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**Analysis of Fission-Product
Effects in a Fast Mixed-Spectrum
Reactor Concept**

J. R. White
T. J. Burns

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DEPARTMENT OF ENERGY

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ANALYSIS OF FISSION-PRODUCT EFFECTS IN A FAST
MIXED-SPECTRUM REACTOR CONCEPT

J. R. White¹

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Date Published - February 1980

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ABSTRACT

The Fast Mixed-Spectrum Reactor (FMSR) concept has been proposed by BNL as a means of alleviating certain nonproliferation concerns relating to civilian nuclear power. This breeder reactor concept has been tailored to operate on natural uranium feed (after initial startup), thus eliminating the need for fuel reprocessing. The fissile material required for criticality is produced, in situ, from the fertile feed material. This process requires that large burnup and fluence levels be achievable, which, in turn, necessarily implies that large fission product inventories will exist in the reactor. It was the purpose of this study to investigate the effects of large fission product inventories and to analyze the effect of burnup on fission product nuclide distributions and effective cross sections. In addition, BNL requested that a representative 50-group fission product library be generated for use in FMSR design calculations.

Results from the brief investigation of the effects of the large fission product inventory in the FMSR indicated that care needs to be taken when modeling the effects of fission products on reactor performance. They not only have direct reactivity effects, but they also affect power and flux distributions and breeding performance. It was found that deletion of the fission products in a reference FMSR design resulted in a 5.4% reactivity gain at the beginning-of-life (equilibrium model) and about a 4.2% gain on the new equilibrium cycle, with the difference being caused by the radial redistribution of the neutron flux.

The effect of burnup on effective fission product cross sections was then analyzed with somewhat surprising results. The final conclusion was that a one-lump fission product model is sufficiently accurate for

reliable FMSR design studies. Although a multi-nuclide fission product set which explicitly accounts for the more important chains would be a more accurate representation, the added complexity and cost would outweigh the additional accuracy. It is felt that the uncertainty in the one-lump model is tolerable for preliminary design calculations and probably lies well within the uncertainty of the individual fission product cross sections.

Finally, based on the above conclusions, a 50-group fission product library was generated. The library contains a single fission product lump for each of six spectral regions of a 1-D FMSR model.

ANALYSIS OF FISSION-PRODUCT EFFECTS IN A FAST
MIXED-SPECTRUM REACTOR CONCEPT

J. R. White

T. J. Burns

1. BACKGROUND

The Fast Mixed-Spectrum Reactor¹ (FMSR) concept has been proposed by BNL as a means of alleviating certain nonproliferation concerns relating to civilian nuclear power. This breeder reactor concept has been tailored to operate on natural uranium feed (after initial startup), thus eliminating the need for fuel reprocessing. The fissile material required for criticality is produced, in situ, from the fertile feed material. This process requires that large burnup and fluence levels be achievable, which, in turn, necessarily implies that large fission product inventories will exist in the reactor. In fact, approximately 15-20% of the initially charged heavy metal to the reactor is fissioned during the fuel residence period.

In the early stages of ORNL's preliminary neutronic analysis of the FMSR,² a large discrepancy was noted in the fission product cross sections utilized at BNL relative to ORNL data. This prompted a more detailed analysis of the effects of the large fission product inventory on criticality and the power distribution of the FMSR. It also indicated the need for FMSR-specific cross-sections for the fission product nuclides, as well as for the primary heavy metal nuclides.

The initial lumped fission product data used in the early scoping calculations apparently were derived from ENDF/B-III data (although its ancestry is unverifiable). Due to this latter fact, and the availability

of ENDF/B-IV data,³ a new composite fission product was generated for use in the FMSR calculations. The details of that work are given in ref. 2.

Although the ENDF/B-IV derived fission product data was considerably more detailed and more reliable, there still remained a question as to the effect of burnup on the final lumped cross sections. A single fission product nuclide density distribution at 56,000 MWD/MTHM was used in the above study. In addition to this concern, BNL requested that they be supplied with a representative fission product library in the LIB-IV⁴ 50-group format.

The following sections discuss the relative importance of fission product data and detail the analysis performed to determine the effects of burnup on the fission product nuclide distributions and effective cross sections. Finally, the actual generation of a representative fission product library for use in BNL calculations is outlined.

