Fission-product Burnup Chain Model for Research Reactor Application

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Abstract

A new fission–product burnup chain model was developed for use in research reactor analysis capable of predicting the burnup–dependent reactivity with high precision over a wide range of burnup. The new model consists of 63 nuclides treated explicitly and one fissile–independent pseudo–element. The effective absorption cross sections for the pseudo–element and the pseudo–element yield of actinide nuclides were evaluated in the this report. The model is capable of predicting the high burnup behavior of low–enriched uranium–fueled research reactors.

요   약

고연소에서도 연소 과정을 정확히 계산할 수 있는 연구로용 핵분열 생성물 연소 체인 모델을 개발하였다. 이 모델은 상세히 취급된 63개 핵분열 생성물 핵종과 핵분열성 물질에 무관한 하나의 가상 원소로 구성되어 있다. 본 논문에서는 가상 원소의 유효 단면적과 악티나이드 핵종별 핵분열 생성율이 평가 되었다. 이 모델은 저농축 우라늄을 핵연료로 하는 연구로의 고연소 해석에서 좋은 결과를 보이고 있다.

1. Introduction

The time–dependent behavior of neutron absorption in fission–products is a major concern in any reactor depletion calculations. The degree of complexity required for an adequate treatment of fission-product buildup depends on several factors, including the neutron energy spectrum, fuel isotope composition, burnup range, etc. .To estimate the reactor's performance as a function of time, it is necessary to model the fuel depletion and the buildup of fission–products. An exact calculation of full fission–product buildup is not feasible since there are several hundred fission–product nuclides, and lattice group constants for core calculation would have to be generated at numer-

ous burnup points. Since the computer time required for a depletion problem depends on the number of nuclides being treated explicitly, simplified many kinds of fission–product burnup chain models have been developed and applied to burnup calculations for various types of reactors.

The Garrison–Roos model[1] consists of three lumps, saturating , slowly saturating and nonsaturating fission–products plus two isotopes of Xe^{135} and Sm^{149}. In the Nephew model[2], X^{135} is always treated explicitly. In addition, six other fission–products with high cross sections, namely Sm^{149}, Sm^{151}, Cd^{113}, Eu^{155}, Gd^{155} and Gd^{157} are collectively referred to as "samarium", and 30 other fission–products are assigned to four groups according to their excess resonance integral. These simple models are applicable to a low burnup and/or a survey calculation for reactor design.

With the development of large and fast computers, it has become possible to treat many nuclides explicitly. A number of different models, which treat $20 \sim 40$ explicit nuclides and one or two pseudo-elements, have been developed and successfully applied to commercial power reactor calculations. For example, the EPRI–CPM code[3] uses a model containing 22 explicit nuclides and two pseudo–elements, and the original WIMS code[4] uses 32 explicit nuclides and one pseudo–element. The models recommended by Iijima et al[5]. and Donnelly[6], use 45 explicit nuclides and one pseudo–element. On the other hand, more than 50 explicit nuclides have been used occasionally for special reactor design calculations such as advanced converter[7], $HTGR^{8)}$, $HCLWR^{9)}$ etc. In many cases, the models developed for power reactor design have been used for research reactor problems. However, practical questions arise when we try to apply the models developed for power reactor to research reactor calculations with high burnup.

Since both the fuel composition and the discharge fuel burnup are very different from any of

the thermal power reactors for which the burnup model was developed, a study on the fission–product burnup chain model for a research reactor with high burnup was initiated. The purpose of this work is to develop an efficient fission–product burnup chain model for research reactor lattice calculations with the aim of predicting the burnup-dependent reactivity with high precision over a wide range of burnup.

2. Importance of Fission–Products in Burnup Calculation

As discussed above, because of a lack of the efficient fission–product burnup model available for research reactor the conventional model for power reactor has been used as a tool for the burnup calculations of research reactors. The characteristics of a current low–enriched, uranium –fueled research reactor are considerably different from those of thermal power reactors. Therefore, a study was performed to investigate the adequacy of a conventional fission–product model for the analysis of the research reactor problems with high burnup. A survey calculation using a pin–cell geometry of the KMRR fuel[10] which consists of 20% uranium enriched U_3Si–Al was performed with the WIMS–KAERI code[11] and the associated WIMKAL–88 library[12]. The model in the library, which is the same as the 1986 version[13] of WIMS library, treats 35 nuclides explicitly together with a single pseudo–element, and the pseudo data are quoted from the old WIMS library[14] without adjustment.

