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Validation of KEDAK-4 Data for Thermal Reactors: Resonance Integrals and Postirradiation Fuel Analysis*

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Abstract

An optimal solution of problems relating to the fission reactor fuel cycle requires the ability to calculate the isotope production as a function of reactor history. For this purpose neutron cross section data for ^{241}Am, ^{242m}Am, ^{243}Am and ^{244}Cm were evaluated for the German nuclear data library KEDAK-4. For some isotopes a large discrepancy between the resonance integrals and the effective thermal cross sections calculated from KEDAK and measurements in a reactor spectrum was observed. In an earlier report the discrepant thermal data were analysed. In this report the results of resonance integral measurements and of post irradiation analysis are critically examined and compared with those obtained from KEDAK-4 data. Good agreement between the calculations and reliable experimental data is observed.

Absicherung der KEDAK-4-Daten für Thermische Reaktoren durch Resonanzintegrale und Nachbestrahlungsanalysen.

Zusammenfassung

Zur Beantwortung von Fragen des nuklearen Brennstoffkreislaufs ist es notwendig, das nukleare Inventar abgebrannter Kernbrennstoffe zuverlässig zu berechnen. Zu diesem Zweck wurden für die Karlsruher Kerndatenbibliothek KEDAK-4 Neutronenwirkungsquerschnitte für die Nuklide ^{241}Am, ^{242m}Am, ^{243}Am und ^{244}Cm berechnet und ausgewertet. Für einige Nuklide gibt es große Diskrepanzen zwischen berechneten KEDAK-Werten für Resonanzintegrale und effektive thermische Wirkungsquerschnitte und den in einem Reaktorspektrum gemessenen Werten. In einem früheren Bericht wurden die diskrepanten thermischen Daten analysiert. In diesem Bericht werden die Messungen von Resonanzintegralen und experimentellen Nachbestrahlungsanalysen kritisch analysiert und mit Berechnungen auf der Basis von KEDAK4-Daten verglichen. Es ist eine gute Übereinstimmung zwischen Berechnungen und verläßlichen experimentellen Daten festzustellen.

Contents

Introduction

An efficient radioactive waste management stipulates an accurate knowledge of the inpile and out of pile characteristics of the burnt fuel. The isotopic composition of the fuel is to be calculated as a function of reactor history. The accuracy of these calculations depends upon the calculational models of the reactor codes and the quality of the nuclear data used. For some of the applications in the field of thermal reactors it is sufficient to know the effective cross sections, which in turn can be derived from the knowledge of thermal cross sections and the resonance integrals. In this paper we examine the present status of the resonance integrals of the isotopes of Am and ^{244}Cm. A comparison of the measured resonance integrals with those calculated from the evaluated neutron data files, such as KEDAK-4, UKNDL and ENDF/BV, is also done wherever the data are available. The compilation of Gryntakis and Kim/1/ with some additions from CINDA form the basis of this survey. The results of the post irradiation fuel analysis which can be an important tool to check the quality of the data used in the burnup calculations are also critically examined in this report.

Resonance Integrals

During the course of the evaluation/2/ of the neutron data for the actinide isotopes a wide range in the published values for the resonance integrals was observed. The published resonance integrals can be classified in 4 different categories:

a) Calculated from the differential data

b) Semiempirical nuclear systematics

c) Direct measurement using more or less pure isotopes

d) Resonance Integrals derived from the isotope production measurements in the reactor fuel.

In this paper the data belonging to the categories a) and b) will not be discussed as it is assumed that the current evaluations include all the scrutinised information about the differential data and nuclear systematics. The measurement of resonance integrals is done using the Cd-difference technique. The probe is protected from the thermal neutron flux with a Cd-cover. The thickness of the Cd-cover determines the lowest energy of neutrons reaching the probe. If strong resonances are present in the region of Cd-cutoff energy the measured resonance integral will strongly depend on the thickness of the cadmium sheet. The measurement and the interpretation of the resonance integrals in the actinide region is hampered due to the presence of strong resonances (Fig.1) near the Cd-cutoff

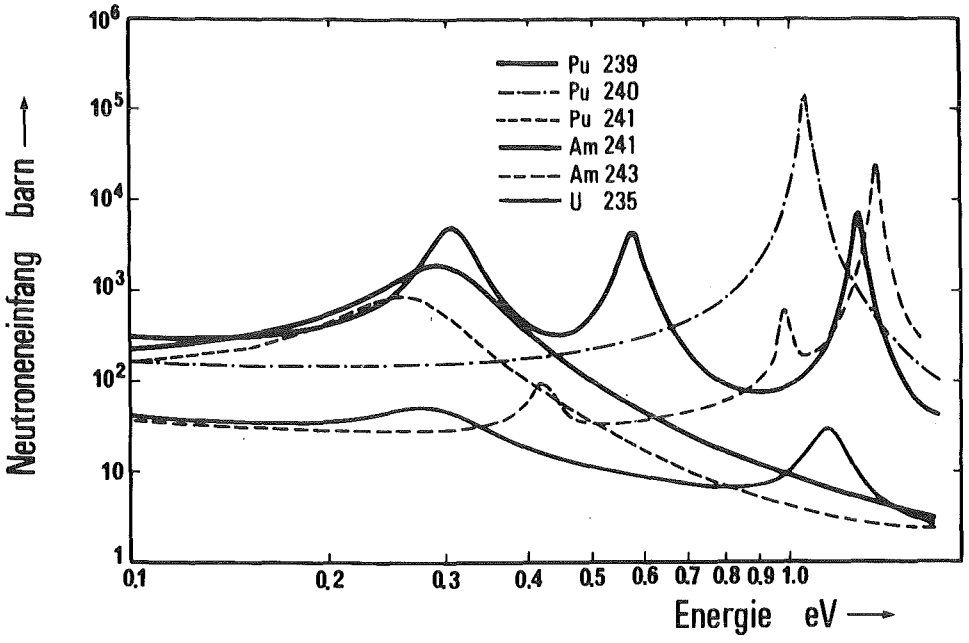

Fig. 1 Capture cross sections for different actinides.

