Evaluation of Thermal Reactor Fission Products Cross Sections

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Abstract

The production of fission products during reactor operation has a very important effect on reactor reactivity .Results of neutron cross section evaluations are presented for the main product nuclides considered as being the most important for reactor calculation and burn-up consideration . Data from the main international libraries considered as containing the most up-to-date nuclear data and the latest experimental measurements are considered in the evaluation processes, we describe the evaluated cross sections of the fission product nuclides by making inter comparison of the data and point out the discrepancies among libraries.

Key words: Absorption Cross-Sections, Nuclear Reactors, Fission Products, International Data Base Library, Fission Yields.

Introduction

The production of fission products during reactor operation has a very important effect on reactor reactivity (i.e. effective multiplication factor).Evaluation of neutron absorption cross sections of fission products is one of the most important and essential subject needed to the save work of reactors. The nuclear properties and particularly the absorption cross sections are of an extreme importance not only for the fission products but also for all reactor components including eventually the moderator and structure materials. An extensive work has been done for low-energy fission up to 15 MeV in order to form a complete nuclear data library for all types of cross section. Data concerning the fission products are needed too as they are also extremely important in other fields of calculations such as understanding the nature of fission process, determining of the fuel burn- up ,performing shielding calculation , calculating decay heating power, estimation of the amount of gas production and nuclear transmutation of fuel in nuclear reactors ,estimation of radiation damage of all reactor material and components ,neutron dosimetry of produced nuclides and other special applications .[1]

The importance of fission products results from the fact that practically all stages of the nuclear fuel cycle are affected by their presence. A number of different national and international comities have been engaged in considerable theoretical and experimental efforts to build their nuclear data libraries. The most recent evaluated neutron nuclear data libraries concerning the fission products are actually JENDL-3.3 from Japan[2,3] EXFOR from IAEA [4],ENDF/B-V1 release 8 from United States[5],JEFF-3.0 and JEF-2.2 from Europe [6] , BROND-2 from

Russia [7] ,and CENDL-3.0 from China [8] . These libraries are considered now as containing the most up-to-date nuclear data of evaluated (recommended) cross sections, spectra, angular distributions, fission product yields, thermal neutron scattering, photo-atomic reactions, and other data which are important in neutron-induced reactions relevant to reactor calculations [9,10] .They contain too cross sections of numbers of fission products nuclides according to their yields , half-lives ,capture , elastic, inelastic and absorption cross sections, for example the Chinese library contain 138 fission product nuclides which are evaluated in the energy range from 1.e-5 eV to 20. MeV, while the Japanese, European, Russian and American libraries, they contain 209, 203, 36 and 199 respectively mostly in the same energy range.

In spite of all these voluminous and important data, the IAEA, (The International Atomic Energy Agency), point out that, there are still considerable discrepancies among the evaluated data sets. [11]

The discrepancies are in particular attributed to the absorption cross sections of the fission products, which are considered as the most important nuclides in the reactor operation. The fission products absorption cross section account for more than 40% of the total neutron absorption cross section of materials composing the reactor core [11] which constitutes the major contribution to the reactivity loss in a reactor, for a medium enriched uranium core (MEU core), it is approximately equal to about 9% of the reactivity value and increase with increasing burn-up or neutron flux. Eventually their concentration increases too with radiation, although they have relatively small capture cross sections, their build up factor near EOL is important to the reactivity balance of the reactor.

At the end of the core lifetime (EOL), about 30% of the nuclear fuel (U-235 or Pu-239) initially loaded, in the core, as fuel elements is burned and converted into fission products.

The data of some of the isotopes given by the libraries were obtained by a special numerical evaluation method based on the fitting of nuclear fragments of mass distributions by several Gaussian functions. [12] The generation of nuclear data based on theoretical physic models is frequently used when no experimental data are available. [13] In particular the total cross sections above the resonance region are generally evaluated using the optical model by fitting the data to the measured total cross sections.

This method allows filling the gaps in the data of experimental results. The IAEA estimate that there is an important lack of the data for higher energy fission to form reliable physical models of nuclear fission to be applied for the nucleon induced fission in 20-200MeV energy region. [14] In this region there still some important discrepancies of neutron cross section data due to experimental errors and statistical fluctuation of all kinds of nuclear reactions or kinds of theoretical models used for the calculation.

Calculation Methods

One of the main important utilization of the fission product cross sections is illustrated in the time-dependent behavior of neutron absorption in fission products which is considered as a major problem concerning the reactor depletion calculation. The degree of complexity required for adequate treatments of fission products build up depends on several factors, such as, energy spectrum, fuel isotope composition and burn up rate, neutron capture and total cross sections, isotopic yields productions...etc.

An exact calculation of full fission products build up is not feasible since there are several hundreds of fission products nuclides, and some number of lattice group constants for core calculation which have to be generated at numerous points.

The calculations are done using a modular program system composed of number of codes capable to do cross section preparation, flux calculations and evaluations, burn-up and depletion calculation in terms of time and reactor power.

Such models, even the most sophisticated, which must take into account the half-life, yield and nuclear transformation due to the decay to other nuclides during the reactor operation, cannot give in spite of the very important and substantial progress in this area a definite cross section values due to the complexity of the problem originated practically by the complicated process of fission.

Fission Product Production

Over the years, a great effort has been devoted to the evaluation of major isotopes pertinent to reactor application including particularly the uranium and plutonium isotopes, however, the effort to evaluate fission product data are less than the effort given for the actinide data.