The result of this calculation is given in Fig. 1. The figure shows the burnup–dependent contribution of main fuel isotopes and total fission–product absorption. The data on minor actinides are not included in this figure. The contribution of total fission–product is on the increase with burnup, reaching nearly 15% of the total absorption at the burnup of 122 GWD/MTU, and 25% at
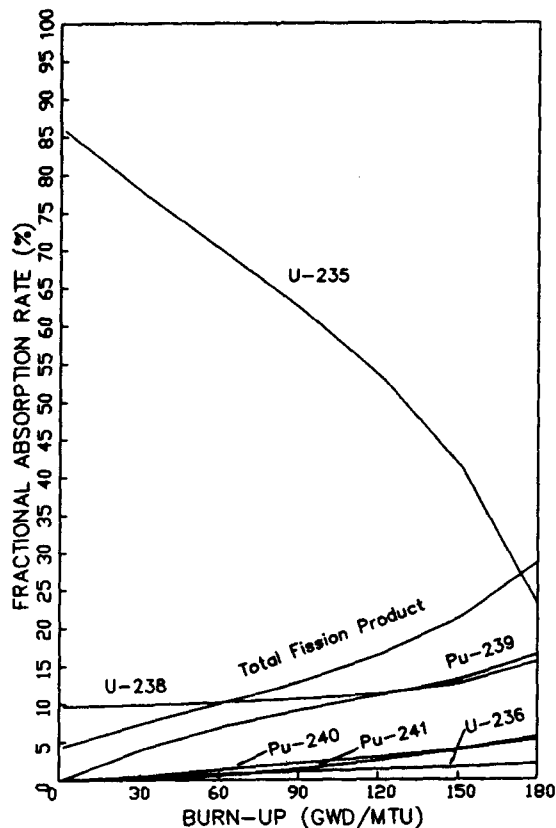
Fig. 1. Contribution of Actinides and Fission—products to Total Neutron Absorption Rate in KMRR Fuel

182 GWD/MTU. Hence, it is expected that the fission—products are very important contributor to the reactivity change with burnup. The contribution of the pseudo—element to total fission—product absorption are estimated to 2.0% at 32 GWD /MTU, 4.8% at 122 GWD/MTU and 7.1% at 182 GWD/MTU. The contribution of pseudo—element absorption at high burnup domain is very high. The fission—product chain must be described in sufficient detail to be adequate for all burnup ranges being considered. This usually requires the chain description to be more complex than would be needed when studying a low burnup characteristics.

To identify the important poisoning fission—product nuclides in research reactor depletion, a

more detailed burnup calculation using a full fission—product representation was performed with the ORIGEN2 code[15]. The code can explicitly calculate the absorption rates over 850 fission —product nuclides produced during irradiation. The ORIGEN2 code is provided with a large number of one—group cross section data base for different types of power reactors but there is no special data base for thermal research reactors. Therefore, the associated library for PWR with uranium cycle and burnup dependence of cross sections to 50,000 MWD/MTU was used for calculating the burnup characteristics of the KMRR fuel.

The calculated result showed that the absorption contribution by 60 and 70 important fission —product nuclides were over 99.0% and 99.5% of total fission—product absorption, respectively. And it was most interesting that 100 important nuclides of the 226 total fission—products considered in the ORIGEN2 calculation account for more than 99.9% of the total fission—product poisoning in the burnup range up to approximately 120 GWD/MTU. As a result of this calculation, it was found that a fission—product chain model treating 100 important fission—products explicitly would be able to predict the high burnup poison effect of a low—enriched uranium research reactor fuel without the pseudo concept.

3. Reference Model

To develop a simplified fission-product burnup model, a lumped pseudo—element could be used with its cross section and fission yield values normalized to more exact explicit description. For this, 105 fission—product nuclides were selected on the base of previous analysis. Besides the important fission—product absorbers, some precursors of strong absorbers in either decay or capture chain were included in the 105 nuclides. Using all the 105 nuclides, a reference chain model was

constructed. The 105 fission–products model was taken as the reference to which simpler treatment was normalized. Table 1 lists the individual nuclides included in the reference chain model.