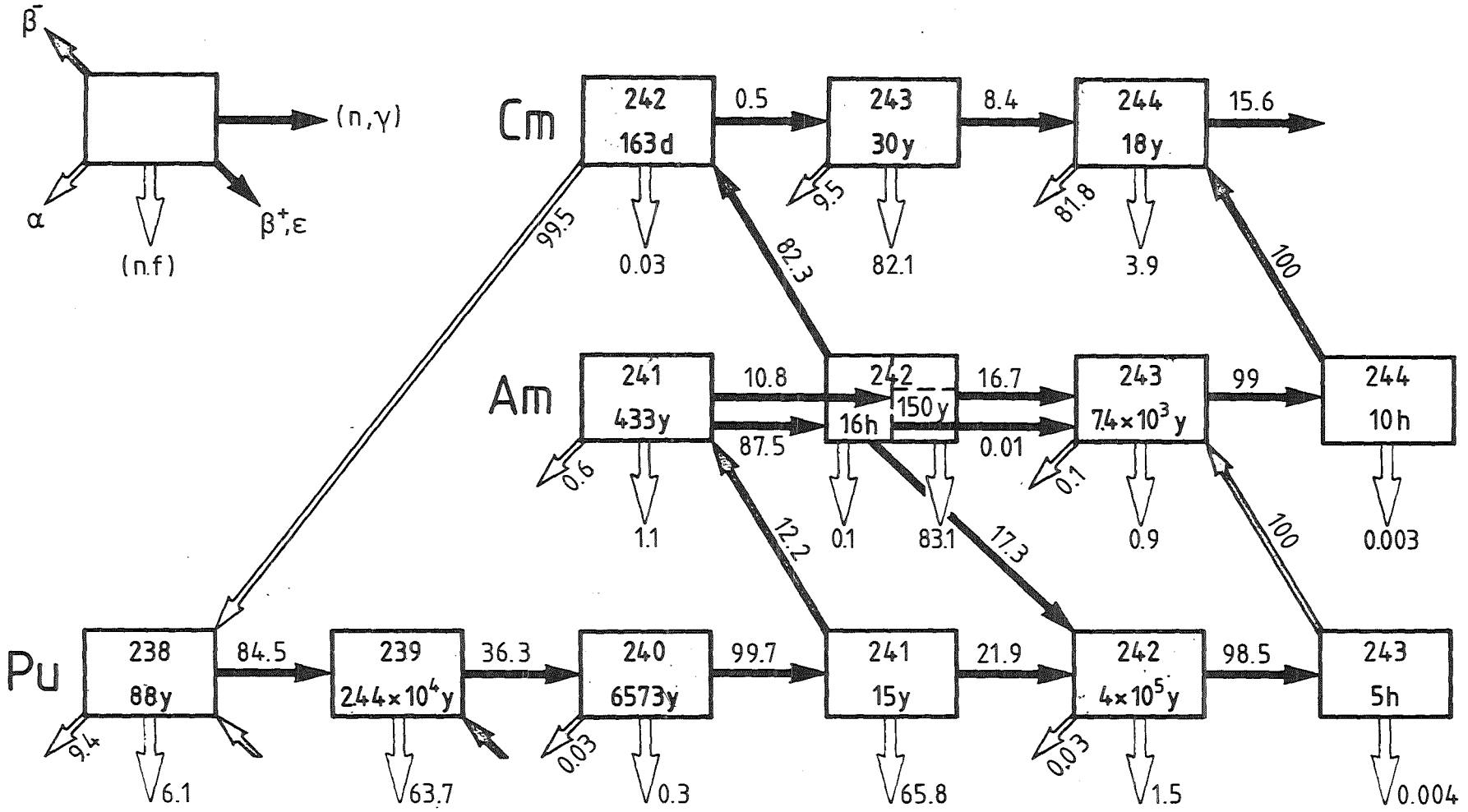

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Fig.2 ISOTOPE PRODUCTION IN A THERMAL REACTOR ($\Phi_{th}=10^{13}$ n / cm^2 · sec)

energy. Thus a precise knowledge of the Cd-cutoff energy is a prerequisit for a proper evaluation of the resonance integral. This information, however, is often missing in the publications. The measurements of the type d) are further complicated due to the mutual resonance shielding i.e. the presence of isotopes with strong resonance in or near the sample causes the shape of neutron flux to deviate from the assumed 1/E behaviour. The production of Am and Cm isotopes in the reactor fuel is a result of multiple neutron absorption which takes place in competetion with the different decay modes of the intermediate isotopes(Fig. 2). Though some of the data uncertainties may compensate each other others may accumulate to give a larger uncertainty of the measured value.

In the following we examine the experimental data for different isotopes.

^{241}Am:

For this isotope there are a number of measurements. Due to the two strong subcadmium resonances at 0.308 eV and 0.577 eV the measured value depends strongly on the cadmium cutoff energy and consequently the range in the published values is large. The resonance integrals change by more than a factor of 2 by varying the Cd-cutoff energy from 0.2 eV to 0.7 eV. A similar effect was observed and discussed previously/3/ for the thermal cross section of ^{241}Am. Table I reproduces the results of different measurements for resonance integrals for ^{241}Am.