The nuclear fission of a fissile material, as uranium or plutonium, is a complicated process in which more than 500 different products of radio nuclides of about 40 elements of the periodic table (ranged from Z=31 to 68) are produced. Most of these fission products may be formed in different ways, as a primary event or as they have extra neutrons, they tend to decay to more stable isotopes through beta emission constituting the fission chains. Each fission chain is formed of a certain number of fission products which are, in general, radioactive such as:

$$^{85}_{31}Ga \xrightarrow[0.09\,s]{} {}^{85}_{32}Ge \xrightarrow[0.54\,s]{} {}^{85}_{33}As \xrightarrow[2.03\,s]{} {}^{85}_{34}Se \xrightarrow[32\,s]{} {}^{85}_{35}Br \xrightarrow[2.9\,m]{} {}^{85}_{36}Kr \xrightarrow[10.7\,y]{} {}^{85}_{37}Rb\ (stable)$$

$$^{131}_{50}Sn \xrightarrow[61\,s]{} {}^{131}_{51}Sb \xrightarrow[23\,m]{} {}^{131}_{52}Te \xrightarrow[25\,m]{} {}^{131}_{53}I \xrightarrow[8.02\,d]{} {}^{131}_{54}Xe(m) \xrightarrow[11.9\,d]{} {}^{131}_{54}Xe\ (stable)$$

Fission products belonging to about 90 important mass chains [15], they are ranging from Nickel to Erbium. Only about 25% of them are creating in both ground and isomeric states. Besides this, but with a very little probability another contribution comes from light fission products (tritium, helium) generated in the ternary nuclear fission.

The description of the formation of fission fragment in the nucleon-induced fission reactions can be explained by the following two main subjects. The first one lies in the definition of the reaction mechanism of all fission stages which is depending on the beam energy .The second is the proper model of fragment formation. Fission transmutation reactions produce mostly short lived fission products that decay into stable element .The following example of plutonium fission shows the production of two short –lived fission products, tellurium and molybdenum. They both undergo a series of beta decays. The decay chain of molybdenum-102 consists of short-lived radio nuclides until it reaches stable (non- radioactive) Ruthenium-102 while Tellurium, Fig (1), decays into long-lived cesium-135. [16]

There are 26 important fission products in LWR which account for about 80% of the fission product absorption in this type of power reactors. This number is determined in the base of their yield, half-life and absorption cross section.

The yields data are extremely important for the calculations of fission products accumulation and inventory at various stages of the nuclear fuel cycle and estimation of decay heat after reactor shutdown or in the spent fuel assemblies, while their half lives are particularly important for shielding problems, processing and extraction treatment and calculation of their quantities in terms of reactor working time. The types and yields of the fission products depend on many factors, but principally, on the type of the fuel element and the neutron energy inducing the fission. The fission products are usually classified according to their half-life or yields, table 1 represents some important fission products arranged which are arranged in descending order of their yields, while in table 2 some others are arranged according to their half-lives [17].

Results

From the large number of fission products and excluding all of those having half-lives in second or minute, we present twelve important fission products having the following characteristics:

Four of them are long lived, their half-life exceed one million year which are:
Europium-151, Neodymium-145, Samarium-147 and Technetium-99.
Six of them have a large cumulative yields, they are:
Cerium-141(5.72%), Cesium-133(6.79%), Cesium-137(6.21%),
Molybdenum-99(6.11%), Strontium-89(4.85%) and Zirconium-95(6.49%) .

The last two fission products are medium lived isotopes, they are:
Krypton-85(10.7y) and Promethium-147 (2.62y) .
Three of the selected fission products have acceptable half-lives for measurement purpose which are Ce-141(32.4d), Sr-89(47.51d) and Mo-99(66.02h), the others have a very large half-life.
In order to obtain an average value for the different libraries data we applied the following weighted mean formula [18] ,

$$Y = \frac{\sum w_i \, y_i}{\sum w_i}$$

Where: $w_i = 1/\sigma_i^2$

σ_i =standard deviation of sample i

y_i = cross section value of sample i

Some libraries declare the uncertainty of calculation which we considered; otherwise we used 10% for the unmentioned error.
Basing on the above formula, we apply the calculation using Matlab.
Different sets of programs are built depending on the number of data used
(From 3 to 7 data), the energy interval considered is from 0.00001 to 15 MeV.
Results of 12 fission products (from different libraries and evaluated data) are presented in graphs as shown in figures (2-13).

Conclusion

The evaluated absorption cross sections of twelve important fission products are presented in figures (2-13).The evaluation is based on the main international data libraries. The average weighted values indicate clearly the necessity to adopt such calculations which are very important for some problems such as neutron dosimetry, fuel burn-up and determination of isotope production.

A complete dependency on individual or even collective experimental results are not recommended due to experimental deviations and errors and the impossibility of measurement for all the detailed interval of energy .This may be one of the important reasons to explain the observed deviations in some energy intervals of the international libraries. Thus using the weighted average one can obtain an accurate, complete and energy detailed cross section values and can implement the essential condensation calculation in terms of any energy interval which are essential for all kinds of reactor calculations

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Table(1): Some fission products ordered by yield produced by thermal neutron fission of U-235 [17]

Nuclide	T ½	Yield %	Nuclide	T ½	Yield %
Cs-133	2.06 y	6.79	Nd-145	stable	2.24
Xe-135	9.1 h	6.54	Sm-147	stable	2.24
Zr-95	64 d	6.50	Sm-149	$1.0\ 10^{16}$ y	1.08
Zr-93	$1.5\ 10^{6}$ y	6.35	Eu-151	Stable	$4.19\ 10^{1}$
I-135	6.57 h	6.28	Sm-151	90 y	$4.19\ 10^{1}$
Cs-137	30.17 y	6.21	Ru-106	1.02 y	$4.02\ 10^{1}$
Te-99	$2.1\ 10^{5}$ y	6.11	Kr-85	10.7 y	$2.83\ 10^{1}$
Mo-99	66 h	6.11	Se-79	$6.5\ 10^{4}$ y	$4.47\ 10^{2}$
Ce-141	32.4 d	5.85	Eu-155	4.71 y	$3.21\ 10^{2}$
Sr-90	29.1 y	5.78	Gd-155	Stable	$3.21\ 10^{2}$
SR-89	47.51 d	4.85	Sb-125	2.76 y	$3.40\ 10^{2}$
I-131	8.04 d	2.89	Sn-126	$1.0\ 10^{5}$ y	$5.61\ 10^{2}$
Pm-147	2.62 y	2.25	Cd-113m	14.6 y	$1.66\ 10^{4}$