Table 1. Nuclides included in the Reference Model

35-Br- 81	46-Pd-106	59-Pr- 141
36-Kr- 82	46-Pd-107	59-Pr- 143
36-Kr- 83	46-Pd-108	60-Nd-142
36-Kr- 84	47-Ag-109	60-Nd-143
36-Kr- 85	48-Cd-110	60-Nd-144
37-Rb- 85	48-Cd-111	60-Nd-145
37-Rb- 87	48-Cd-112	60-Nd-146
38-Sr- 89	48-Cd-113	60-Nd-147
38-Sr- 90	49-In- 115	60-Nd-148
39-Y - 89	51-Sb-121	60-Nd-150
39-Y - 91	51-Sb- 123	61-Pm-147
40-Zr- 91	53-I -127	71-Pm-148
40-Zr- 92	53-I -129	61-Pm-148
40-Zr- 93	53-I -131	61-Pm-149
40-Zr- 94	53-I -135	61-Pm-151
40-Zr- 95	54-Xe-131	62-Sm-147
40-Zr- 96	54-Xe-132	62-Sm-148
41-Nb- 95	54-Xe-133	62-Sm-149
42-Mo- 95	54-Xe-134	62-Sm-150
42-Mo- 96	54-Xe-135	62-Sm-151
42-Mo- 97	54-Xe-136	62-Sm-152
42-Mo- 98	55-Cs-133	62-Sm-153
42-Mo- 99	55-Cs- 134	62-Sm-154
42-Mo-100	55-Cs- 135	63-Eu- 153
43-Tc- 99	55-Cs- 137	63-Eu- 154
44-Ru-100	56-Ba-134	63-Eu- 155
44-Ru-101	56-Ba- 138	63-Eu- 156
44-Ru-102	56-Ba- 140	64-Gd-154
44-Ru-103	57-La- 139	64-Gd-155
44-Ru-104	57-La- 140	64-Gd-156
44-Ru-105	58-Ce-140	64-Gd-157
45-Ru-103	85-Ce-141	64-Gd-158
45-Rh-105	58-Ce-142	65-Tb- 159
46-Pd-104	58-Ce-143	66-Dy-161
46-Pd-105	58-Ce-144	66-Dy-162

69–group absorption cross sections of the 105 nuclides at 600°K were prepared from ENDF/B–V data[16] using the processing code NJOY[17], and the burnup related information such as chain scheme, fission yield data, β–decay constants, power per fission etc. are compiled in the library. All the data were added to the WIMKAL–88 library. More detailed procedure for generating the multigroup data can be found in Ref. 12. It can be expected that this reference model would account for over 99.9% of a full fission–product poisoning. With these data, a burnup calculation for a 20% enriched U₃Si–Al fuel was performed using the WIMS–KAERI code up to approximately 200 GWD/MTU. The total fission–product absorption rates calculated at various burnup steps have been used as reference data in the derivation of a simpler chain model.

4. New Fission–product Chain Model

From the study based on the above result a new chain model containing 63 explicit nuclides and one lumped pseudo–element was constructed as shown in Fig. 2. In this model, 63 nuclides of the 105 fission–products are explicitly treated during burnup, and the remaining 42 nuclides are lumped together as a single pseudo–element.

In thermal reactor calculation, it has been rather customary to use the pseudo–element cross sections not depending on the irradiation time and/ or the fissioning nuclides. The fissile dependence is taken into account by the effective pseudo yields. Therefore, those characteristics were incorporated in the construction of pseudo–element data, that is, a stable nuclide that would be destroyed by capture, fissile–independent cross sections, and a distinct fission yield for each fissioning nuclide.