TABLE I Measured resonance integrals for ^{241}Am

Author	year	E_{Cd}	capture	fission
Bak +/4/	1967	(0.3)	2400 ±200	21 ± 2
Schuman/5/	1969		1100 ±100	
Eberle+/6/	1972		1390	21
Harbour+/7/	1973	0.369	1538 ±135	
Zhuravlev+/8/	1975			27.7 ±1.6
Gavrilov+/9/	1977	0.68	1800 ±100	22.5 ±0.25

Now let us examine the different data critically. Bak et al./4/ have measured the capture resonance integrals for ^{241}Am and ^{243}Am and fission integral for ^{241}Am.
For the measurement of ^{241}Am resonance integrals a "pure" sample was used but in the publication no information about the purity of the probe is given. Information about the cutoff energy is also missing. In Ref. /3/ it was shown that the effective cutoff energy in this experiment is likely to be arround 0.3 eV. Thus both resonances at 0.308 eV and 0.577 eV would contribute to the resonance integral. The value of both capture and fission resonance integrals are consistent with this cutoff energy (Fig. 3).

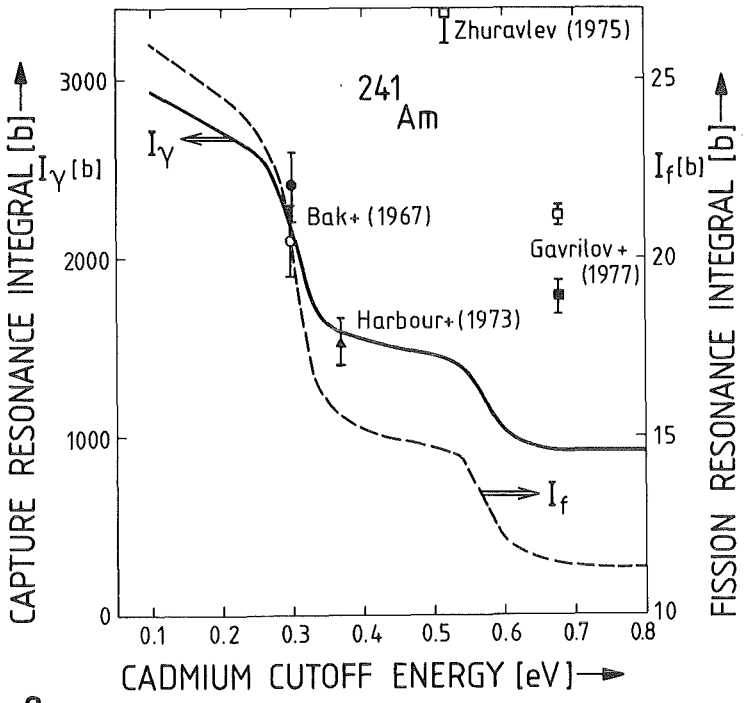

Fig. 3

RESONANCE INTEGRALS FOR ^{241}Am AS A FUNCTION OF Cd-CUTOFF ENERGY

FULL SYMBOLS ARE FOR CAPTURE.
EMPTY SYMBOLS ARE FOR FISSION.

The aim of Schuman's work/5/ was to produce samples rich in ^{242}mAm. Pure samples of ^{241}Am were irradiated in high flux core positions of ETR. The values given for resonance integrals are not corrected for variable Cd-shields. The value obtained for the isometric ratio in this experiment is 0.773. This is much lower than the value obtained in other experiments and the presently recommended value of the isomeric ratio. For these reasons the results of this experiment are to be discarded.

Eberle et al./6/ is an example of the category d) measurements. Pu-Al-aloy containing 1.5% Pu was irradiated in a reactor until all the Pluonium was burnt and the production of different actinides was measured. The initial composition of the sample was 90.91% ^{239}Pu,

8.22% ^{240}Pu, 0.83% ^{241}Pu and 0.04% ^{242}Pu. The nuclear data were taken from the literature and adjusted to reproduce the experimentally determined isotopic compositions of the irradiated probe. Moreover, the ratio of thermal to epithermal flux was also varied to obtain an optimal agreement between experiment and calculations. In most cases the isotope production was a result of multiple neutron absorption in competion with different decay modes of the intermediate nuclei. Thus an accurate knowledge of constants for all intermediate processes is required to interprete the experiment properly. This type of experiment is of only a limited use to extract cross sections. Though it can be an important tool to check the consistency and the accuracy of the nuclear data for a definite purpose.

The best available measurement for the capture resonance integral of ^{241}Am is that of Harbour et al./7/. They have measured the equivalent thermal capture cross section and capture resonance integral on a highly pure (> 99.9% ^{241}Am) probe and have carefully examined the effect of epithermal to thermal flux ratio and the cadmium cutoff energy on the measured data. They give a value of 1538 ± 135 b for a Cd-cutoff energy of 0.369 eV. The value calculated form KEDAK-4 data for this cutoff energy is 1549 b which is in excellent agreement with the result of Harbour et al.

Zhuravlev et. al./8/ have measured the fission resonance integrals for a number of isotopes ranging from ^{239}Pu to ^{249}Cf. Though the details of the calculation are not published , corrections for various effects have been applied to the data. Nevertheless their results for ^{241}Am with strong subcadmium resonances are up to a factor of 2 higher than the values obtained from differential data. The Cd-cutoff energy is given to be

0.52 eV. Even reducing this energy to an extremely low value will not match their results to the differential data. We believe that there is some unrecognised error in this experiment and discard their data from present evaluation.

