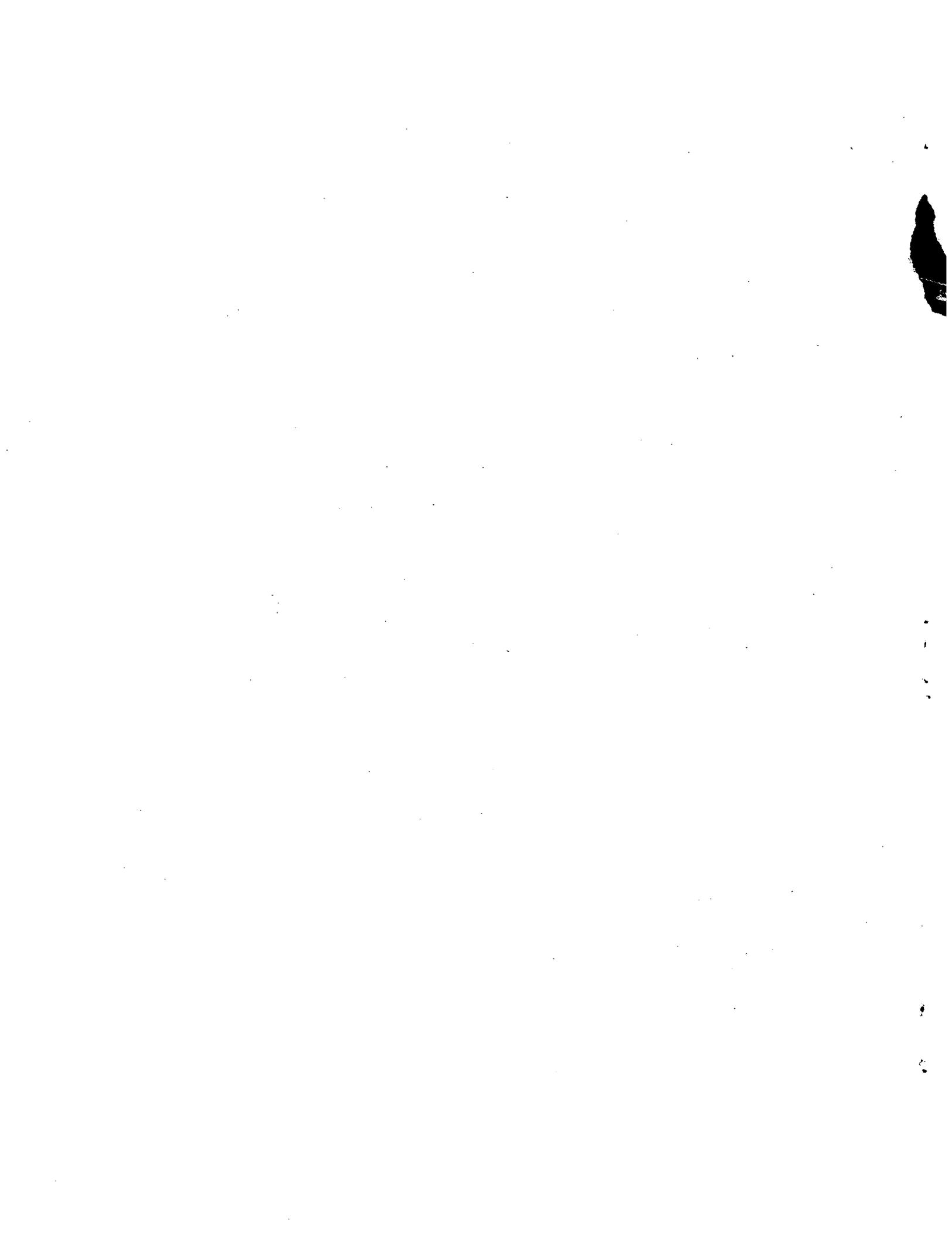
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APPENDIXES

## APPENDIX A

### MATHEMATICAL NOTATION

A.1. Vector Notation. For this study, vector fields are denoted by underlining the variable, such as  $\underline{N}(\hat{r}, t)$ . Vectors denoting points in a phase space (i.e., *independent* variables) are denoted with a caret, such as  $\hat{r} = (x, y, z)$ . Matrices are denoted with two underlines, such as  $\underline{\underline{M}}$ .