In principle, there are many possible approaches to preparing the multigroup cross sections of a lumped pseudo fission–product. The
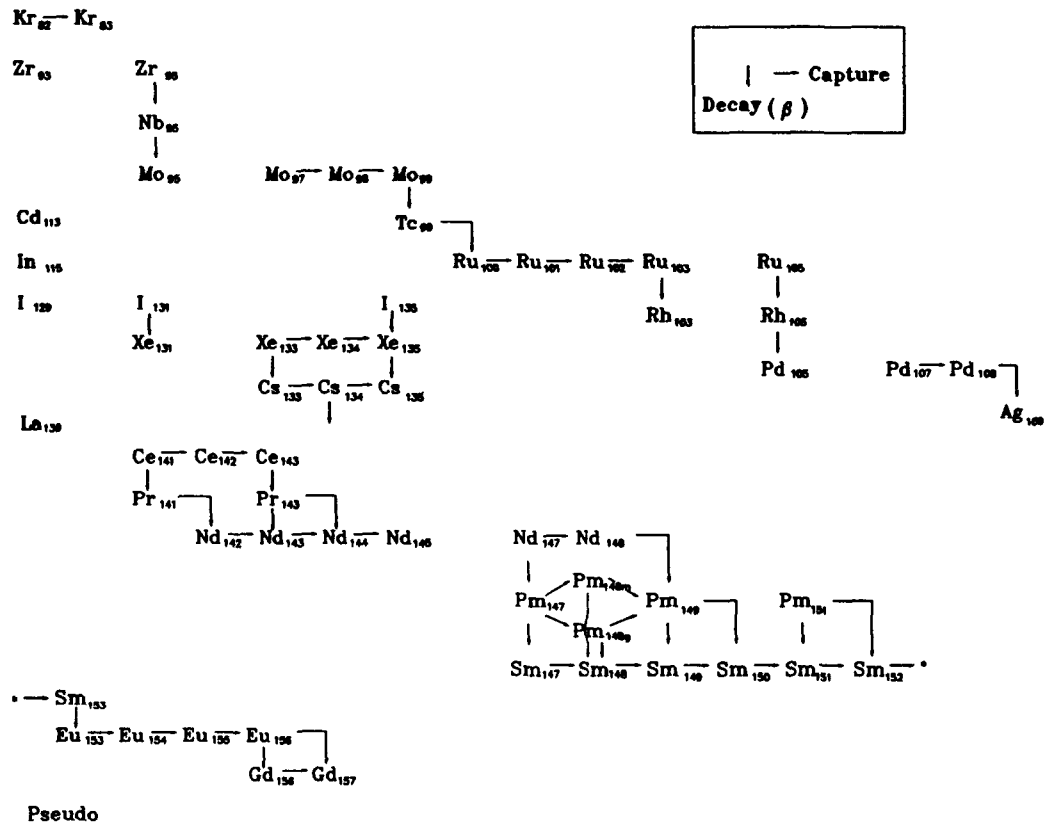
Kr$_{82}$— Kr$_{83}$

Zr$_{83}$ Zr$_{95}$

| — Capture
Decay (β)

Nb$_{95}$

Mo$_{95}$ Mo$_{97}$— Mo$_{98}$— Mo$_{99}$

Cd$_{113}$

Tc$_{99}$

In$_{115}$

Ru$_{100}$— Ru$_{101}$— Ru$_{102}$— Ru$_{103}$ Ru$_{104}$

I$_{129}$ I$_{131}$ I$_{135}$

Rh$_{103}$ Rh$_{104}$

Xe$_{131}$ Xe$_{133}$— Xe$_{134}$— Xe$_{135}$

Pd$_{104}$ Pd$_{107}$— Pd$_{108}$

Cs$_{133}$— Cs$_{134}$— Cs$_{135}$

Ag$_{109}$

La$_{139}$

Ce$_{141}$— Ce$_{142}$— Ce$_{143}$

Pr$_{141}$ Pr$_{143}$

Nd$_{142}$— Nd$_{143}$— Nd$_{144}$— Nd$_{145}$ Nd$_{147}$— Nd$_{148}$

Pm$_{147m}$

Pm$_{147}$ Pm$_{149}$ Pm$_{151}$

Pm$_{148g}$

Sm$_{147}$— Sm$_{148}$— Sm$_{149}$— Sm$_{150}$— Sm$_{151}$— Sm$_{152}$ •

• — Sm$_{153}$

Eu$_{153}$— Eu$_{154}$— Eu$_{155}$— Eu$_{156}$

Gd$_{156}$— Gd$_{157}$

Pseudo

Fig. 2. Fission–product Chain for 63 Explicit plus One Pseudo Model

ission yield fractions for individual nuclides or the ission–product concentrations at a burnup step have been mainly used as weighting functions in the derivation of the effective multigroup cross sections for a lumped pseudo–element. Since research reactor fuel generally achieves high burup, it is expected that an application of the conentration weighting is desirable.

An effective cross section, σ_a, for the lumped uclide can be obtained by weighting the cross ections, σ_a^i for the individual nuclide by the oncentration, N_i, at a burnup step :

$$\bar{\sigma}_a = \frac{\sum_i N_i \sigma_a^i}{\sum_i N_i} \qquad (1)$$

where the summation is over all nuclides included i the lumped pseudo–element. The effective ross sections for the pseudo–element were evalu-

ated by applying Eq.(1) to each group in 69–group of the WIMS code. And a preliminary pseudo yield fractions of each actinide nuclide was selected by using Eq.(2). That is, if y_i is the yield for nuclide i, then the yield fraction, Y, of pseudo–element is

$$Y = 2 - \sum_i y_i \qquad (2)$$

where the summation is over all nuclides treated explicitly in the model.