Gavrilov et al./9/ have measured the capture and fission resonance integrals for ^{241}Am and ^{243}Am. They have also used almost pure isotopes and the thickness of the Cd-shield is given to be 1 mm which corresponds to a cadmium cutoff energy of 0.68 eV for pure 1/v absorbers. Since the investigated isotopes have strong subcadmium resonances the effective cutoff energy is likely to be lower than this value. The authors claim a good agreement between their results and those of Harbour et al/7/. This again is not consistent with the cutoff energy of 0.68 eV. As is seen in Fig. 3, the capture integral reduces by about 40% by changing the cutoff energy from 0.369 eV to 0.68 eV. Gavrilov et al.'s value on the contrary is 17% larger than that of Harbour et al. The authors have observed subcadmium neutrons while checking the method on ^{239}Pu. These were then avoided by reducing the detector size. Since the subcadmium resonance in ^{239}Pu is at a lower energy than those of ^{241}Am and ^{243}Am neutrons which do not appreciably effect the fission integral for ^{239}Pu may still enhance the resonance integral for Am isotopes.

^{242m}Am

For this isotope there are 6 entries in Ref. /1/. Out of these only two are of the category c), namely Schuman/5/ for the absorption resonance integral and Zhuravlev et al./8/ for the fission resonance integrals. Schuman did this measurement concurrent to that of ^{241}Am. The value derived for the ^{242}mAm depends

strongly on the cross sections adopted for ^{241}Am. Since these are in large error the same is also true for his value of 7000 ± 2000 b for the absorption resonance integral of ^{242m}Am. Thus the only reliable data for the capture resonance integral for this isotope are due to differential measurements of the capture cross section. The value derived from the KEDAK-4 evaluation is 280 b for the capture integral and 1909 b for the absorption integral.

For the fission resonance integral the only measurement is that of Zhuravlev et al/8/. They give a value of 2260 ± 100 b for ^{242m}Am. This value is about 40% higher than the value of 1629 b derived from KEDAK-4 which is based upon the differential data. The sample contained only about 1% of ^{242m}Am. Thus the contribution to the fission count rate of the probe due to fission in ^{241}Am and ^{243}Am, in spite of their comparitively low cross sections, is of the same order as that due to ^{242m}Am. Their value for fission integral of ^{241}Am as shown above is too large. If a smaller value for fission integral of ^{241}Am is taken to extract the fission integral of ^{242m}Am its value will be still larger. This discrepancy can not be presently resolved. It is recommended to use the value derived from the differential data.

^{243}Am

Table II gives experimental data for ^{243}Am. It is seen that the results of different authors for capture integral agree well within the experimental uncertainties. Fig. 4 shows that the resonance integral for this isotope, in spite of a resonance at 0.4 eV, is not very much dependant on the cadmium cutoff energy. The value of the capture resonance integral for ^{243}Am based

on differential data (KEDAK-4) however is 1847 which is about 20% lower than the results of the integral mesurements. An increase in the resonance parameters to match this value can not be justified at present. An explanation for this discrepancy has yet to be found.

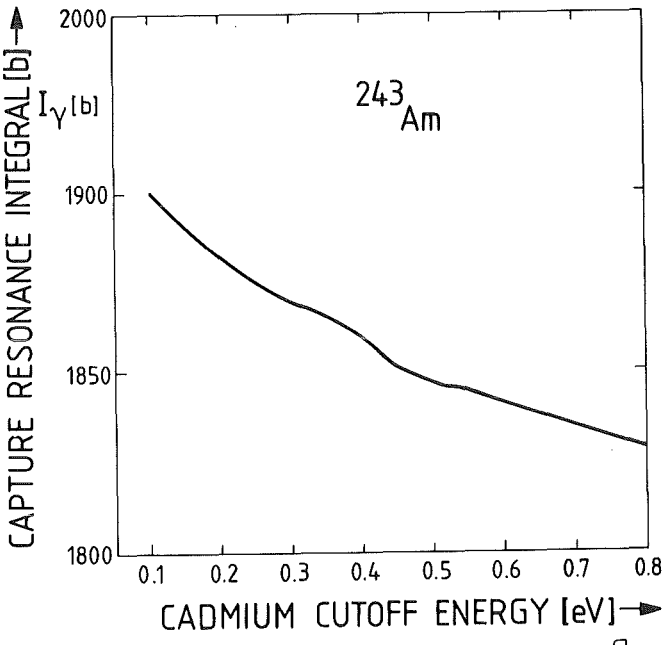

Fig.4

RESONANCE INTEGRALS FOR ^{243}Am AS A FUNCTION OF CADMIUM CUTOFF ENERGY

TABLE II Measured resonance integrals for ^{243}Am

Author	year	capture	fission
Butler +/10/	1957	2290 ± 50	
Bak +/4/	1967	2300 ± 200	
Folger +/11/	1968	2250 ± 50	
Zhuravlev+/8/	1975		9 ± 1
Gavrilov+/9/	1977	2210 ± 150	17.1 ± 1.3

For the fission resonance integral of ^{243}Am the differential information is lacking and the two integral measurements differ by a factor of 2. For the reasons given above we discard the data of Zhuravlev et al./8/ and recommend the results of Gavrilov et al./9/.

^{244}Cm

For this isotope there are 6 category c) measurements for resonance integrals. The eldest measurement is due to Folger et al./11/. Folger et al. and Smith et al./12/ are twin papers on this subject. The value measured in Ref. /11/ is 700 b. Ref. /12/ uses a value of 631 b for the analysis of their experiment. A reason of this inconsistency is not given in the

publications. Both these values are, nevertheless, consistent with the result of Schuman/13/. Since the first resonance for this isotope is at 7.67 eV the results are not affected by the variable Cd-shields. Schuman reports a large uncertainty in the ^{245}Cm content of the sample. It is not clear whether this is included in the uncertainty of ± 50 b or not.

