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The TRADE Experiment: Importance of Neutron Cross-Sections for Transmutation

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Accurate neutron cross-section data is fundamental to the reliable design of any transmutation device, and, in particular, of an Accelerator-Driven System (ADS). Calculations of the behaviour of the core depend strongly on the cross-section data: parameters such as the multiplication coefficient, power densities or reactivity may vary significantly depending on the nuclear data (ND) library used. These potential discrepancies justify the need to improve the present data for several isotopes and reaction channels, for a wide range of neutron energies from thermal to high-energy.

The present work expands the analysis performed in the context of the n_TOF-ND-ADS project of the EURATOM 5th Framework Program, to other isotopes of interest such as ^{233}U, ^{243}Am, $^{244,\,245}$Cm and the long-lived fission fragments (LLFFs) ^{99}Tc and ^{129}I. A direct comparison of nuclear data libraries to indicate the spread between values was performed. The paper also extends the sensitivity analysis mentioned above to moderated systems, such as TRADE (Triga Accelerator-Driven Experiment): a 1 MW Triga Reactor coupled with a 140 MeV–2 mA proton cyclotron. Study of the discrepancies in the thermal and epithermal regions is essential for the design of systems for the transmutation of LLFF (transmutation by adiabatic resonance crossing, TARC) and also important for minor actinides (MAs) for which sub-threshold fission should not be neglected.

KEYWORDS: TRADE facility, ADS, n_TOF-ND-ADS project, Transmutation of LLFP and MA, TARC method, Nuclear data libraries.

1. Introduction

Accurate cross-section (XS) information is fundamental for reliable prediction of the behaviour of a nuclear reactor core. In particular, for Accelerator-Driven Systems the level of uncertainty in $\Delta\rho$/cycle is essential (e.g. at $k_{src} \approx 0.98 \pm 2\%$ uncertainty in $\Delta\rho$/cycle has serious consequences for safety analysis and accelerator current requirements). Thus, for a subcritical device, uncertainties in the nuclear data limit the safety margins within which such a device could operate. Hence, for $0.90 \leq k_{src} \leq 0.99$ the precision should be similar to that required for critical cores (±100 pcm for UO_2, ±40 pcm for MO_X). However, the various XS libraries show significant differences between their respective XS data, for a wide range of energies. The use of different libraries can cause significant deviations in the results of design calculations. This analysis aims to identify the isotopes and reaction channels responsible for the largest discrepancies in the main neutronic parameters.

A neutron XS sensitivity analysis [1] was previously performed using the EA-MC Monte

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Carlo code package [2] to study the Energy Amplifier Demonstration Facility [3,4] (EADF), an 80 MW_{th} Pb-Bi cooled reactor, comparing the results of calculations based on different nuclear data libraries, i.e. JAR-95 [5] (CERN), ENDF/B-VI (US), JENDL-3.2 (Japan). The results showed discrepancies in $\Delta k/k$ ranging from 500 to 2500 pcm for different types of fuels and ND libraries. Large neutron balance discrepancies for some reactions and isotopes (e.g. 25% for capture in ^{241}Pu, 16% for fission in ^{240}Pu) were discovered.

The release of new versions of the ND libraries (ENDF/B-VI.8, JEFF-3.0 and JENDL-3.3) led to this new study, extending the previous work to other isotopes of interest. In addition, progress in the design of TRADE [6] generated interest in studying the effects of XS data for thermal ADSs, thus widening the energy range examined in the study.

The present analysis was performed using ENDF/B-VI.8 as a reference library and systematically changing the ND library of a single isotope at a time, while keeping the ND intact for the rest of the isotope inventory. In this way, the effects of the discrepancies in the ND for every specific isotope were isolated, thus avoiding spectrum effects.

It should be emphasized that the results presented here show the relative discrepancies between libraries, and do not imply an assumption that one library is more accurate than another. The results simply estimate the level of uncertainty for every isotope and reaction.

2. The TRADE Project

TRADE is a pilot experiment, to provide a global demonstration of the ADS concept and based on an original idea of Carlo Rubbia. The experiment entails the coupling of an existing 1 MW Triga reactor to a 140 MeV, 2 mA proton cyclotron at ENEA Casaccia, Rome. A preliminary study of the experiment is presented in the Final Feasibility Report [6]. The reference configuration (Figures 1a and 1b) predicts a multiplication coefficient $k_{src} = 0.977$, a beam power $P_{beam} = 11$ kW and an energy gain $G = 19$, with a neutron yield for 140 MeV protons in tantalum of 0.8 n/p as shown in Table 1.