Considering the burnup dependence, three kinds of temporary cross section sets for the pseudo–element were prepared using the concentrations at each burnup step of 62, 122 and 182 GWD/MTU as weighting function. Then preliminary test calculations were performed using these sets. Burnup dependence of k_∞–values were com-
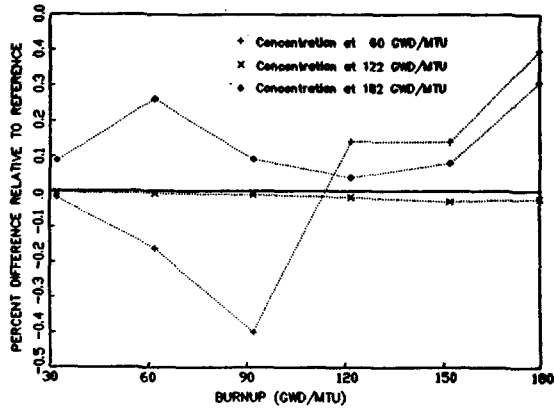
Fig. 3. Effects of Burnup–dependent Concentration Weighting for Generating Lumped Pseudo–element

pared with the reference data in Fig.3. Since the dependence of lumped cross sections on burnup is not significant, it is clear that one can arbitrarily lump the fission–products at any given burnup step and apply the data to all other burnup steps. So the cross section set at burnup of 122 GWD/ MTU was fixed as an effective one. And then, the preliminary pseudo yields were adjusted to conserve the absorption rates. The effective 69–group absorption cross sections of the pseudo–element and the associated pseudo yield data estimated from the above processes are given in Fig.4 and Table 2, respectively.
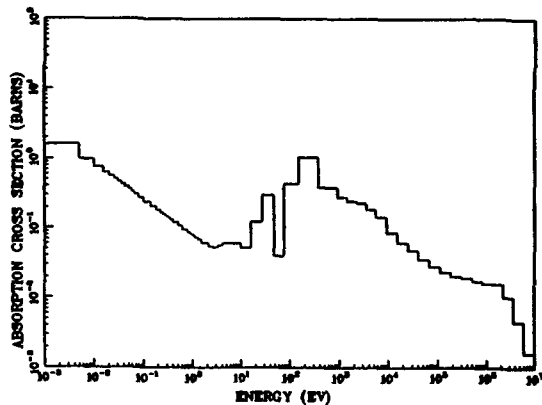

Fig. 4. Absorption Cross Section of Pseudo–element in New Model

Table 2. Recommended Pseudo–element Yield Data for Actinides

Nuclide	Fission Yield	Nuclide	Fission Yield
U–235	7.60768E–01*	Pu–239	6.59652E–01
U–236	7.32749E–01	Pu–240	6.51088E–01
U–238	6.56068E–01	Pu–241	6.40982E–01
Np–237	6.97372E–01	Pu–242	6.45311E–01

* Read as 7.60768×10^{-1}

5. Verification of New Model

The new model developed in the present work was verified by performing a cell burnup calculation for a research reactor fuel using the WIMS –KAERI code. The burnup dependence of total fission–product absorption rate and k_∞ are compared with the reference values. The results are summarized in Table 3 and 4, respectively.

The absorption differences of the WIMKAL–88 are getting worse with the increase in the depletion time. However, the trend of the new model shows a stable result. Percent differences of total fission–product absorption relative to reference data are within 0.02 for 63 explicit model. The k_∞ –values show that the differences of the WIMKAL–88 are also on the increase with burnup. The WIMKAL–88 model overpredict the k_∞ by as much as 0.1% at low burnup and 2% at high burnup. On the other hand the new model predicts the behavior of k_∞ within the difference of 0.01% Δk to the reference up to 182 GWD/ MTU. And the contribution of pseudo–element to total fission–product absorption, as shown in Table 5, are decreased within 1.0% in the burnup range considered. Compared to 7.1% difference of the old model, the new model shows a very stable result.