The experiment of Thompson et al./14/ was essentially performed to measure the fission resonance integrals of Cm isotopes. It was done in two parts, short irradiation measurement to determine the fission integral with respect to ^{235}U and long irradiation measurement to obtain a consistent set of cross sections for Cm isotopes. In short irradiation measurement samples with 93 - 95% enriched ^{244}Cm were irradiated in Cd-capsules together with ^{235}U for about 5 hours. The buildup of ^{131}I was measured with γ-spectroscopy thus obtaining a direct measurement of fission integral with respect to ^{235}U. In the long irradiation experiment samples with different compositions were irradiated long enough to transmute a substantial amount of the probe. From the isotope production calculations the value of capture integral of 650±50 b for ^{244}Cm could be confirmed. The value of fission integral was found to be 12.5 ± 2.5 b. The paper states that this value of resonance integral has been confirmed by fission chamber measurements with a 99.02% ^{244}Cm sample. This measurement with 99.02% ^{244}Cm sample was performed by Benjamin et al./15/. The measurement was done relative to ^{235}U with fission chambers and with solid state track detectors. The value of fission resonance integral for ^{244}Cm is given to be 18 ± 1 b. We recommend this value as the latest result from SRL for the fission resonance integral of ^{244}Cm.

Gavrilov and Goncharov/16/ have published a short note

indicating the measurement of capture resonance integrals for Cm isotopes. The work of Zhuravlev et al. has already been discussed. Table III shows results of different authors. It is seen that capture integral values of different authors agree well with each other.

TABLE III Measured resonance integrals for ^{244}Cm

Author	year	capture	fission
Folger +/11/	1968	700	
Schuman /13/	1969	650 ± 50	
Thompson+/14/	1971	650 ± 50	12.5 ± 2.5
Benjamin+/15/	1972		18.0 ± 1
Zhuravlev+/8/	1975		13.4 ± 1.5
Gavrilov+ /16/	1978	626 ± 53	

Comparison of Different Evaluations

In Table IV and V the resonance integrals calculated from different libraries of evaluated data are compared. The resonance integrals for UKNDL and JENDL are due to Mattes/17/. It is seen that the agreement among the different data is good. Except for ^{241}Am, where there

TABLE IV Capture resonance integrals from different evaluations

	^{241}Am	^{242m}Am	^{243}Am	^{244}Cm
KEDAK-4	1452	280	1847	637
UKNDL	1415		1846	
JENDL	1299	207	1816	593
ENDF/B V	1420	286	1818	595

TABLE V Fission resonance integrals from differen evaluations

	^{241}Am	^{242m}Am	^{243}Am	^{244}Cm
KEDAK-4	14.8	1629	4.4	18.1
UKNDL	15.02		5.95	
JENDL	14.7	1575	11.4	17.8
ENDF/B V	13.4	1886	6.15	18.7

is a sufficient experimental data base the agreement is rather deceiving and reflects the limited data base. For example for the evaluation of ^{243}Am data in the resonance region there is practically only one publication, namely that of Simpson et al./18/, on which different evaluations are based. The measured capture resonance integral for ^{243}Am is about 20% larger than the values given in Table IV. The KEDAK value of the capture resonance integral for ^{244}Cm is higher than other evaluations. This is due to the inclusion of the data of Kalebin et al./19/. Including these data in the resonance analysis leads to a larger value for the neutron width of the first few resonances in ^{244}Cm (see Ref.2).

Post Irradiation Fuel analysis

As has been stated before the isotopic composition of the spent fuel can be used as a tool to check the data used in burnup calculations. Whether this is a good check or not depends upon: a) the quality of calculational methods and b) the reliablity of the experimental results.

At KfK the isotopic composition of the spent fuel is calculated using the computer code KORIGEN/20/. This is an improved version of the well known code ORIGEN/21/. The improvements concern physics of the code as well as the improved and extended data base. The main feature of these improvements is that the effective cross sections are not derived from the thermal cross sections and resonance integrals rather they are calculated from a 83 group (30