A.2. Inner Product of Vectors and Functions. All vector multiplication used in this work refers to the inner product operation:

$$\underline{A} \cdot \underline{B} = A_1 B_1 + A_2 B_2 + \dots + A_n B_n .$$

The inner product of two functions is defined analogously:

$$[g(x) \cdot f(x)]_x = \int g(x) \cdot f(x) dx .$$

A.3. Vector Derivative (gradient). The derivative of a scalar function with respect to a vector is defined by

$$\frac{\partial f}{\partial \underline{A}} (\underline{A}) = \left( \frac{\partial f}{\partial A_1}, \frac{\partial f}{\partial A_2}, \dots, \frac{\partial f}{\partial A_n} \right) \quad (\text{A-1})$$

This operation maps a scalar into a vector.

A.4. Functional Derivative (gradient). This is a generalization of the concept of a vector derivative. This operator transforms a

functional (a scalar) into a function (a vector). If  $K[f(x)]$  is a functional defined by  $K = \left[ F[f(x)] \right]_x$ , where  $F$  is a density quantity which is a *composite function of  $f(x)$* , then we have (see ref. 54 for details) for the functional derivative per unit  $x$ ,

$$\frac{\partial K}{\partial f(x)} = \frac{\partial F}{\partial f(x)} \quad (\text{A-2})$$

A.5. Functional Variation (differential). A functional variation is a generalization of the concept of a differential. It is defined by

$$\delta K[f(x)] = \left[ \frac{\partial K}{\partial f} \cdot \Delta f \right]_x = \left[ \frac{\partial F}{\partial f} \cdot \Delta f \right]_x. \quad (\text{A-3})$$

In this expression it is assumed that  $\Delta f$  is small, such that second-order terms can be ignored. A functional is stationary at some function  $f_0(x)$  if the functional gradient (and hence the variation) vanishes there. At such a point,  $K$  will either have an extremum or an inflection point (55).

A.6. Functional Taylor Series. Using the definitions in A.4 and A.5, a Taylor series expansion of a functional is defined analogously to a Taylor series for a function of a finite dimensional vector:

$$K[f + \Delta f] = K[f] + \left[ \frac{\partial K}{\partial f} \cdot \Delta f \right]_x + \frac{1}{2} \left[ \frac{\partial^2 K}{\partial f^2} \cdot \Delta f^2 \right]_{x,x'} + \dots \quad (\text{A-4})$$

## APPENDIX B

### NONLINEAR OPERATOR NOTATION

Let  $y$  be some function of the independent variables  $(x, t)$ . Also assume that  $y$  is specified by the relation

$$F(x, t, y, y_x, y_t, \dots) = 0, \quad \text{B-1}$$

where  $y_x \equiv \frac{\partial}{\partial x} y$ , etc.

and where all partial derivatives are assumed to exist.  $F$  is, in general, a nonlinear operator which maps the function  $y(x, t)$  into the zero function. In this study we deal with a special case of Eq. B-1 characterized by asymmetric time behavior:

$$F(y) = G(y) - y_t, \quad \text{B-2 (a)}$$

or

$$G(y) = \frac{\partial}{\partial t} y \quad \text{B-2 (b)}$$

where again  $G(y)$  is some operator which now is assumed to contain no time derivatives. In the case where  $G(y)$  is linear in  $y$ , Eq. B-2 can be written as

$$M \cdot y = \frac{\partial}{\partial t} y, \quad \text{B-3 (a)}$$

with

$$G(y) = M \cdot y, \quad \text{B-3 (b)}$$

where  $M$  is now a linear operator, possibly containing derivative and integral expressions. This factoring of  $G(y)$  into the product of an operator times the dependent variable is necessary in order to define an adjoint operator  $M^*$  by the relation

$$[fMg]_{x,t} = [gM^*f]_{x,t} \quad \text{B-4}$$

for arbitrary functions  $f, g$  that satisfy the necessary continuity and boundary conditions.