Table (2) : Some fission products ordered by half-life produced by thermal neutron fission of U-235 [17]

Nuclide	T ½	Yield %	Nuclide	T ½	Yield %
I-135	6.57 h	6.28	Sm-151	90 y	0.419
Xe-135	9.10 h	6.54	Cs-137	30.17 y	6.19
I-131	8.04 d	2.89	Se-79	$6.5\ 10^{4}$ y	$4.47\ 10^{2}$
Ru-106	1.02 y	0.40	Sn-126	$1.0\ 10^{5}$ y	$5.61\ 10^{2}$
Cs-133	2.06 y	6.79	Tc-99	$2.1\ 10^{5}$ y	6.11
Pm-147	2.62 y	2.25	Zr-93	$1.5\ 10^{6}$ y	6.35
Sb-125	2.76 y	$3.40\ 10^{2}$	Cs-135	$2.3\ 10^{6}$ y	6.54
Eu-155	4.71 y	$3.21\ 10^{2}$	In-115	$5.0\ 10^{14}$ y	$1.22\ 10^{2}$
Kr-85	10.7 y	$2.83\ 10^{1}$	Cd-115	$9.0\ 10^{15}$ y	$1.40\ 10^{2}$
Cd-113m	14.6 y	$1.66\ 10^{4}$	Sm-149	$1.0\ 10^{16}$ y	1.08
Sr-90	29.1 y	5.78	Gd-155	Stable	$3.21\ 10^{2}$

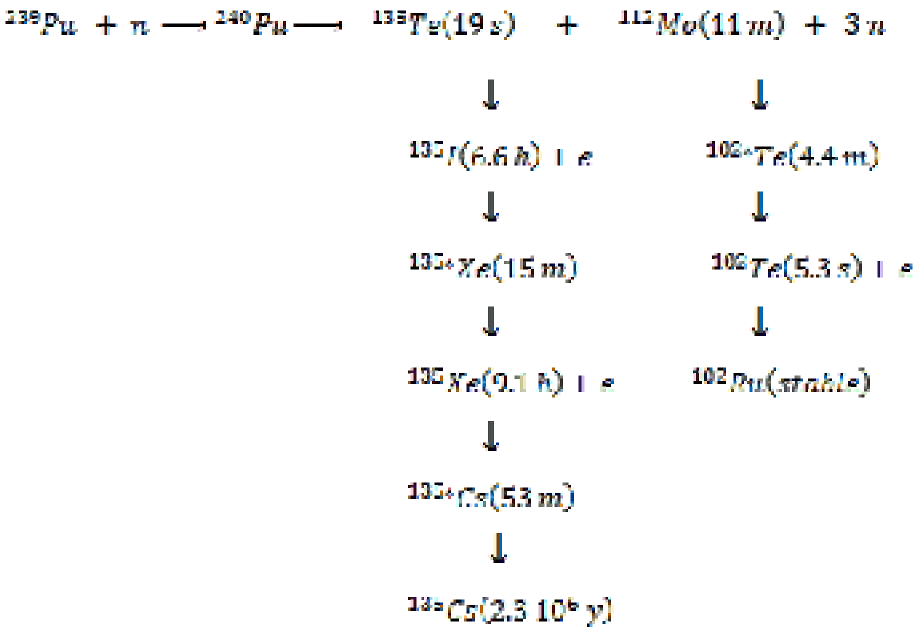

Fig. (1): Schematic diagram of a fission products chains of Plutonium [16]