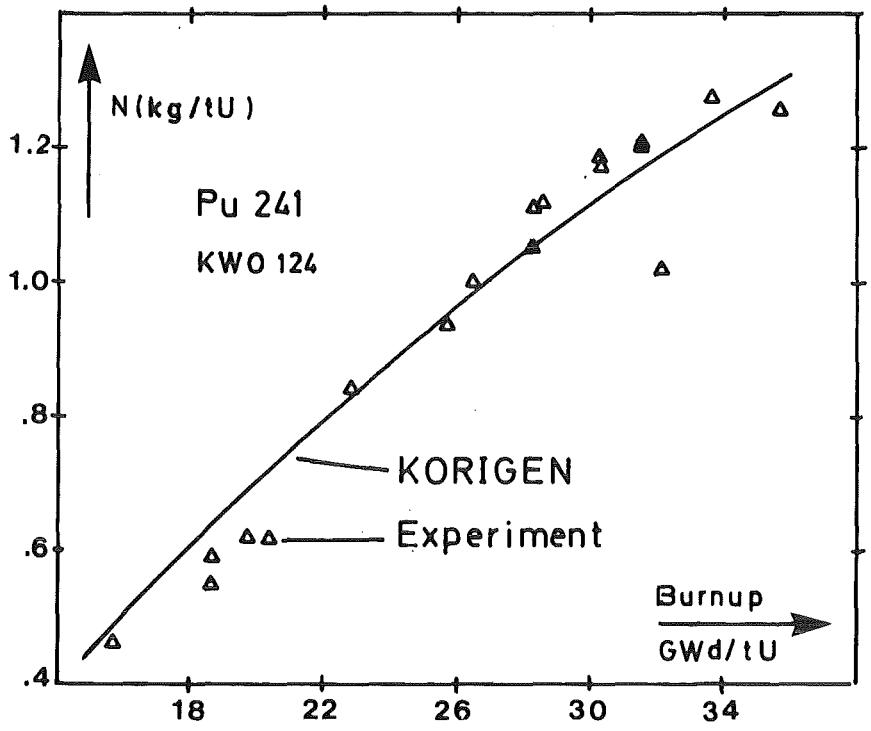

Fig. 5 ^{241}Pu concentration in the spent fuel of the PW reactor KWO

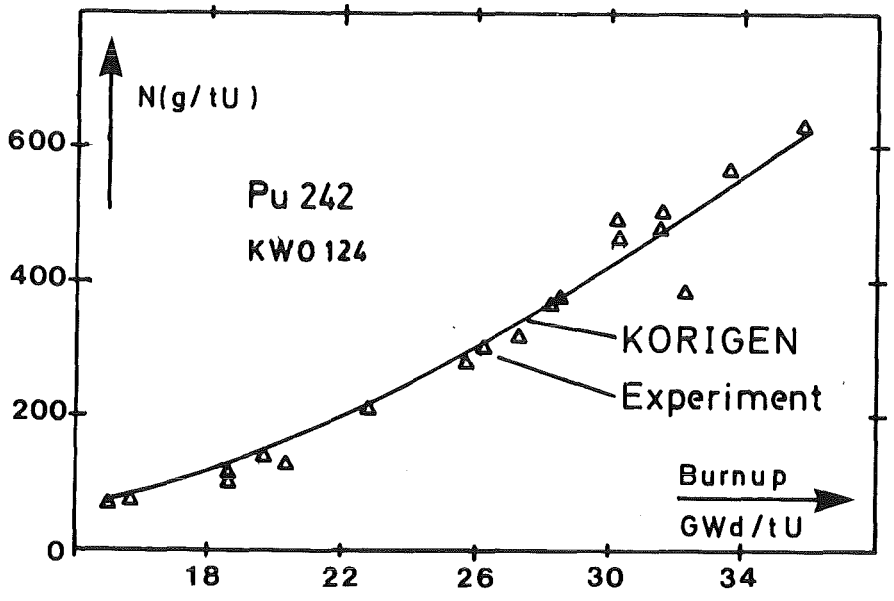

Fig. 6 ^{242}Pu concentration in the spent fuel of the PW reactor KWO

thermal and 53 epithermal groups) cross section set. This cross section set is in turn generated from the differential data library KEDAK-4. The effective cross sections are calculated at each burnup stage using the current neutron spectrum obtained with time dependant cell calculations. This enables to take account of effects like resonance shielding, spectrum hardening etc. as a function of burnup in a consistent manner. To calculate the isotopic composition of the spent fuel a good knowledge of its irradiation history is essential. This information is very often of commerical nature. Therefore, the analysis is limited to those cases where we have access to the irradiation data. Post irradiation fuel analysis was performed for different reactors. In a subsequent report/20/ an extensive overview is given. In this report the results for the prototype 350 MWe PWR at Obrigheim (KWO) are presented. The experimental composition and the irradiation data are taken from Ref. /22/, /23/ and /25/.

The validity of the code has been checked by reproducing the measured isotopic composition of the major actinides i. e. for the isotopes of U and Pu for different reactors. Fig. 5 and 6 compare the experimental and calculated concentrations of isotopes ^{241}Pu and ^{242}Pu in the spent fuel of the reactor KWO.

The measurement of the isotopic compostion of the spent fuel is done after a cooling period whose length is determined by technical and managerial problems. Generally it is of the order of 2 to 3 years. During this period ^{242}Cm with its half life of 162 days has decayed to ^{238}Pu. A substantial amount of ^{241}Am has been generated through the decay of ^{241}Pu with the half life of 14.35 $\pm$ 0.02 years/24/. The measurement of isotopic