6. Conclusion

An efficient high precision fission–product bur-

Table 3. Comparison of Fission–product Models on Total Poison Fraction of All the Fission–products as a Function of Burnup

Burnup (GWD/MTU)	Fission–product Model		
	Reference	WIMKAL–88 Library	New Model
32	6.87491E–02*	0.150**	–0.001
62	9.43018E–02	–2.289	–0.002
92	1.21650E–01	–3.943	–0.005
122	1.53289E–01	–5.287	0.005
152	1.92844E–01	–6.665	0.023
182	2.43102E–01	–8.658	0.012

* Volume integrated absorption rate (sec^{-1}), Read as 6.87491×10^{-2}

**Percent difference relative to the reference

Table 4. Comparison of Fission–product Models on K–infinite Values as a Function of Burnup

Burnup (GWD/MTU)	Reference (K)	WIMKAL–88 Library	New Model
32	1.509906	–0.103*	0.000
62	1.419802	0.077	0.000
92	1.316655	0.310	0.000
122	1.188645	0.638	–0.002
152	1.006706	1.163	–0.009
182	0.726918	2.085	–0.005

*Percent difference relative to the Reference

Table 5. Fractional Absorption Rates of Pseudo–element to Total Fission –product Absorption(%)

Burnup (GWD/MTU)	Fission–product Model	
	WIMKAL–88 Library	New Model
32	2.01	0.35
62	3.11	0.52
92	3.97	0.64
122	4.78	0.74
152	5.72	0.84
182	7.11	0.98

nup chain model for research reactor lattice cal-

culations was developed. The model consists of 63 nuclides treated explicitly and one fissile–independent pseudo–element. In the new model, the fraction of the pseudo–element to total fission –product absorption was reduced within 1% in all the burnup range. This model is capable of predicting the exact burnup–dependent behavior of low–enriched uranium fueled research reactors, up to approximately 200 GWD/MTU. With the results mentioned above, it is concluded that the 63 explicit nuclides plus one pseudo–element model developed in the present work can offer an effective and/or accurate means for fission–product representation in thermal research reactors over a wide range of burnup. Also, this model should be checked, in future, with experimental data and/or new nuclear data for fission–product poisoning as a final verification of its adequacy.

References

1. J.D. Garrison et al., "Fission–Product Capture Cross Sections", Nucl. Sci. Eng. 12, 115–134(1962)

2. E.A. Nephew, "Thermal and Resonance Absorption Cross Sections of the U^{233}, U^{235}, and Pu^{239} Fission Products".

ORNL-2869(1960)

3. M. Edenius et al., "The EPRI-CPM Data Library", Chap. 4, Part II, EPRI CCM-3(1977)

4. M.J. Halsall, "A summary of WIMSD4 Input Options", AEEW-M-1327(1980)

5. S. Iijima et al., "Fission Product Model for BWR Lattice Calculation Code", Jour. Nucl. Sci. Tech. 19, 96~106(1982)

6. J.V. Donnelly, "Modifications to WIMS-CRNL Burnup Calculations", AECL-9195(1986)

7. M.W. Rosenthal et al., "A Comparative Evaluation of Advanced Converters", ORNL-3686(1965)

8. E. Teuchert et al., "V.S.O.P.-Computer Code System for Reactor Physics and Fuel Simulation", Jul-1649(1980)

9. H. Takano et al., "The Effect of Fission Products on Burnup Characteristics in High Conversion Light Water Reactors", Nucl. Tech. 80, 250~262(1988)

10. D.H. Kim et al., "KMRR Design and Construction Project", KAERI/RR-813/88(1988)

11. H.R. Kim, "WIMS-KAERI for the Extended KMRR Physics Calculations", KAERI Internal Report(1987)

12. Jung-Do Kim et al., "Generation and Benchmarking of a 69-group Cross Section Libratry for Thermal Reactor Applications", Journal of Korean Nuclear society, 21, 245~258(1989)

13. M.J. Halsall et al., "The '1986' WIMS Nuclear Data Library", AEEW-R 2133(1986)

14. C.J. Taubman, "The WIMS 69-group Library Tape 166259", AEEW-M1324(1975)

15. C. Allen et al., "ORIGEN2 : A Versatile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials", Nucl. Tech. 62 335-352(1983)

16. O. Schwerer et al., "ENDF/B-5 Fission Products Library", IAEA-NDS-25 (Rev. 2) (1984)

17. R.E. MacFalane et al., "The NSOY Nuclear Data Processing System", LA-9303-M, Vol.1(1982)