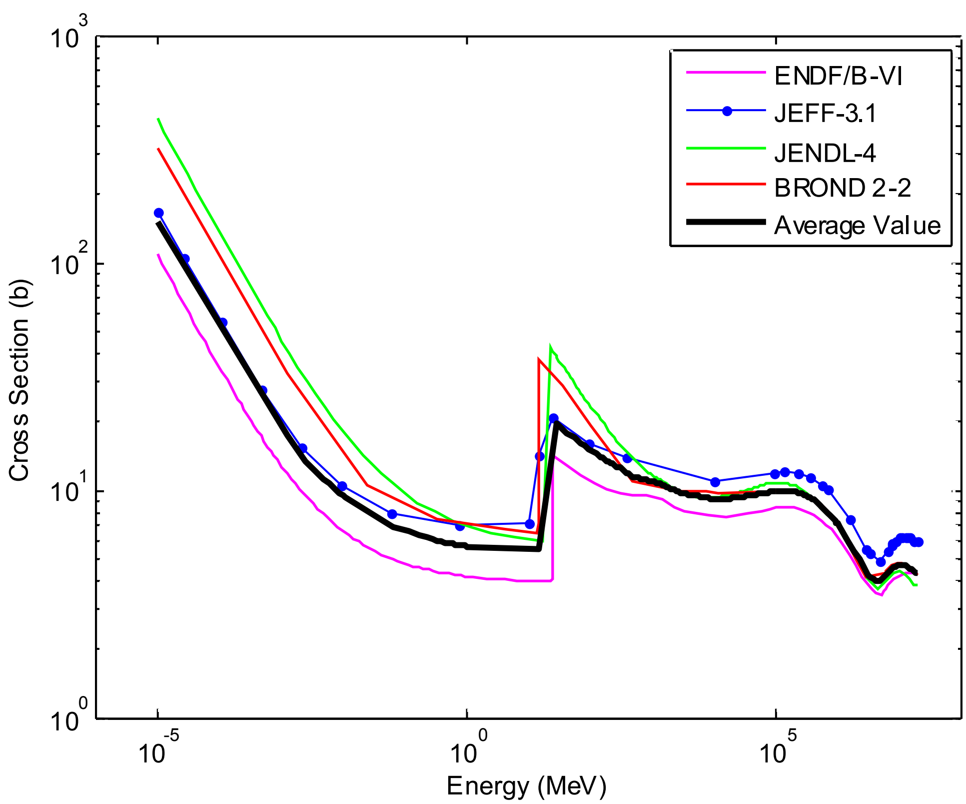

Fig.(2): Absorption Cross Section of Mo-99

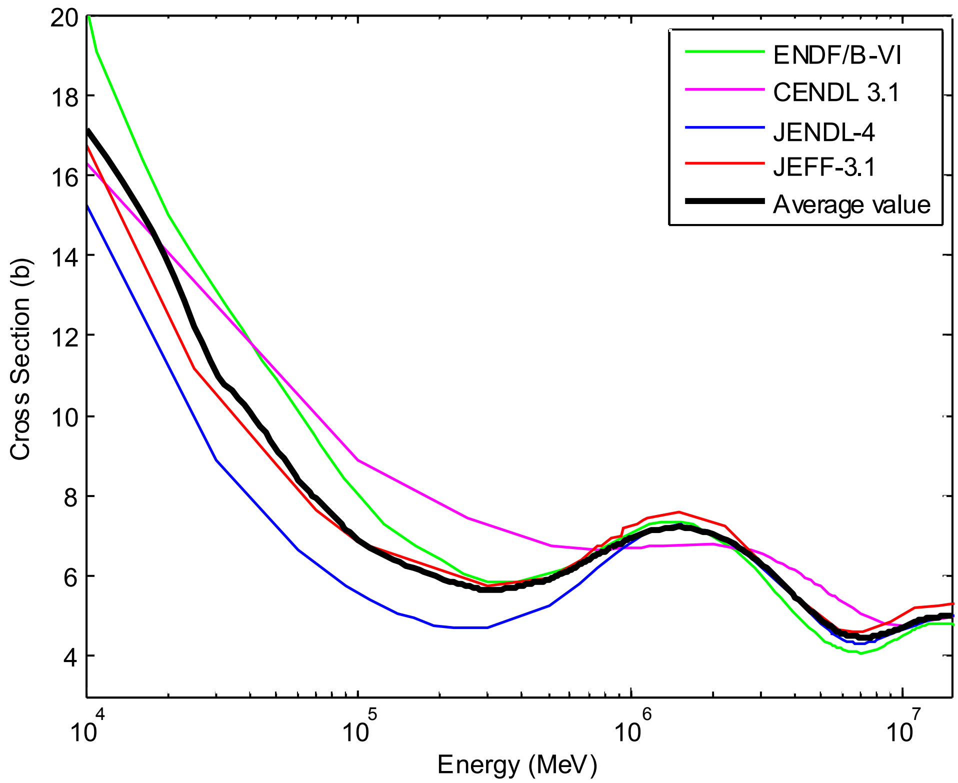

Fig.(3): Absorption Cross Section of Nd-145

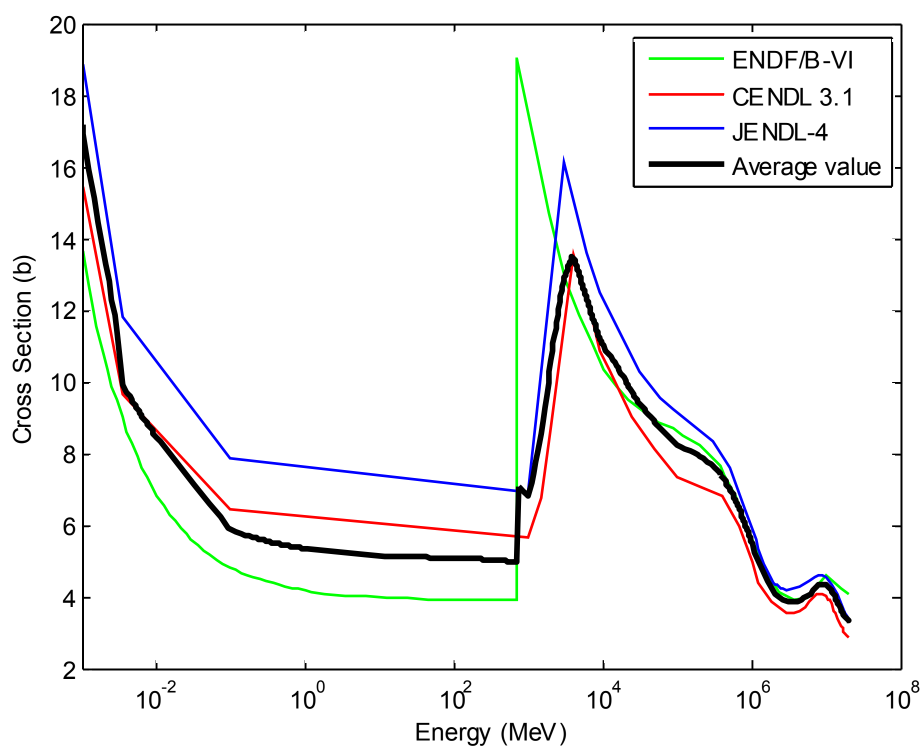

Fig.(4): Absorption Cross Section of Kr-85

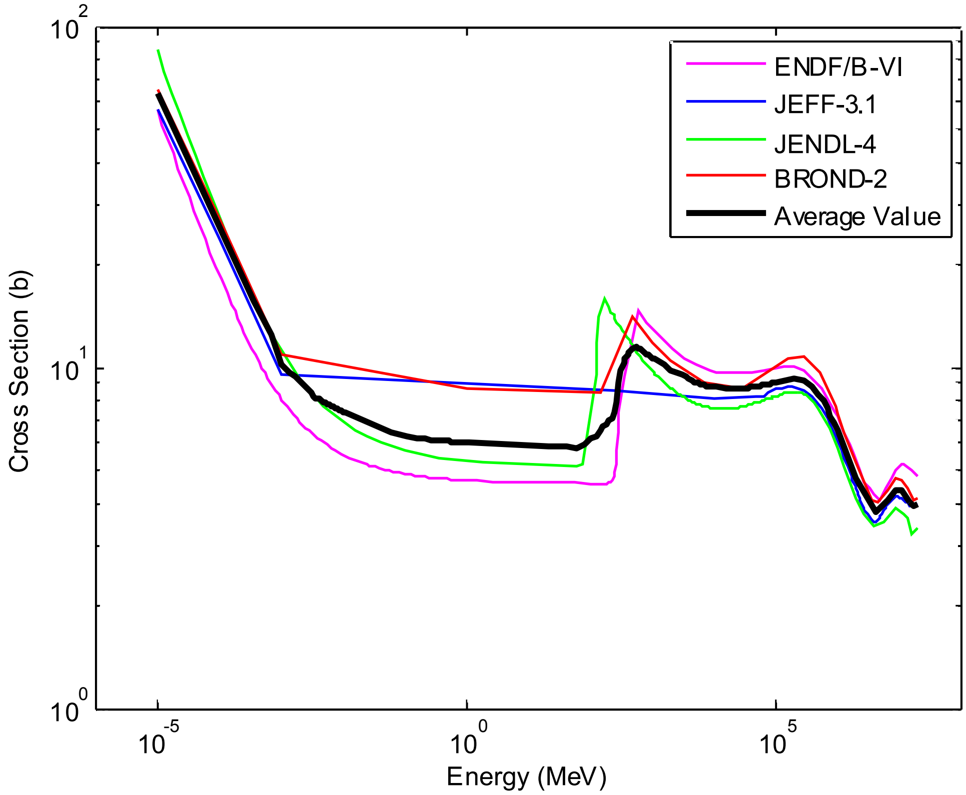

Fig.(5): Absorption Cross eSction of Zr-95

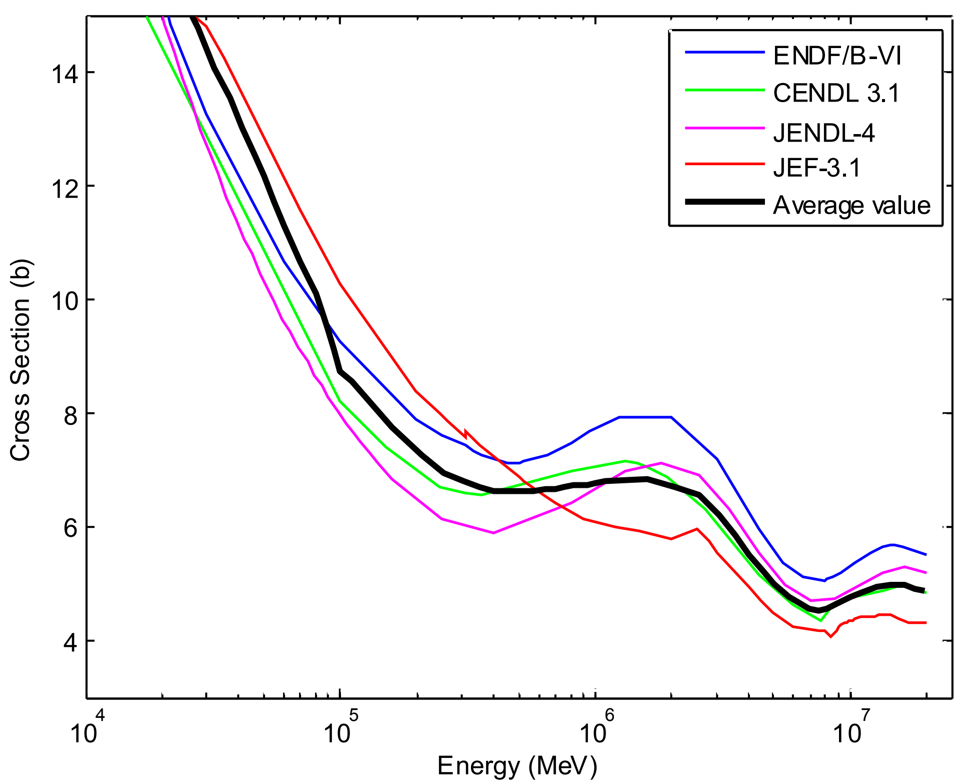