2. FMSR BENCHMARK WITH AND WITHOUT FISSION PRODUCTS

Before performing a detailed analysis of the burnup effect, a brief study was initiated to determine the importance of the fission product uncertainty in the overall neutronics design of the FMSR. Most of this work is documented in ref. 2. However, a final set of calculations was performed to estimate the upper limit on the reactivity effect of the lumped fission product concentration in the reactor. This was accomplished by setting the initial fission product concentration to zero and disallowing buildup through the fission process. Tables 2.1-2.3 and Figs. 2.1-2.3 contrast the more important neutronic characteristics of the FMSR with

Table 2.1 First Cycle Comparisons of Fission Product
and No Fission Product Cases

<u>1st cycle</u>	Fission product	No fission product	% Diff
Keff			
BOC	0.979	1.032	5.41
EOC	1.003	1.054	5.08
Breeding ratio			
BOC	1.662	1.608	-3.25

Table 2.2 Equilibrium Cycle Comparison of Fission
Product and No Fission Product Cases

<u>8th cycle</u>	Fission product	No fission product	% Diff
Keff			
BOC	0.967	1.008	4.24
EOC	0.989	1.032	4.35
Breeding ratio			
BOC	1.701	1.672	-1.70

Table 2.3 Total Nuclide Reaction Rates at EOL*

Nuclide	Reference			No fission products		
	Absorption	Capture	Neutron production	Absorption	Capture	Neutron production
^{235}U	1.885-2	4.464-3	3.522-2	2.143-2	5.256-3	3.954-2
^{238}U	5.501-1	4.754-1	2.000-1	5.697-1	4.939-1	2.111-1
^{239}Pu	2.890-1	4.803-2	7.078-1	3.135-1	5.762-2	7.510-1
^{240}Pu	1.739-2	1.167-2	1.753-2	2.393-2	1.789-2	1.850-2
^{241}Pu	4.118-3	5.911-4	1.051-2	4.736-3	7.294-4	1.192-2
^{242}Pu	1.587-4	9.771-5	1.847-4	2.352-4	1.716-4	1.946-4
Be 9**	-3.708-3	-3.708-3	-	-5.407-3	-5.407-3	-
Struc	5.005-2	5.005-2	-	5.755-2	5.755-2	-
Fissp	5.915-2	5.915-2	-	-	-	-
Total	9.851-1	6.457-1	9.793-1	9.857-1	6.277-1	1.032

*Normalized to 1 neutron lost (absorption and leakage).

**Negative values reflect the inclusion of the (n,2n) reaction.

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Fig. 2.1. Radial power distribution for cases with and without fission products (see ref. 2 for model description).

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Fig. 2.2. Zone 14 ^{239}Pu concentration for reference and no fission product cases (see ref. 2 for model description).

Fig. 2.3. ^{239}Pu distribution for reference and no fission product cases (see ref. 2 for model description).

and without consideration of the equilibrium cycle fission product inventory. Table 2.1 and 2.2 give comparisons of reactivity and breeding ratio for the first and equilibrium cycles, respectively. In addition, the overall neutron balance at the beginning-of-life (BOL) for the two cases are contrasted in Table 2.3. As can be seen, the deletion of the fission products at the BOL resulted in a 5.4% reactivity gain. However, on the equilibrium cycle the reactivity increase relative to the reference calculation with fission products was only about 4.2%. The reason for this difference is that the fission product concentration was not uniform in the reference model. Therefore, elimination of the total fission product inventory resulted in a radial redistribution of the power density which, in turn, caused a redistribution of the equilibrium ^{239}Pu concentration in the reactor.

These effects are displayed graphically in Figs. 2.1-2.3. Figure 2.1 shows a decrease in the relative power production in the central regions and a large jump in power density near the fast core/moderator region interface. Since the ^{239}Pu distribution is unchanged at the BOL, this power shift is also indicative of the flux redistribution due to the absence of the fission products. Thus, there will be a decrease in the ^{239}Pu buildup rate in the center of the core and an increase in ^{239}Pu production toward the outer moderated regions.

