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ESTIMATES OF FISSION-PRODUCT INVENTORIES IN FUEL ELEMENT E06-01 OF THE PEACH BOTTOM CORE 2: RESULTS, COMPARISONS, AND SENSITIVITY STUDY

Melvin Tobias

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ORNL-TM-4705
(UC-77 Gas-Cooled
Reactor Technology)

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Reactor Division

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OCTOBER 1974

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ESTIMATES OF FISSION-PRODUCT INVENTORIES IN FUEL ELEMENT E06-01 OF THE PEACH BOTTOM CORE 2: RESULTS, COMPARISONS, AND SENSITIVITY STUDY

Melvin Tobias

Abstract

The fission-product inventory of fuel element E06-01 of the Peach Bottom Core 2 has been estimated using the ORIGEN code to provide a theoretical basis for the present program of experimental measurements. The input numbers used, the results obtained, and a comparison with numbers provided by GAC for some of the fission-product nuclides are shown. The results of a study intended to show the sensitivity of ORIGEN calculations to the input data are also discussed.

Keywords: fission products, fuel element, Peach Bottom reactor.

I. Introduction

In support of an experimental program currently underway to measure fission-product inventories in fuel element E06-01 in Peach Bottom Core 2, a calculation has been performed using the ORIGEN[1] code. The results obtained are displayed and compared with the experimental measurements for selected nuclides and with results obtained by General Atomic Company (GAC).

The sensitivity of ORIGEN results to changes in input data is discussed with the aim of clarifying possible sources of discrepancy.

II. Calculation of E06-01 Inventories

Input Data

The irradiation history of this Peach Bottom Core 2 element was provided in a letter of January 4, 1974 by David Hoppes[2] of GAC (Table 1). The flux data comes from a simulation of the actual power history

Table 1. Flux history for fuel element E06-01 Peach Bottom core 2

(Fluxes are averages for fuel element computed by GAUGE code)

Four-group energy ranges (eV)

ϕ_1: 14.96×10^6 - 8.65×10^4

ϕ_2: 8.65×10^4 - 17.6

ϕ_3: 17.6 - 2.38

ϕ_4: 2.38 - 0.0

Core power (MW)	Time at power (days)	Mean fuel-element temperature (°C)		Fluxes at E06.01 (10^{13} neutrons/cm^2 sec)			
				ϕ_1	ϕ_2	ϕ_3	ϕ_4
107.6	5	913	(1675°F)	3.81	6.42	1.02	2.65
107.6	25	913	(1675°F)	3.86	6.50	1.03	2.69
107.6	37	913	(1675°F)	3.98	6.74	1.07	2.85
Shutdown	4.5			----	----	----	----
89.9	7.5	854	(1570°F)	3.27	5.52	0.88	2.32
Shutdown	5			----	----	----	----
111.0	69	921	(1690°F)	4.28	7.28	1.16	3.19
111.0	69	921	(1690°F)	5.02	8.56	1.36	3.85
95.5	61	860	(1580°F)	3.70	6.31	1.00	2.89
Shutdown	80			----	----	----	----
93.0	5	857	(1575°F)	3.41	5.79	0.92	2.61
93.0	25	857	(1575°F)	3.44	5.84	0.92	2.66
93.0	24	857	(1575°F)	3.46	5.88	0.92	2.70
93.0	10	857	(1575°F)	3.91	6.54	1.05	3.28
93.0	24	857	(1575°F)	3.59	6.13	0.97	2.87
93.0	24	857	(1575°F)	3.61	6.15	0.97	2.91
Shutdown	21			----	----	----	----
104.5	40	907	(1665°F)	4.88	8.12	1.31	4.26
103.2	7	904	(1660°F)	4.19	7.20	1.14	3.55
Shutdown	162			----	----	----	----

of Peach Bottom Core 2, using the 2-D diffusion code GAUGE.[3] In addition, the four-group cross sections of certain of the nuclides were made available and are shown in Table 2; these cross sections result from weighting over the Peach Bottom neutron energy spectrum, as calculated for a temperature of 1200°K (1700°F).