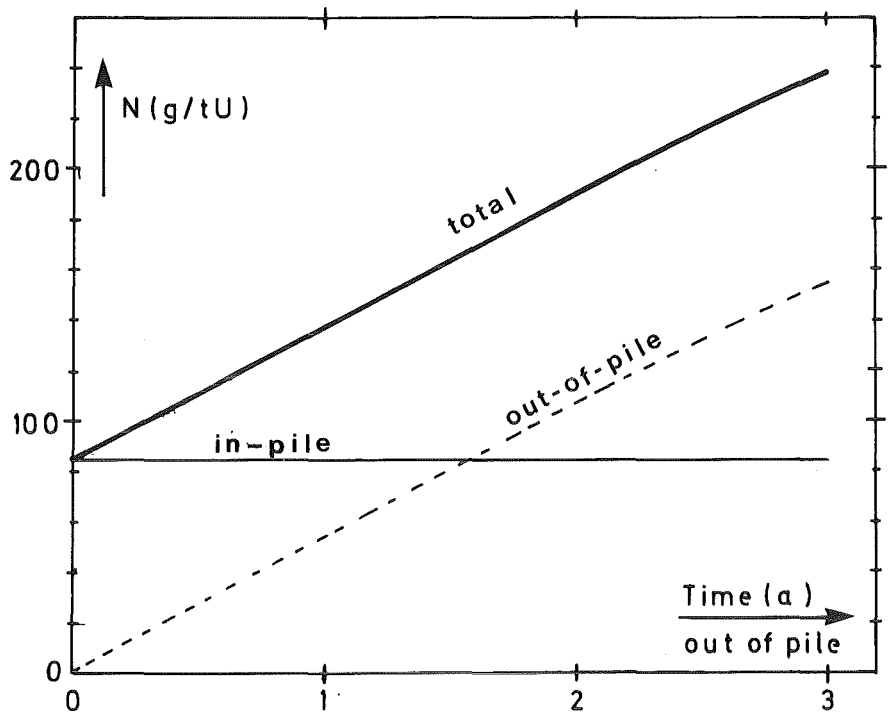

Fig. 7 Out of pile accumulation of ^{241}Am in spent fuel

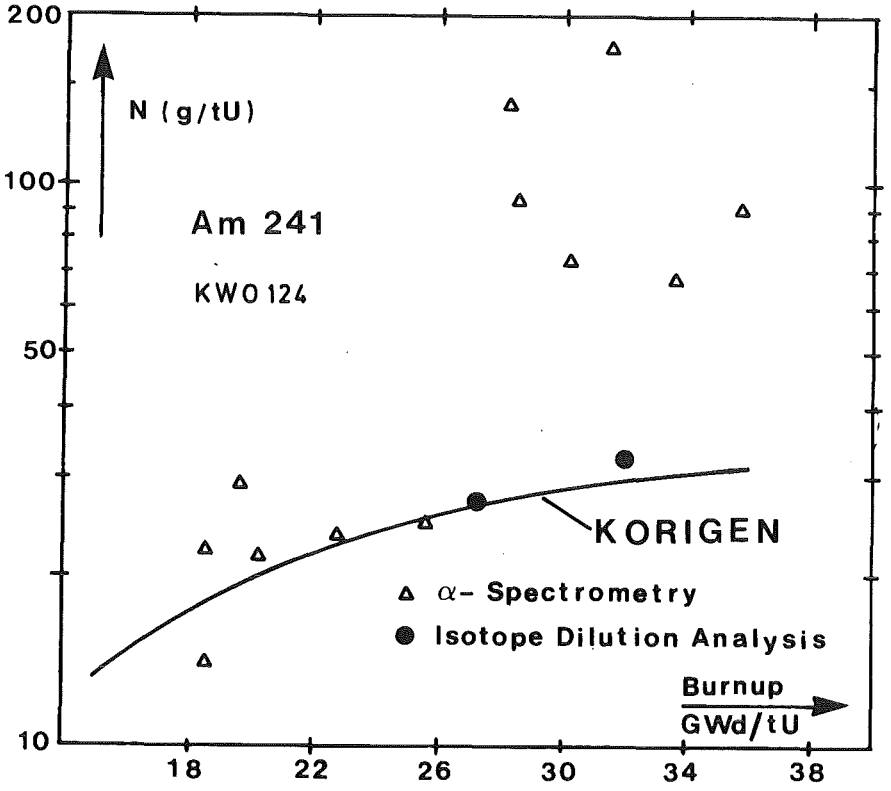

Fig. 8 ^{241}Am Concentration in the spent fuel of KWO

concentration of Am and Cm isotopes is usually performed with the methods of α-spectroscopy and mass analysis.The α-peaks of ^{241}Am are overshadowed by ^{238}Pu, which is formed in the reactor mainly via the reaction ^{237}Np(n,γ)^{238}Np--β-->^{238}Pu and the decay of ^{242}Cm as mentioned above. This is the main source of error in the determination of ^{241}Am concentration as it is obtained by measuring the ^{238}Pu-^{241}Am peak before and after the extraction of Pu from the spent fuel. This is a difference of approximately equal numbers and accordingly the error in the ^{241}Am measurement is large. Moreover the account of he ^{241}Am formed due to the out of pile decay of ^{241}Pu has to be made. Fig. 7 shows the out of pile increase in the ^{241}Am concentration in the spent fuel due to the decay of ^{241}Pu. It is seen that at the time of experimental analysis the ^{241}Am produced in the reactor accounts for only 25% of the measured ^{241}Am concentration. Due to the uncertainty in the measured ^{241}Am concentration which is of the same order, this isotope is not suitable for data check. Only recently a measurement/25/ using the isotope dilution technique for the determination of the concentrations of Am isotopes in the spent fuel has become available. This technique due to higher experimental accuracy is more suitable to determine the ^{241}Am concentration. Results of these are in good agreement with the calculated ^{241}Am concentrations (Fig. 8), whereas results of α-spectroscopy measurements show a large spread.In the past ^{243}Am was measured with mass analysis relative to ^{241}Am and thus the error in the ^{241}Am concentration also dominates the error in the ^{243}Am concentration. In the measurements based on the isotope dilution technique the ^{243}Am concentrations are free from the errors in the ^{241}Am concentration. Fig. 9 compares the results of isotope dilution measurement/25/, which is in good agreement with the earlier measurement