The effect of this flux shift over several depletion cycles can be seen in Figs. 2.2 and 2.3. The first of these shows the relative ^{239}Pu concentration versus time in the first moderator region. As can be seen, the equilibrium concentration is considerably increased for the no fission product case relative to the reference calculation. Since ^{239}Pu from this region is shuffled into the fast core, an increase there should also be expected. This can be seen in Fig. 2.3 where the BOL and equilibrium ^{239}Pu distributions are given. However, this initial increase is more than offset by the decreased breeding performance of the central fast core for the no fission product case. From Fig. 2.3 one sees that about four cycles residence time in this region is required to reduce the ^{239}Pu concentration levels below that obtained in the reference calculation.

Results from this brief investigation of the effects of the large fission product inventory in the FMSR indicate that care needs to be taken when modeling the effects of fission products on reactor performance. They not only have direct reactivity effects, but they also affect power and flux distributions and breeding performance. However, it must

be remembered that the above study looked at the extreme case of a 100% decrease in the fission product concentration (or cross section). The uncertainty associated with the lumped fission product cross section utilized in the reference calculation is more likely on the order of 10-20%. Thus, although still important, the changes from the reference state would be greatly reduced from those observed in this study.

3. ANALYSIS OF FISSION PRODUCT CROSS SECTIONS VERSUS BURNUP

A. Generation of Nuclide Distributions Versus Burnup

The number densities required for the generation of the burnup-dependent lumped fission product cross sections were calculated using the ORIGEN⁵ computer code. An equilibrium cycle calculation for the "benchmark" refueling and shuffling strategy was performed using the CITATION⁶ code. The 34 cycle power history of a single batch was then input into ORIGEN, together with the appropriate initial heavy metal number densities, to determine the time-dependent distribution of individual fission product nuclides. Since ORIGEN tracks a total of 831 fission products, an edit of the 181 fission product nuclides for which multigroup data exists was performed to address the adequacy of the 181 representation. For all burnups, the 181 nuclides represent better than 99.8% of the total fission products.

It should be noted, however, that the use of the ORIGEN code does represent a degree of approximation. The ORIGEN code utilizes a zero-dimensional one-group model for the configuration in question. Four basic libraries were available; LWR, HTGR, LMFBR, and MSR. The cross sections in these libraries have been collapsed using spectra typical of

the reactors specified. The basic library utilized for this study was the LMFBR library, since it was judged to be more representative of the FMSR than the others. It must be noted, however, that an LMFBR spectrum is harder than that of the FMSR's moderated region and softer than that in the fast core region. Thus, the number densities determined via ORIGEN are subject to some spectral uncertainty. However, this uncertainty is probably of the order of the uncertainty inherent in the basic cross sections themselves.

B. Analysis of Burnup Dependent Nuclide Distributions and Effective Cross Sections

An analysis addressing the question of burnup dependent cross sections was performed employing the fission product density distributions determined in the ORIGEN calculation described above. The goals of the analysis were to determine the influence of burnup on an effective fission product cross section and to determine the best way to efficiently and accurately model this dependence.

The data available for the analysis were the density distributions versus burnup and a 126 group fission product library consisting of 181 individual isotopes. To make the analysis more manageable only the eleven discrete burnups listed in Table 3.1 were utilized. In addition, the cross sections were reduced to the 1-group level using two different spectra representative of the fast and moderated regions of the reference FMSR. Figure 3.1 displays the spectra used in the analysis and also emphasizes the need for spatially dependent cross sections for accurate FMSR studies due to the large variations in the region spectra. The next step was to calculate the 1-group macroscopic absorption cross

Fig. 3.1. Spectra used to collapse fission product cross sections.

section of the 181 individual fission products at each burnup interval for both spectra shown in Fig. 3.1. These reaction rates were then sorted in order of decreasing importance. In this manner the most important nuclides could be identified. Tables 3.2 and 3.3 list the ten most important nuclides at three different burnups in the fast and moderated regions, respectively. Also tabulated are the percent of the total reaction rate that each nuclide represents.

Several important observations can be made from the data presented in Tables 3.2 and 3.3. Most striking, of course, is the very large importance associated with SM-149 in the moderated flux region. This

Table 3.1 Discrete Burnups Used in the Present Analysis

Index	Refueling interval	Depletion time (days)	Burnup (MWD/MTHM)
1	1	185	22.7
2	4	740	113.7
3	8	1480	616.9
4	11	2035	2240
5	14	2590	10163
6	16	2960	22950
7	20	3700	55314
8	23	4255	82749
9	26	4810	111844
10	30	5550	141467
11	34	6290	170812

one nuclide accounts for approximately 45% of all neutron losses to the fission products. Just as important, however, is the fact that in the fast region, SM-149 is only of moderate importance at low burnups and accounts for less than 3% of the total absorptions at very large burnups.