To define an adjoint operator for a nonlinear operator, the same criterion as in Eq. B-4 is used; therefore it is desirable to express the general nonlinear operator  $G(y)$  in a form similar to B-3 for the linear case:

$$G(y) \Rightarrow M(y) \cdot y. \quad \text{B-5}$$

The operator  $M$  is now nonlinear, and depends on  $y$ . The assumption in B-5 was made by Lewins (21) in his study of adjoint nonlinear operators; however, one must be careful about the implications of replacing a nonlinear operator by the product of another nonlinear operator times the dependent variable. In the most general case  $G(y)$  cannot be uniquely expressed in a term such as B-5. This fact can be illustrated by the simple expression  $y^2 y_x$ , which can be expressed in several ways, such as

$$(y y_x) \cdot y \Rightarrow M(y) = y y_x$$

$$\left(y^2 \frac{\partial}{\partial x}\right) \cdot y \Rightarrow M(y) = y^2 \frac{\partial}{\partial x}$$

etc.

There is obviously ambiguity in deciding which  $y$ 's are contained in the nonlinear operator and which one is to be operated on.

This presents a troublesome difficulty when trying to define an "exact adjoint operator" for  $M$ , since  $M$  is not unique. In practice the difficulty is overcome by using "first-order adjoint operators" derived from the linearized expression for  $G(y)$ . In this case there is a unique operator  $M(y)$  which operates on  $\Delta y$ . Therefore, even though an exact adjoint operator may not exist uniquely, the first-order adjoint operator will exist uniquely.

However, there is an important class of problems (into which the equations in this study fall) for which the nonlinear operator  $G(y)$  can be uniquely expressed as the product of a nonlinear operator times the dependent variable. This is the case in which the nonlinear operator only depends implicitly on the past behavior of the dependent variable through feedback mechanisms, so that at time  $t$ ,

$$G(y(t)) = M[y(t^{\leftarrow}t)] \cdot y(t)$$

B-6

Now there is no ambiguity of how to define  $M$  at any instant  $t$  because it does not explicitly contain  $y(t)$ , only past values of  $y$ . Nonlinear operators of this type appear frequently in reactor physics and account for such diverse phenomena as Doppler feedback, voiding feedback,

depletion and poison feedback, etc., which occur with a wide range of time-lag constants.

The nonlinear operators discussed in Chapter II are assumed to be of this type, and hence it is assumed that it is always possible to determine  $M(y)$ . This being the case, the "exact adjoint operator" for the nonlinear operator is defined as being analogous to Eq. B-4 for the linear case:

$$[fM(y)g]_{x,t} = [gM^*(y)f]_{x,t} \quad \text{B-7}$$

Now that the definitions of a nonlinear operator and its corresponding exact adjoint operator have been stated for the case of interest, we proceed to an examination of the effects of perturbations on nonlinear operators. This requires introducing the concept of a variation of an operator (55).

The variation (differential) of an operator  $G(y)$  in the "direction"  $\Delta y$  can be written (55)

$$\delta G(\Delta y) = \lim_{\epsilon \rightarrow 0} \frac{d}{d\epsilon} G(y + \epsilon \Delta y) \quad \text{B-8}$$

This quantity is related to the derivative of the operator by (56),

$$\delta G = \frac{dG}{dy} \Delta y \quad \text{B-9}$$

In general, the  $i^{\text{th}}$  order variation in a nonlinear operator is given by

$$\delta^i G = \lim_{\epsilon \rightarrow 0} \frac{d^i}{d\epsilon^i} G(y + \epsilon \Delta y) \quad \text{B-10}$$

Now consider an operator which is perturbed by a change in the dependent variable  $y \rightarrow y + \Delta y$ :

$$G(y) \rightarrow G(y+\Delta y) \quad \text{B-11}$$

The value for the perturbed operator can be expressed by a Taylor series expansion (55):

$$G(y+\Delta y) = \sum_{i=0}^{\infty} \frac{1}{i!} \delta^i G, \quad \text{B-10}$$

assuming that the infinite series converges. For the case in which  $G$  can be written as in Eq. B-6,

$$G(y+\Delta y) \equiv (My)' = \sum \frac{1}{i!} \delta^i (M \cdot y) \quad \text{B-11}$$