Fig.(6): Absorption Cross Section of Eu-151

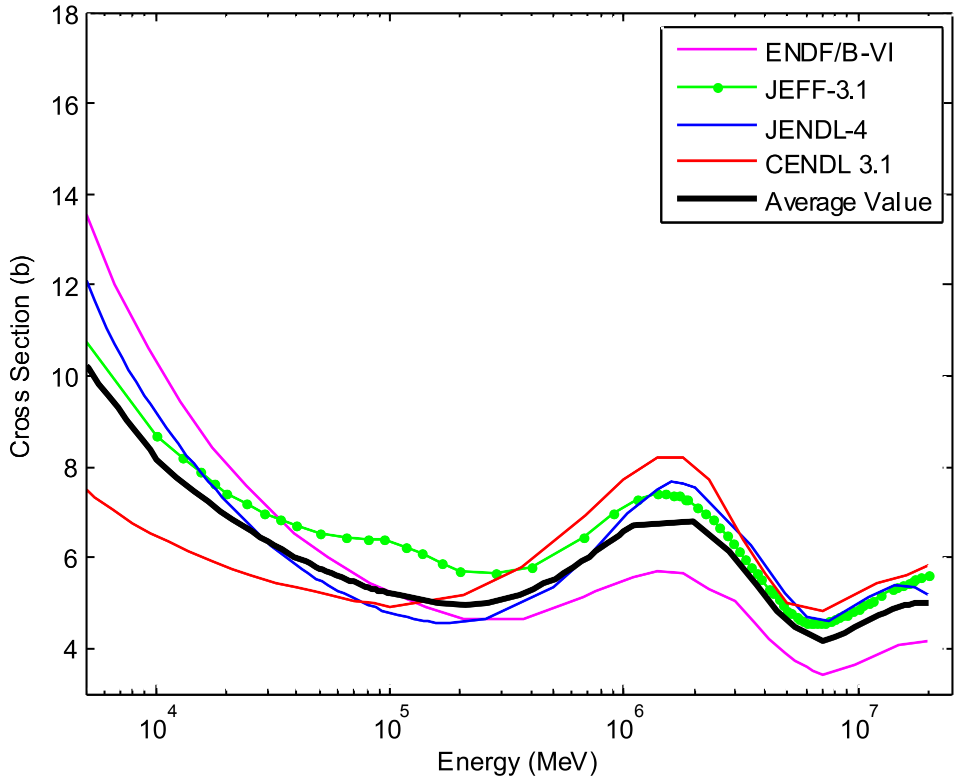

Fig.(7): Absorption Cross Section of Ce-141

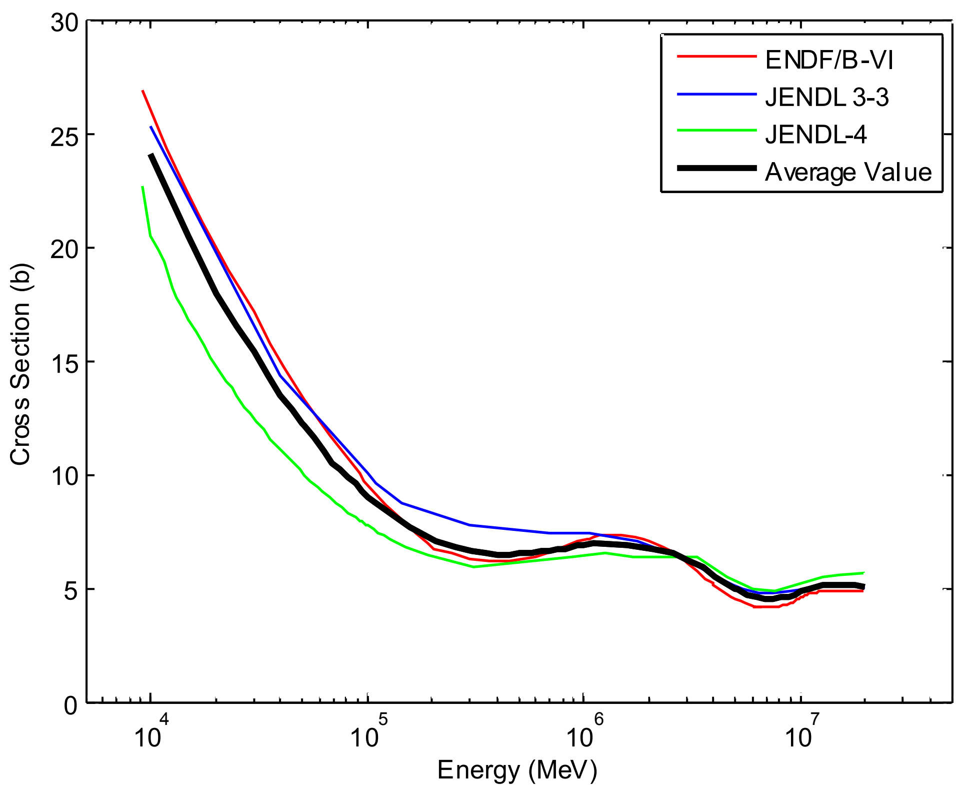

Fig.(8): Absorption Cross Section of Sm-147

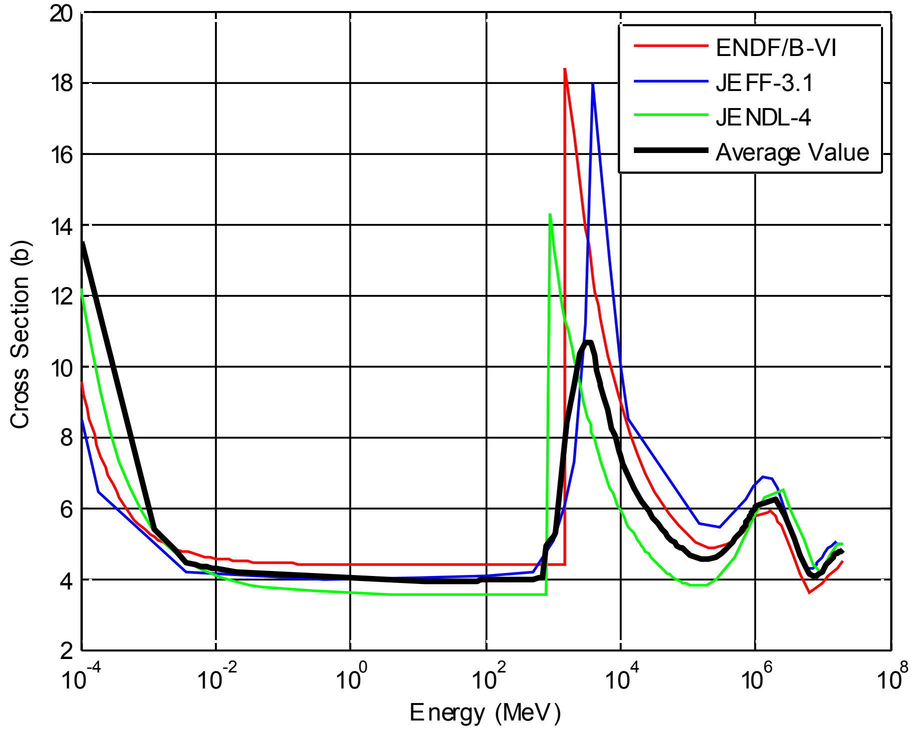

Fig.(9) : Absorption Cross Section of Cs-137

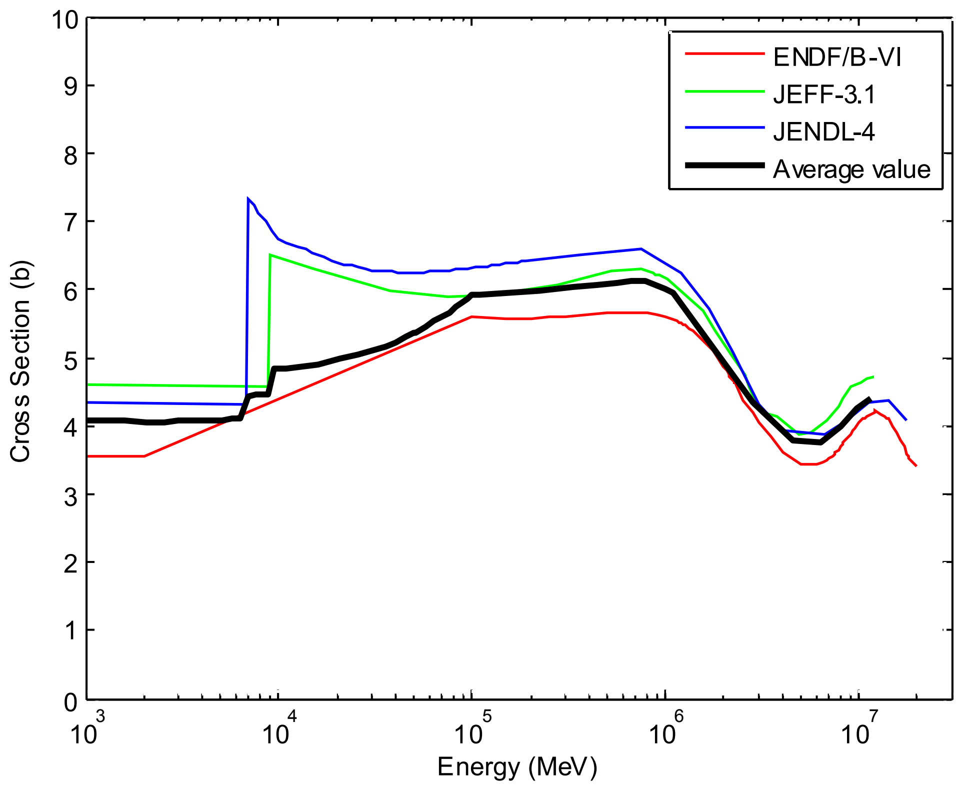