Taking this comparison one step further, one sees that many of the nuclides important in the moderated region are not even included in the top ten nuclides for the fast region. For example, SM-151, XE-135, and CS-135 which contribute considerably in the moderated region are not even tabulated in Table 3.2. Another important area needing consideration is the relative importance of various nuclides as a function of burnup.

Table 3.2 Importance-Ranked Nuclides in Fast Flux Region

Burnup =						
10163 MWD/MTHM			82747 MWD/MTHM		170812 MWD/MTHM	
Index i	Nuclide	$\Sigma a_i / \Sigma a_t$ (%)	Nuclide	$\Sigma a_i / \Sigma a_t$ (%)	Nuclide	$\Sigma a_i / \Sigma a_t$ (%)
1	RH-103	9.88	RH-103	10.95	RH-103	11.77
2	RU-101	8.85	PD-105	9.20	PD-105	8.96
3	PE-105	8.78	RJ-101	8.05	RU-101	7.47
4	CS-133	7.01	CS-133	6.77	CS-133	6.63
5	TC-99	6.27	TC-99	5.87	TC-99	5.72
6	SM-149	4.33	SM-149	3.74	RU-102	3.80
7	PM-147	3.92	PD-107	3.68	PD-107	3.77
8	MC-97	3.56	RU-102	3.59	MO-97	3.46
9	RU-102	3.38	MD-97	3.30	MO-95	3.22
10	NE-143	<u>2.90</u>	MD-95	<u>2.87</u>	ND-143	<u>2.94</u>
Total		58.88				57.74

Table 3.3 Importance-Ranked Nuclides in Moderated Flux Region

Burnup =		10163 MWD/MTHM	82747 MWD/MTHM		170812 MWD/MTHM	
Index i	Nuclide	$\Sigma a_i / \Sigma a_t$ (%)	Nuclide	$\Sigma a_i / \Sigma a_t$ (%)	Nuclide	$\Sigma a_i / \Sigma a_t$ (%)
1	SM-149	47.67	SM-149	45.75	SM-149	45.88
2	SM-151	8.81	SM-151	8.96	SM-151	8.89
3	XE-135	4.80	CS-135	3.64	CS-135	3.98
4	PM-147	3.66	XE-135	2.85	ND-143	2.85
5	CS-135	3.25	ND-143	2.74	PD-105	2.77
6	ND-143	2.72	PD-105	2.63	TC-99	2.34
7	PD-105	2.26	PM-147	2.57	RH-103	2.14
8	TC-99	2.13	TC-99	2.22	RU-101	2.12
9	RU-101	2.08	EU-155	2.16	PD-107	2.07
10	EU-155	<u>1.71</u>	RU-101	<u>2.11</u>	EU-155	<u>2.07</u>
Total		79.09		75.63		75.11

For example, in the moderated region XE-135 accounts for 4.8% of the total absorption at low burnups, 2.8% at burnups around 80,000 MWD/MTHM, and only about 1.0% at 170,000 MWD/MTHM. The reason for this is that the XE-135 concentration reaches an equilibrium level depending on the flux level relatively quickly; but other isotopes, like CS-135, are non-saturating and, therefore, their concentrations increase approximately linearly with burnup.

A single lumped fission product pair absorption cross section versus burnup was then calculated using:

$$\overline{\sigma}_{ab} = \frac{\sum_i (N_i \sigma_{ai})_b}{\sum_i (N_i)_b} * 2.0 ,$$

where the subscript b refers to a specific burnup. Results of this calculation using both the fast and moderated spectra are displayed in Fig. 3.2 and 3.3, respectively. The effective lumped cross section is essentially constant with burnup in the fast regions of the FMSR. The moderated region's cross section, however, shows a fairly linear decrease with burnup. The slope of this line is quite small and gives a 13% decrease in the lumped fission product cross section over the range of about 40,000 to 170,000 MWD/MTHM.