The input numbers THERM, RES, and FAST* were calculated to conform to the flux spectrum indicated in Table 1, and the cross sections for ^{233}U, ^{235}U, and ^{232}Th were altered to conform to the numbers in Table 2. In this way, presumably, the power generated, as calculated by ORIGEN, would be the same as the power calculated by GAC. (The inventories of many of those isotopes on a direct yield chain are strongly dependent on the power level and a correct power level is therefore essential.) The initial nuclide inventories are displayed in Table 3 and are for the whole fuel element.

III. Computed Results

Table 4 displays the results computed by ORIGEN for a selected list of nuclides. For certain of these, experimental comparisons are available.[4] The deviations between the calculated and measured activities show no particular bias. In Table 5, on the other hand, there appears to be a definite tendency for the GAC values to be lower than the ORIGEN results. Time did not permit checking the difference thoroughly. One possible explanation is that a lower power may have been used in the GAC calculations. Also, for lack of enough information, the calculations take no account of the possible effects of axial flux distributions upon results. The experimental results are normalized to the inventory of a full fuel element of 30 compacts by simple proportion.

IV. Results of Sensitivity Study

To evaluate the sensitivity of results obtained by ORIGEN to the input parameters THERM, RES and FAST, a series of cases was run with

---

*See Appendix A for definitions of THERM, RES, and FAST.

Table 2. Four-group cross sections for selected nuclides

Group	^{235}U		^{232}Th	^{233}U	
	σ_f	σ_c	σ_c	σ_f	σ_c
1	1.32	0.179	0.159	2.02	0.12
2	13.88	7.21	5.404	17.7	3.1
3	35.07	29.16	0.15	47.38	17.4
4	160.00	30.63	2.24	222.6	28.3

Group	^{238}U		^{239}Pu		^{10}B
	σ_f	σ_c	σ_f	σ_c	σ_c
1	0.127	0.129	1.67	0.1	0.86
2	---	13.3	12.02	8.34	30.15
3	---	63.8	56.59	36.21	245.6
4	---	0.95	766.3	477.4	1279.02

Table 3. Fuel element E06-01 initial inventories[7]

Nuclide	Inventory, grams
C	8550*
^{232}Th	1373.8
^{234}U	3.74
^{235}U	232.47
^{236}U	1.25
^{238}U	12.11
^{103}Rh	6.156**

*Thirty fuel compacts at 285 g C each (Table II-1, Ref. 8).

**Eighteen of the compacts contain rhodium.

Table 4. Results of ORIGEN calculation for selected fission products at end of irradiation period

	Gram atoms	Curies	Measured activity[6] (curies)	% Deviation*
^{85}Kr	7.98E-4	264		
^{89}Sr	1.71E-3	429		
^{90}Sr	1.31E-2	166		
^{95}Zr	2.58E-3	5180	5639 (30)**	-8.1
^{103}Ru	8.05E-4	2660	2128 (2)	+25.0
^{106}Ru	5.52E-4	196	229.5 (30)	-14.6
^{110m}Ag	4.35E-7	0.224		
^{111}Ag	1.42E-6	24.7		
^{125}Sb	4.87E-5	6.45	9.7 (19)	-33.3
^{127m}Te	2.49E-5	29.9		
^{129}Te	4.09E-7	1110		
^{129}I	2.39E-3	5.03E-5		
^{131}I	2.06E-4	3340		
^{134}Cs	9.23E-4	161	129.0 (30)	+24.8
^{137}Cs	1.41E-2	168	185.7 (30)	-9.5
^{140}Ba	6.81E-4	6950		
^{141}Ce	1.54E-3	6230		
^{144}Ce	6.90E-3	3180	3155 (30)	+0.8
^{152}Eu	6.47E-7	0.0193		
^{154}Eu	1.47E-4	3.3	3.07 (21)	+7.5
^{155}Eu	1.99E-5	3.94		

*% Deviation $= \left(\frac{\text{Calculated activity}}{\text{Measured activity}} - 1\right) \times 100\%$.