In general the  $i^{\text{th}}$  variation,  $\delta^i$ , will contain powers of  $\Delta y$  and/or its derivatives up to the  $i^{\text{th}}$  order,

$$\delta^i = \delta^i(\Delta y)$$

and hence can be viewed as a nonlinear operator in terms of  $\Delta y$ . An exact adjoint operator for  $\delta^i$  is defined by

$$[y^* \delta^i(\Delta y)]_{x,t} = [\Delta y \delta^{i*}(\Delta y) \cdot y^*]_{x,t} \quad \text{B-12}$$

For a given value of  $i$ , there may be multiple operators which satisfy the above relation. An exception to this is the case for  $i = 1$ , for which there is a unique adjoint operator that is independent of  $\Delta y$ .

Also notice that for  $i > 1$ ,  $\delta^{i*}$  is an operator in terms of  $\Delta y$ . As shown in Chapter II, this implies that it is impossible to have an exact adjoint equation for a nonlinear equation which is independent of the perturbation in the forward solution.

## APPENDIX C

### GENERALIZED ADJOINT SOLUTION FOR INFINITE HOMOGENEOUS MEDIA

The purpose of this appendix is to prove that for an infinite homogeneous medium the value for  $\Gamma^*(E)$ , which is orthogonal to the forward fission source, is given by the first term in a Neumann series expansion; i.e.,  $\Gamma^*(E)$  can be found from a fixed-source calculation without considering any multiplication. The idea for this proof was suggested to the author by R. L. Childs (57).

The equation for the shape adjoint function, as derived in the text, is given for an infinite homogeneous medium by

$$L^* \Gamma^*(E) - \lambda F^* \Gamma^*(E) = Q^*(E) \quad (C-1)$$

along with the constraint conditions

$$\int_0^{\infty} \psi(E) Q^*(E) dE = 0, \quad (C-2)$$

and

$$\int_0^{\infty} \Gamma^*(E) F \psi(E) dE = 0 \quad (C-3)$$

The forward equation for the flux shape is

$$L \psi(E) - \lambda F \psi(E) = 0 \quad (C-4)$$

The adjoint shape function can be expressed as a Neumann series by

$$\Gamma^*(E) = \Gamma_0^*(E) + \Gamma_1^*(E) + \dots, \quad (C-5)$$

where the terms in the infinite series are found from

$$L^* \Gamma_0^*(E) = Q^* \quad (C-6)$$

$$L^* \Gamma_1^*(E) = \lambda F^* \Gamma_0^*(E) \quad (C-7)$$

⋮

Multiply Eq. (C-4) by  $\Gamma_0^*$ , and Eq. (C-6) by  $\psi$ , integrate both over energy and subtract:

$$\lambda \int_0^\infty \Gamma_0^*(E) F \psi(E) dE = \int_0^\infty \psi(E) Q^*(E) dE \quad (C-8)$$

Therefore, from Eq. (C-2) we see that

$$\int_0^\infty (\Gamma_0^* F \psi) dE = 0 = \int_0^\infty \psi(E) v \Sigma_f(E) dE \cdot \int_0^\infty \chi(E') \Gamma_0^*(E') dE' \quad (C-9)$$

This equation shows that  $\Gamma_0^*(E) \perp \chi(E)$ , since

$$\int_0^\infty \chi(E') \Gamma_0^*(E') dE' = 0 \quad (C-10)$$

Now consider the term on the right-hand side of Eq. (C-7):

$$F^* \Gamma_0^* = v \Sigma_f(E) \int_0^\infty \chi(E') \Gamma_0^*(E') dE' = 0, \quad (C-11)$$

by Eq. (C-10). Since  $L^*$  is a nonsingular operator, we conclude that  $\Gamma_1^*(E) = 0$ . This argument is easily extended to the higher iterates, and the result is that

$$\Gamma^*(E) = \Gamma_0^*(E), \quad (C-12)$$

where  $\Gamma_0^*$  is the solution to Eq. (C-6).

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