This constant relationship of the cross sections with burnup is emphasized in Figs. 3.4 and 3.5. The macroscopic effective cross section and total fission product concentration is plotted versus burnup. The macroscopic cross section is given by:

$$\Sigma_{ab} = N_{Tb} \overline{\sigma}_{ab}$$

where

$$N_{Tb} = \sum_i (N_i)_b .$$

Fig. 3.2. 1-group absorption cross section versus burnup for fast region spectra.

Fig. 3.3. 1-group absorption cross section versus burnup for moderated region spectra.

Fig. 3.4. Lumped fission product concentration and absorption rate versus burnup.

Fig. 3.5. Lumped fission product concentration and absorption rate versus burnup plotted on a log-log scale.

Figs. 3.4 and 3.5 display the same data in slightly different form. The key point to note is the near linear relationship with burnup of both the fast and moderated region effective cross sections.

At this point in the study it was suggested that two lumped cross sections be utilized, one representing the more important odd atomic numbered isotopes and one representing the even isotopes. In this way one might be able to account for the neutron capture of an odd isotope going to an even isotope. This might model the effect observed in Fig. 3.3.

This suggestion was investigated further in the following manner. First a plot of the fission product distribution as a function of the atomic number was drawn for different burnups. Fig. 3.6 shows one such plot. A shift in the distribution to higher atomic number with burnup is clearly apparent. The concentrations for the even and odd isotopes were then summed separately and plotted as a function of burnup in Fig. 3.7. At zero burnup the normalized density of both the even and odd isotopes were nearly unity. The normalized even isotope density increased linearly with burnup with about a 5% increase over the 170,000 MWD/MTHM burn. Obviously, a corresponding decrease in the odd atomic number nuclides was observed. This odd-even transmutation effect, therefore, has a maximum effect of only 3-4% on the macroscopic absorption rate. In addition, if the reference burnup for a 1-lump model was taken in the center of the burn, there would be only a $\pm 2\%$ variation from the reference.

Individual odd and even microscopic lumped cross sections also were generated so that their behavior with burnup could be investigated. Figures 3.8 and 3.9 display the results of this exercise. The conclusion

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Fig. 3.6. Fission product density versus atomic number as a function of burnup.

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Fig. 3.7. Even and odd lumped fission product densities versus burnup.

Fig. 3.8. Even and odd lumped absorption cross sections versus burnup for fast region spectra.

Fig. 3.9. Even and odd lumped absorption cross sections versus burnup for moderated region spectra.

is that since the even isotope's contribution to the total microscopic absorption cross section is small, the one lumped cross section essentially has the same shape as the odd isotope cross section. Thus, the variation of the nuclide distribution within the odd lump is the major cause of the variation of the one lumped cross section with burnup.

This observation is more apparent in Figs. 3.10 and 3.11. Through examination of the important nuclide concentrations it was determined that, in the moderated region at least, ^{135}Xe , ^{147}Pm , and ^{149}Sm were the key isotopes. These three isotopes' variation with burnup contributed to much of the cross section variation observed in Fig. 3.3. Therefore, an analysis was performed on a partial lumped fission product that excluded the nuclides in the ^{135}Xe , ^{147}Pm , and ^{149}Sm chains. The chains excluded were:

and

Therefore, there was a total of 13 fission product nuclides, 3 from the Xe chain, 9 from the Pm and Sm chain, and 1 partial lumped fission product. It is the variation of this partial lumped cross section with burnup that is displayed in Figs. 3.10 and 3.11. After an initial transient period, the partial lumped cross section is relatively constant with burnup in both fast and moderated flux spectra. Thus, this indicates that it is the variation of the Xe, Pm, and Sm chains that account for most of the variation of the one lumped fission product pair cross section (in the moderated region).

Fig. 3.10. Partial lumped fission product cross section versus burnup for fast region spectra.

Fig. 3.11. Partial lumped fission product cross section versus burnup for moderated region spectra.

C. Conclusions

The results just discussed were somewhat surprising, especially Fig. 3.5. The fact that the macroscopic cross section curves almost exactly parallel the fission product concentration line indicates that a one lumped fission product pair cross section is a very good model of the 181 individual nuclide cross sections. The slight deviation observed at low burnups is not too disconcerting since the density, and therefore macroscopic cross section, is small in the low burnup regions. Thus, uncertainties at low burnups have little effect on the overall neutronic performance of the reactor.