**Figures in this column are normalized to 30 compacts. Number of fuel compacts actually measured shown in parentheses.

Table 5. Comparison of ORIGEN and GAC inventories at end of 162-day shutdown

Isotope	GAC result (gram atoms)	ORNL result (gram atoms)	Ratio, $\frac{GAC}{ORNL}$
^{95}Mo	1.2968E-02	1.34E-02	0.97
^{99}Tc	1.2370E-02	1.36E-02	0.91
^{103}Rh	5.1693E-02	4.59E-02	1.13
^{131}Xe	5.4778E-03	5.95E-03	0.92
^{143}Nd	1.1694E-02	1.28E-02	0.91
^{133}Cs	1.3756E-02	1.41E-02	0.98
^{145}Nd	7.7666E-03	8.82E-03	0.88
^{147}Pm	1.7284E-03	2.59E-03	0.67
^{148}Pm*	2.7960E-06	3.07E-08	91.0
^{149}Sm	1.0367E-04	1.20E-04	0.86
^{150}Sm*	2.1512E-03	3.25E-04	0.66
^{151}Sm	4.7977E-04	3.14E-04	1.53
^{152}Sm	8.0980E-04	1.0E-03	0.81
^{153}Eu	4.2855E-04	7.18E-04	0.60
^{154}Eu*	6.2240E-05	1.45E-04	0.43
^{155}Eu**	7.9184E-06	1.68E-05	0.47

*Production requires neutron capture. (Other isotopes part of direct yield chain.)

**ORNL result based on 1.8 yr half-life. A more recent value is 4.65 yrs.

the Peach Bottom total inventory as a basis. Figure 1 shows a few of the fission-product chains for those nuclides for which neutron absorption may significantly alter results. Table 6 gives the cross section and decay properties as they exist in the ORIGEN library; for reference the available values from BNL-325 are displayed in parenthesis. The approximate flux level is 3×10^{13} neutrons/(cm^2 sec).

Table 7 shows the results of varying the power level, and THERM, RES, and FAST individually. The "standard" values are 112.7 MW, THERM = 0.3888, RES = 0.4027, and FAST = 0.58. (These numbers are quite different from those used in the calculation for the element E06-01. They are more in character with a large HTGR.)

The most stable quantity in Table 7 is the concentration of ^{137}Cs, which in a constant power case like this is unaffected by any of the substantial changes in input except for the power change in case N.* The inventory is then approximately one-tenth of the higher power case. The other isotopes display a far more irregular behavior with the changes. No other isotope in case N shows a one-tenth reduction.

Since all the cases in Table 7 are at constant power, a rise in any of the input parameters, THERM, RES, or FAST causes a drop in the thermal flux in the calculation and a rise in the effective cross sections. Furthermore, the production rate of leading members of fission-product chains remains unchanged at constant power. The inventory depends upon the value of $\sigma\phi$ which rises or falls depending on the individual nuclide. In the case of the FAST changes (cases L and M), the effects are rather small since the fission-product cross sections are not affected by this change. The effects are due entirely to the flux change. Much more pronounced changes occur with alterations in RES and THERM. The explanation of what is seen is highly nuclide dependent. In the case of THERM, the neutron capture rates in ^{109}Ag, ^{133}Cs and ^{153}Eu fall as THERM is reduced in case J, cutting down formation of ^{110m}Ag, ^{134}Cs, and ^{154}Eu in the 900-day time range. (None of these isotopes is at saturation levels.) In the case of ^{152}Eu, the reduction in inventory is caused by

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*^{137}Cs is primarily produced by the rapid decay of direct-fission yield isotopes.