Fig.(10): Absorption Cross Section of Sn-126

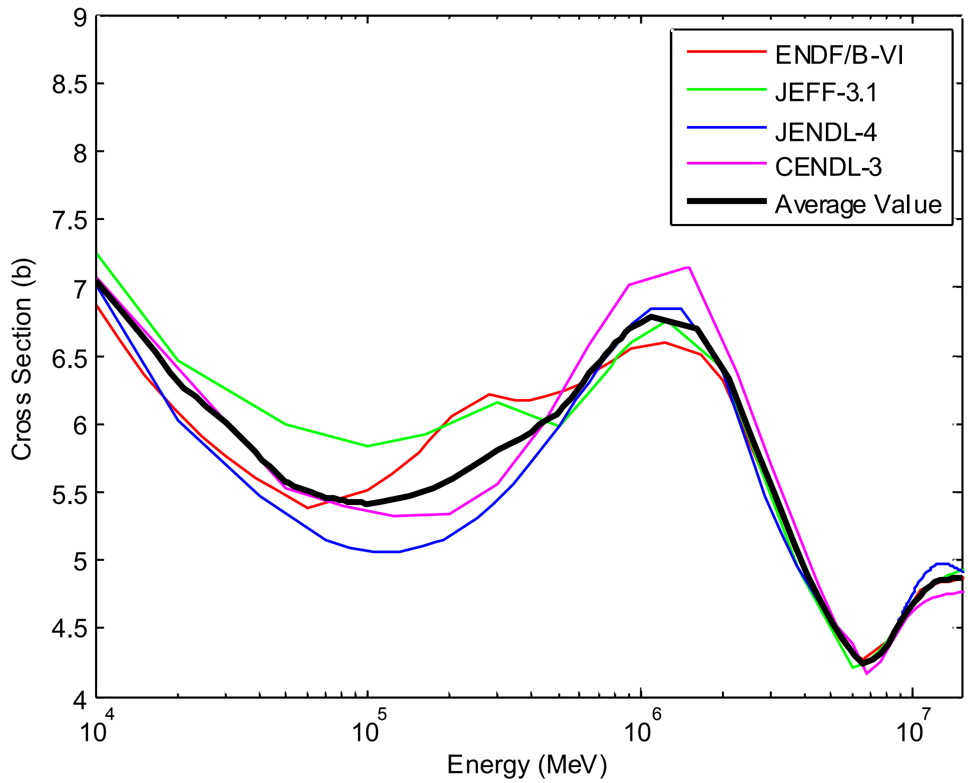

Fig.(11): Absorption Cross Section of Cs-133

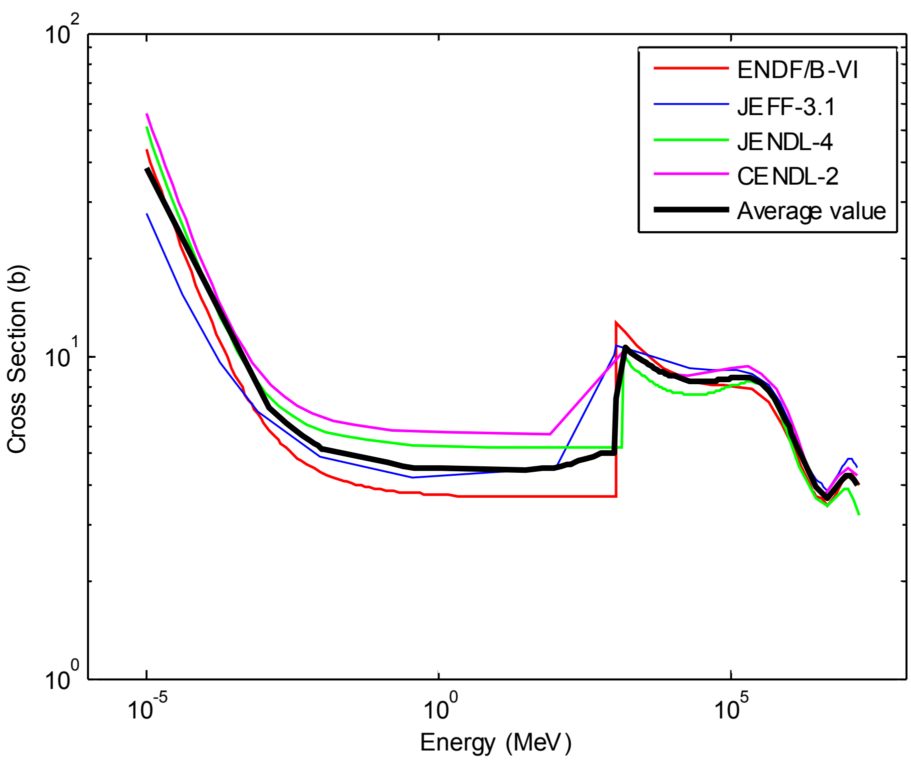

Fig.(12): Absorption Cross Section of Sr-89

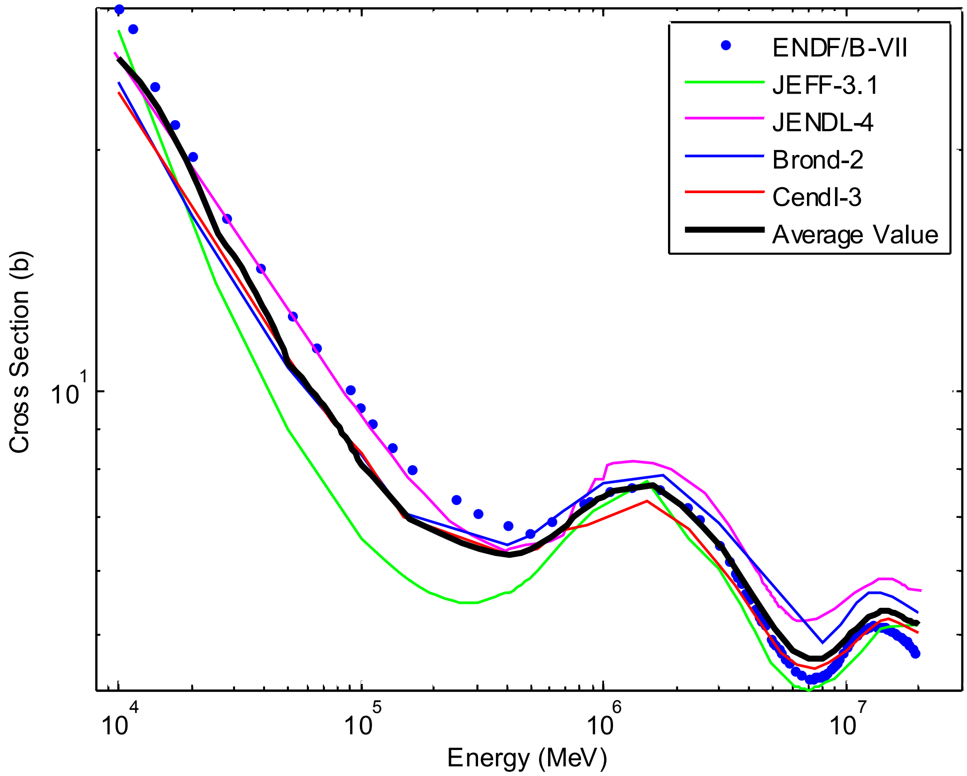

Fig.(13): Absorption Cross Section of Pm-147

تقييم المقاطع العرضية لنواتج الانشطار في المفاعلات الحرارية

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استلم البحث في: 2، كانون الاول ، 2010
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الخلاصة

يعد تاثير نواتج الانشطار من العوامل المؤثرة وبشكل كبير جدا على عمل المفاعل النووي، اذ تساهم مقاطع امتصاصها النووية في الحسابات المفاعلية .اجريت دراسة لقيم المقاطع العرضية النيوترونية لمجموعة من نواتج الانشطار المؤثرة في حسابات احتراق الوقود النووي بالاعتماد على اهم المكتبات العالمية المتخصصة بالمقاطع العرضية، وكذلك على احدث قيم التجارب الموثقة لدى الوكالة الدولية للطاقة الذرية .توضح نتائج حسابات قيم معدل المقاطع العرضية الموزونة اهمية اعتماد طريقة المعدل الحسابي الموزون في الحسابات النيوترونية لنواتج الانشطار.

الكلمات المفتاحية : مقاطع اقتناص عرضية ، مفاعلات نووية ، نواتج انشطار ، مكتبات دولية ، نسب انشطار نووية