The final conclusion of this study is that the one lump fission product model is sufficiently accurate for reliable FMSR design studies. Although a multi-nuclide fission product set which explicitly accounted for the ^{135}Xe , ^{147}Pm , and ^{149}Sm chains would be a more accurate representation, the added complexity and cost would outweigh the value of this additional accuracy. The multi-nuclide representation may be more applicable for detailed startup, shutdown, and other short-term transient analyses. It is felt that the uncertainty in the one-lump model is tolerable for initial design calculations and probably lies well within the uncertainty of the individual fission product cross sections.

4. GENERATION OF LUMPED FISSION PRODUCT CROSS SECTIONS

The conclusion from the above analysis was that a one-lump fission product model was sufficiently accurate for initial FMSR design calculations. However, it also pointed out that large variations in regional spectra are inherent in the FMSR design. Due to this fact, six

50 group lumped fission product sets were generated, each representing a different geometrical region of a 1-dimensional FMSR model.

Figure 4.1 is a flow chart of the scheme used to obtain a six nuclide 50 group fission product library. Table 4.1 gives the burnup by region at which the collapse was performed. Increasing region number is in the direction of increasing radius. A burnup of approximately 83,000 MWD/MTHM was chosen for the fast region because this was near the mid-point of the expected burn. However, the moderated regions of the reference FMSR configuration never see high burnup fuel. Therefore, a nuclide distribution at about 10,000 MWD/MTHM was utilized for the two moderated region lumped fission products.

The collapse from the 126 group format to a 47 group library was done in the standard way utilizing the XSDRNPM⁷ module of the AMPX⁷ modular code system. The collapse to a 47 group level was first performed because of certain energy group boundary requirements for the 50 group library format specified by BNL. The 50 group boundaries were not a subset of the available 126 group fission product library. In most cases, the boundaries were close enough as not to cause any significant errors. However, in three distinct cases the 50 group library had a finer group structure than the 126 fine-group library. For example, group 34 and 35 in the 50 group set were contained in group 111 of the 126 group library. To overcome this difficulty, the collapse in XSDRNPM was performed to a 47 group format, essentially ignoring the finer group structure in the 50 group library for the three cases of interest. The 47 group library was then extended to the 50 group format specified by BNL by dividing groups 34, 35, and 42 into two equal lethargy group widths. This was performed in a small FORTRAN code written specifically

Fig. 4.1. Flow chart for generation of 50-group lumped fission product cross sections.

Table 4.1 Burnup and Spectral Region
by FMSR Zone

FMSR zone	Burnup (MWD/MTHM)	Spectral region
1	82749	central fast core
2	82749	fast core
3	82749	fast core
4	82749	outer fast core
5	10163	inner moderated region
6	10163	outer moderated region

for this purpose, with special consideration given to handling the scattering matrix in a consistent manner.

The fission product cross sections generated utilizing the above scheme are plotted in Figs. 4.2 and 4.3, displaying the microscopic absorption and total cross sections respectively. The first zone in the fast and moderated regions were chosen as representative of the whole region. In fact, the 50-group representation is sufficiently detailed that the differences within a region (fast or moderated) are negligible. Thus, at the 50-group level only two lumped fission products are really needed rather than six lumps. Also, as observed from Figs. 4.2 and 4.3, differences between the fast and moderated regions are relatively small at low energies and completely negligible at high energies. The variation at the low end of the spectrum is mainly due to the different burnup/nuclide distribution employed in the collapse to one lumped fission product.

It should be emphasized that Figs. 4.2 and 4.3 confirm the results obtained from the 1-group analysis discussed previously. From these plots one can conclude that at high energies the fission product microscopic absorption and total cross section is constant with burnup. At low energies, however, the microscopic cross sections are weak functions of burnup with a maximum variation of about 10% over the burnup interval of 10,000 to 80,000 MWD/MTHM. These observations support the conclusion that a one-lump fission product representation is sufficient for FMSR design analysis.

Finally, it should be noted that due to time limitations the final fission product cross sections generated in this study have not been utilized at ORNL. However, no problems with use of the fission product library is anticipated.

Fig. 4.2. 50-group fission product pair microscopic absorption cross section.

Fig. 4.3. 50-group fission product pair microscopic total cross section.

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