ORNL-DWG 74-8866

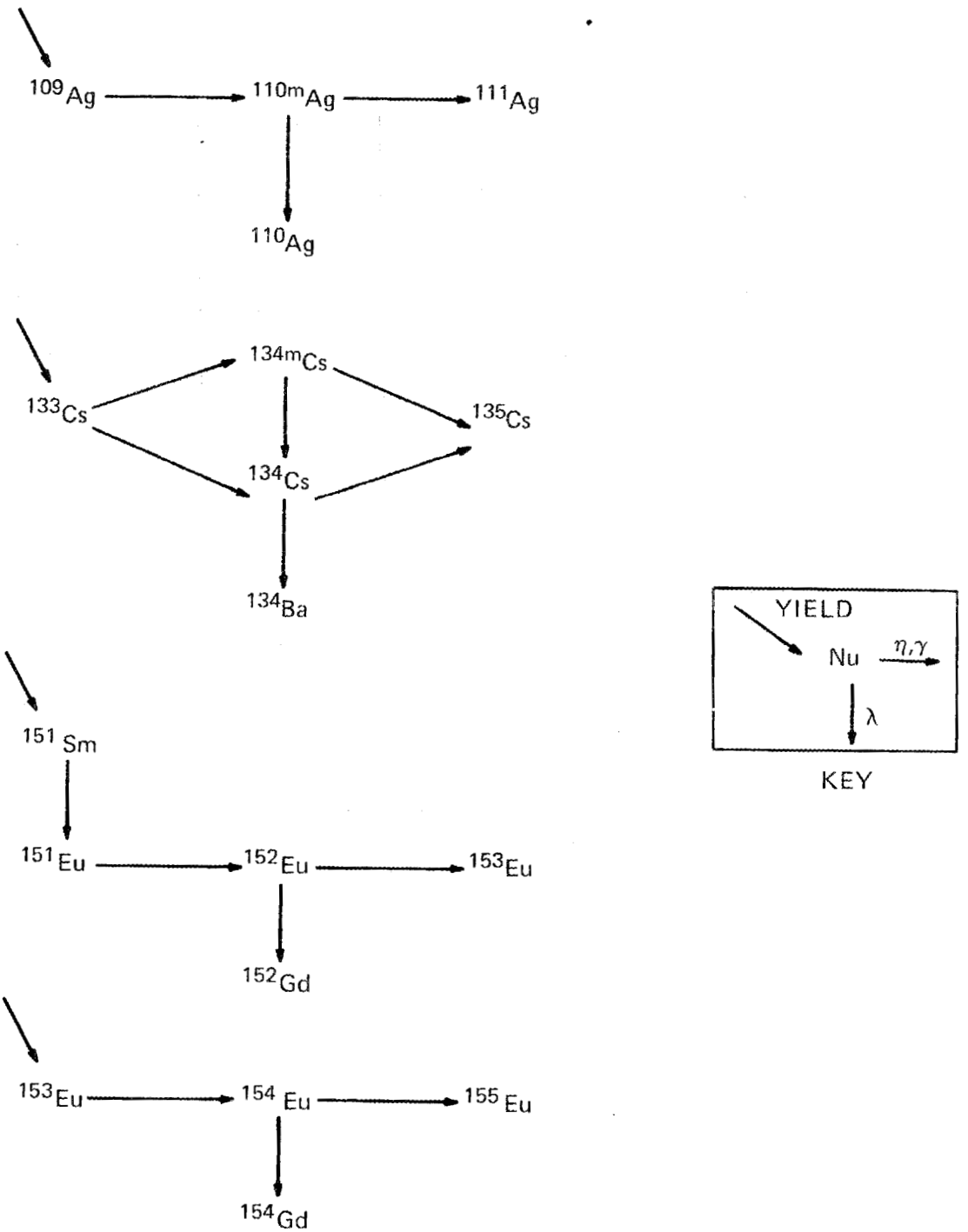

Fig. 1. Fission-product chains for ^{110m}Ag, ^{134}Cs, ^{152}Eu, and ^{154}Eu. (Neutron absorption rates may significantly alter inventories for these nuclides.)

Table 6. Cross sections of certain fission-product nuclides as listed in ORIGEN library

(Numbers in parentheses from BNL-325, 3d ed., vol. 1)

Nuclide	λ (sec^{-1})	Half-life	$\sigma_{n,\gamma}$ (2200 mps) (barns)	Resonance integral (barns)
^{110m}Ag	3.17E-08	253.0 days (252)	82.0 (82)	0.0
^{133}Cs	---	0.0	31.6 (29.0)	460.0 (415)
^{134m}Cs	6.639E-05	2.9 hr	2.6	0.0
^{134}Cs	1.071E-08	2.05 years (2.06)	134.0 (140)	66.0
^{137}Cs	7.32E-10	30 years	0.11 (0.11)	0.3
^{151}Sm	2.526E-10	87.0 years (93)	6120 (15000)	2500.0 (3300)
^{151}Eu	---	0.0	8800 (9200)	0.0 (3300)
^{152}Eu	1.8304E-09	12.0 years (4.8*)	5000 (2300)	0.0
^{153}Eu	---	0.0	390 (390)	1430.0 (1635)
^{154}Eu	1.373E-09	16.00 years (16.0)	1500 (1500)	594.0

*Probable erroneous entry.