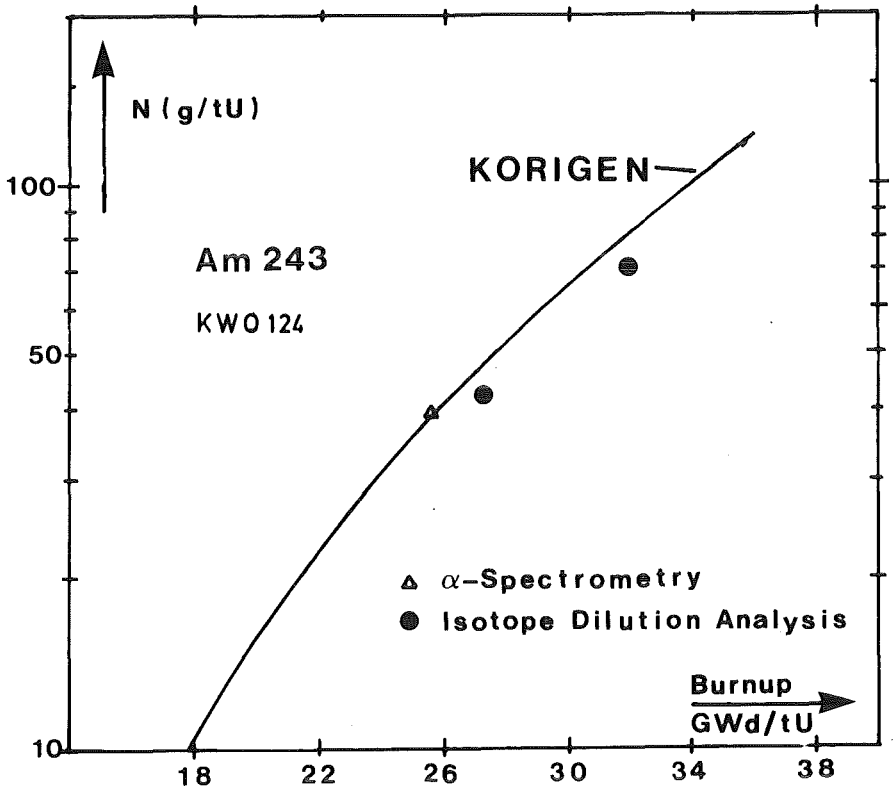

Fig. 9 ^{243}Am concentration in the spent fuel of KWO

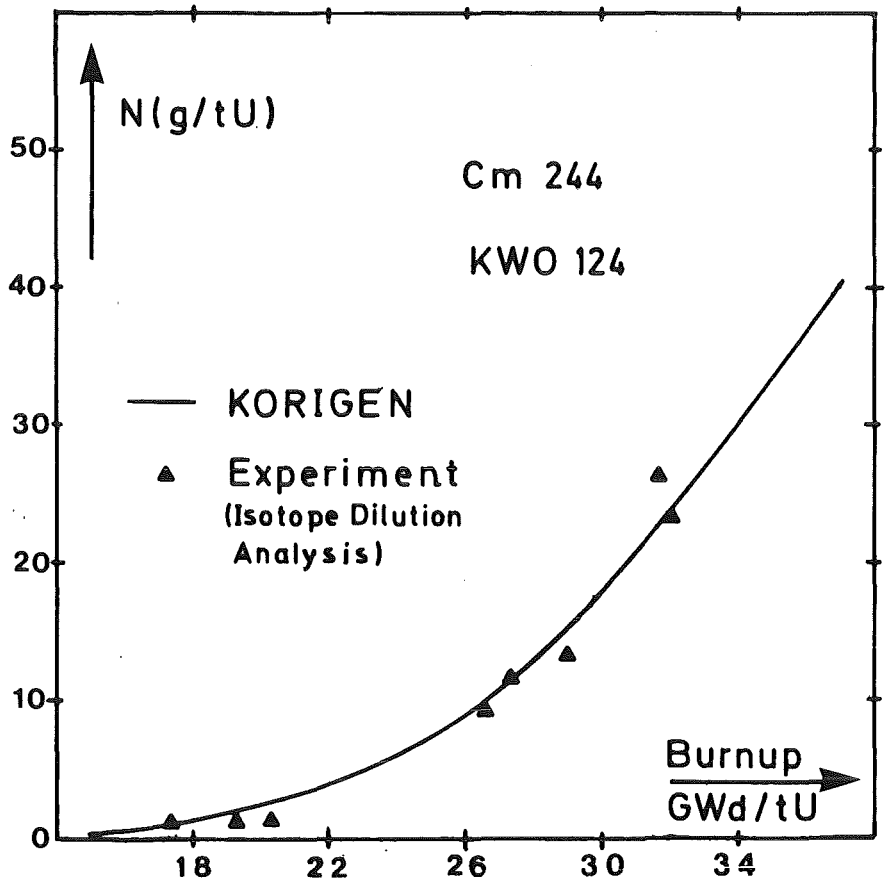

Fig. 10 ^{244}Cm concentration in the spent fuel of KWO

based on α-spectroscopy/22/, with the results of the code KORIGEN. A good agreement is observed.

For ^{244}Cm a similar good agreement is observed (Fig. 10) when compared with the measurements due to isotope dilution technique. Since more than 99% of ^{244}Cm is built via ^{242}Pu and ^{243}Am the good agreement between calculated and measured concentration of Pu isotopes as well as ^{243}Am and ^{244}Cm indicate the validity of the data for the involved processes, namely the neutron cross sections for $^{242}Pu(n,\gamma)$, $^{243}Am(n,\gamma)$ and $^{244}Cm(n,abs.)$. To verify the same for ^{241}Am it is necessary that isotopic analysis of the spent fuel is made soon after irradiation i.e. before the ^{241}Am formed out of pile dominates the experimental value.

Conclusion and Recommendations

The validity of the KEDAK-4 data as used in the burnup code KORIGEN to predict the isotopic formation of higher actinides in the spent fuel of thermal reactors has been demonstrated in the preceeding chapter.

For ^{241}Am different evaluations are consistent with each other and with integral data except that of Zhuravlev et al./8/.

For ^{242m}Am the data basis is poor but the importance of this isotope in burnup calculation is low.

For ^{243}Am there is a discrepancy of about 20% between the capture resonance integral value calculated from differential data and those measured in a reactor.A new measurement seems necessary to resolve this discrepancy.

For ^{244}Cm the data are in good shape. The recent evaluation of differential data (KEDAK-4) give about 5% higher value for the capture resonance integral of ^{244}Cm. This value is supported by the recent measurement of Gavrilov and Goncharov/16/ and the post irradiation fuel analysis calculations.

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The authors thank Dr. Küsters for his support and many extensive and fruitful discussions.