Table 7. Sensitivity of ORIGEN results to input parameters

"Standard": Power = 112.7 MW, THERM = 0.3888,
RES = 0.4027, FAST = 0.58

Case identification	Parameter	Nuclide inventory, gram atoms				
		^{110m}Ag	^{134}Cs	^{152}Eu	^{154}Eu	^{137}Cs
O	"Standard"	2.28E-03	4.97	8.59E-04	0.941	27.3
K	THERM = 0.3478	2.44E-03	5.25	9.12E-04	0.998	27.3
J	THERM = 0.5	1.94E-03	4.36	7.54E-04	0.816	27.3
H	THERM = 0.8865	1.33E-03	3.14	5.83E-04	0.575	27.3
N	Power = 11.27	1.74E-05	4.65E-02	1.88E-04	5.96E-03	2.71
G	RES = 0.8	3.34E-03	6.71	1.29E-03	1.34	27.3
F	RES = 0.0	2.03E-04	0.562	3.48E-04	0.148	27.2
L	FAST = 0.0	2.28E-03	4.98	8.57E-04	0.943	27.3
M	FAST = 5.8	2.24E-03	4.86	8.74E-04	0.918	27.2

a rise in the neutron capture rate for that nuclide. Similar individualized discussions are required for the RES results as well.

The above cases were all at constant power. A few cases were done for the case of ^{155}Eu using a BASIC program to check the ORIGEN results. Although ^{155}Eu comes from the β decay of ^{155}Sm, which is a directly yielded fission product, about half of the ^{155}Eu inventory comes from neutron captures in ^{153}Eu and ^{154}Eu. The BASIC program took no account of this source; lack of time prevented elaborating the program to include the techniques indicated in Appendix B.

Table 8 shows the strong effect of the neglect of the ^{153}Eu and ^{154}Eu production of ^{155}Eu. Case 3, using a more up-to-date half life,[5] shows hardly any change in the final inventory, although the activity in curies would be far different, as would the concentration after a lengthy decay period. Cases 4 and 5 show the strong dependence of the inventory upon the cross section.

V. Conclusions

1. The ORIGEN libraries are in considerable need of being brought up to date, and this is being done, with the use of ENDF/B-IV data.
2. The calculations and the sensitivity study have shown the profound importance of correct input spectra.
3. The ORIGEN code should be altered to permit changes in THERM, RES, and FAST with time.
4. A simple ORIGEN library correction scheme should be implemented for all nuclides. At present, only the actinide elements can be altered easily.

The above needs concerning ORIGEN have been noted by others besides this writer. Work is currently in progress to implement these and other changes as part of the efforts of the CSEWG Fission-Product Task Force.[6]

Table 8. Sensitivity of ^{155}Eu inventory to various parameters at the end of the 543-day run of Table 1

Case No.	$T_{1/2}$ (years)	Effective σ (barns)	Program	Inventory (grams atoms)
1	1.8	7580	ORIGEN	1.59E-05
2	1.8	7580	BASIC	0.88E-05
3	4.65	7580	BASIC	0.91E-05
4	4.65	8338	BASIC	0.82E-05
5	4.65	6822	BASIC	1.00E-05
6	5.0	7580	BASIC	0.91E-05

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8. Peach Bottom Atomic Power Station, Final Hazards Summary Report, Part C, Vol. II.

APPENDIX A

Definitions of THERM, RES, and FAST

The following definitions have been taken from pages 22 and 23 of the ORIGEN report, ORNL-4628:

THERM = ratio of the neutron reaction rate for a 1/v absorber with a population of neutrons that has a Maxwell-Boltzmann distribution of energies at absolute temperature, T, to the reaction rate with 2200 m/sec neutrons

$$= \sqrt{\frac{\pi}{4}\frac{T_o}{T}}\ , \quad T_o = 293.16°K.$$

RES = ratio of the resonance flux per unit lethargy to the thermal neutron flux.

FAST = 1.45 times the ratio of the flux above 1 MeV to the thermal neutron flux.

APPENDIX B

A Method for Computing Isotope Concentrations in Fission-Product Decay Chains

Suppose a sequence of isotopes generated in chains represented by the following differential equations:

$$\frac{dN_1}{dt} = P_1 - (\lambda_1 + \sigma_1 \phi)N_1 \ ,$$

$$\frac{dN_2}{dt} = P_2 - (\lambda_2 + \sigma_2 \phi)N_2 + \sigma_1 \phi N_1 \ , \qquad (1)$$

$$\cdots\cdots\cdots\cdots$$

$$\frac{dN_j}{dt} = P_j - (\lambda_j + \sigma_j \phi)N_j + \sigma_{j-1} \phi N_{j-1} \ ,$$

where P_j = (yield of isotope j) times fission rate assumed constant over the time interval of interest.

Chains of equations like these can be dealt with in a sequential way that can greatly simplify the solution. Specifically, suppose the concentrations of the isotopes are known at the beginning of a time interval which runs from t_i to t_f. Then Eq. (1) has the solution

$$N_1(t_f)e^{K_1 t_f} - N_1(t_i)e^{K_1 t_i} = \frac{P_1[e^{K_1 t_f} - e^{K_1 t_i}]}{K_1} \ , \qquad (2)$$

where $K_1 \equiv \lambda_1 + \sigma_1 \phi$.

It will be necessary to know in what follows what the integral

$$\int_{t_i}^{t_f} e^{K_i t} N_1(t)dt \equiv I_i \qquad (3)$$

is. Integrating by parts:

$$I_i = \frac{e^{K_i t_f} N_1(t_f) - e^{K_i t_i} N_1(t_i)}{K_i} - \int \frac{e^{K_i t}}{K_i} \frac{dN_1}{dt} dt \tag{4}$$

Since $\frac{dN_1}{dt} = P_1 - K_1 N_1$ the value of I_i can be calculated, thus

$$I_i = \left\{ \frac{e^{K_i t_f} N_1(t_f) - e^{K_i t_i} N_1(t_i)}{K_i} - P_1 \frac{e^{K_i t_f} - e^{K_i t_i}}{K_i^2} \right\} \Big/ \left(1 - \frac{K_1}{K_i}\right) \tag{5}$$

Thus, the integral desired is obtainable from the end point values already obtained.

The solution of the jth equation is

$$N_j(t_f)e^{K_j t_f} - N_j(t_i)e^{K_j t_i} = P_j \frac{e^{K_j t_f} - e^{K_j t_i}}{K_j} + \int_{t_i}^{t_f} K'_{j-1} e^{K_j t} N_{j-1}(t)dt , \tag{6}$$

where

$$K'_{j-1} \equiv \sigma_{j-1}\phi.$$

The value of

$$\int_{t_i}^{t_f} K'_{j-1} e^{K_j t} N_{j-1}(t)dt$$

can be found from the value of

$$\int_{t_i}^{t_f} e^{K_j t} N_{j-2}(t)dt$$

by writing the $(j-1)$St equation, thus (with $e^{K_j t}$ multiplying both sides):

$$\frac{d(N_{j-1}\, e^{K_j t})}{dt} - N_{j-1} K_j e^{K_j t}$$

$$= P_{j-1} e^{K_j t} - K_{j-1} e^{K_j t} N_{j-1} + K'_{j-2} N_{j-2} e^{K_j t} \tag{7}$$

or, integrating from t_i to t_f

$$(K_j - K_{j-1}) \int_{t_i}^{t_f} N_{j-1} e^{K_j t}\, dt = + N_{j-1}(t)\, e^{K_j t} \Big|_{t_i}^{t_f}$$

$$- \int_{t_i}^{t_f} P_{j-1} e^{K_j t}\, dt - K'_{j-2} \int_{t_i}^{t_f} N_{j-2} e^{K_j t}\, dt \,. \tag{8}$$

Therefore, given the solution of the first equation in the sequence by formula (2), the second can be obtained from Eq. (6) with $j = 2$, and formula (5) with $i = 2$. The remaining equations can be solved by applying Eqs. (6) and (8) as needed. Care, of course, needs to be exercised to prevent numerical difficulties, that is, for large values of $K_j t$. Also, the procedure will not work as given if $K_j = K_{j-1}$, but this is only of academic interest.

The above procedure is not claimed to be original. It is displayed here for reference in the hope that interested readers will not have to re-invent it.

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