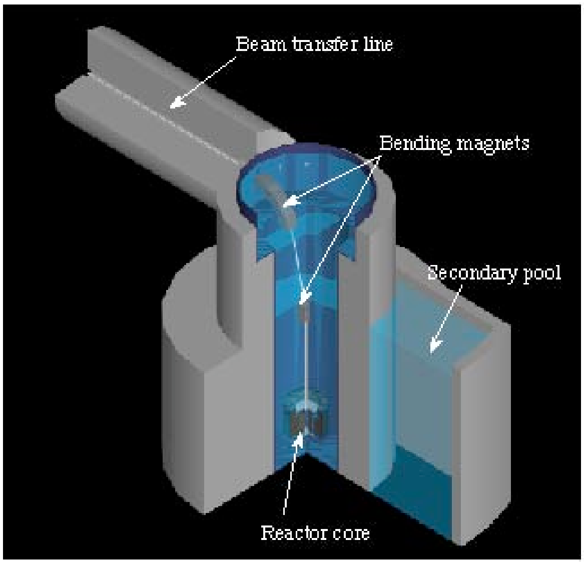

Fig.1a General view of the Monte Carlo of model of the TRADE project

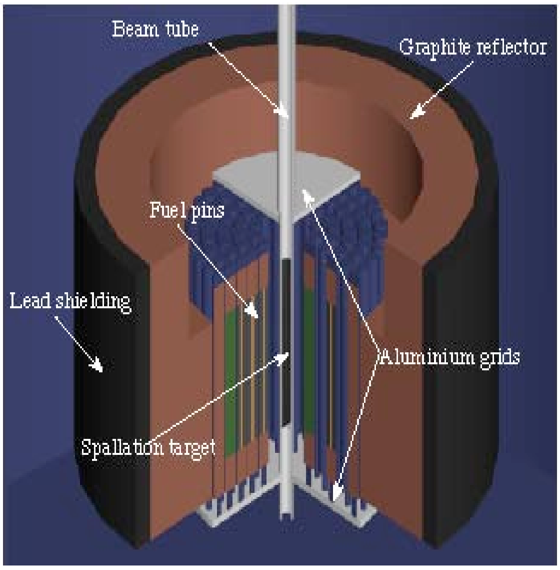

Fig.1b Close-up view of the Triga reactor core of TRADE

Table 1 Main reactor parameters in TRADE for 200 kW of thermal power output

Global parameters	Symbol	Reference case	Low k_s	High k_s	Critical
Proton beam energy	E_p (MeV)	140			-
Spallation neutron yield	N (n/p)	0.75			
Multiplication coefficient	k_s	0.977	0.903	0.988	1.000
Energy gain	G	18.8	4.0	28.1	-
Accelerator current	I_p (mA)	0.08	0.61	0.07	
Beam power	P_{beam} (kW)	11.2	66.9	7.4	
Core Power Distributions					
Av. fuel power density	P_{th}/V_{fuel} (W/cm^3)	4.9	6.5	4.7	4.8
Maximum linear power	P_l(W/cm)	68.0	118.2	70.5	70.2
Radial peaking factor	P_{max}/P_{ave}	1.30	1.76	1.46	1.45
Linear peaking factor	P_{max}/P_{ave}	1.51	1.78	1.48	1.45

The EA-MC code allows for the straightforward production of neutron flux spectra at selected locations. Figure 2 shows some spectra, which allow further insight into the neutronic characteristics of the device. Note the uniform distributions in the different fuel regions (ring C to G).

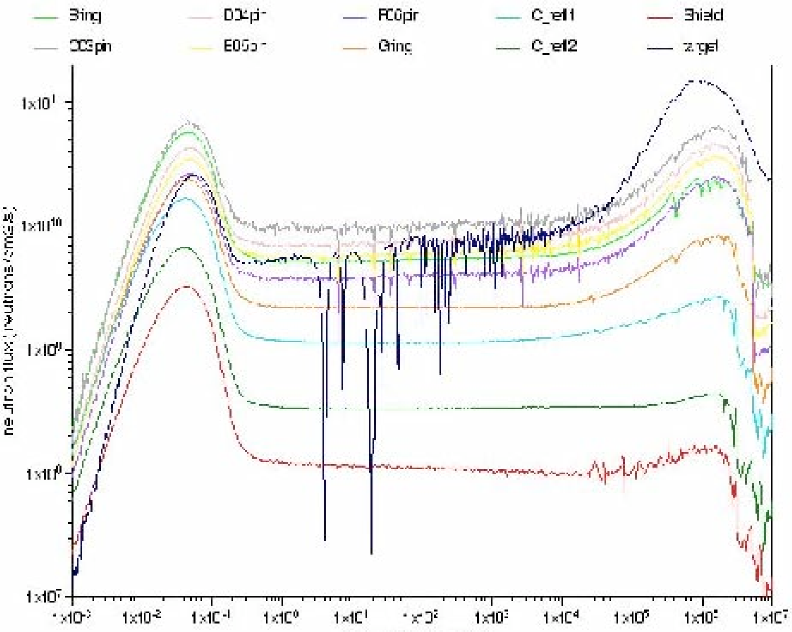

Fig.2 Neutron energy spectra in different positions of the core of TRADE. From the center of the core to the periphery: target, rings B-C-D-E-F-G, carbon reflectors and lead shielding

3. Sensitivity Analysis for Different Neutron Cross-Section Data Libraries

The effects of the different cross-sections of ^{235}U and ^{238}U were considered first, together with a global analysis of the different ND libraries. Table 2 presents the results of this analysis. Substantial discrepancies for ^{235}U between ENDF and JENDL can be observed. When different libraries are used for all the isotopes, large discrepancies on the reactivity appear between ENDF and JEFF, well above the requested limits reported in Introduction.

Table 2 Neutron XS data effects on k_{src} for TRADE

Isotopes		ENDF/B-VI.8	JEFF-3.0	JENDL-3.3
U-235	k_{src}	0.97389	0.97362	0.97201
	error	±0.00029	±0.00018	±0.00017
	Δk/k (pcm)		–28	**–193**
U-238	k_{src}	0.97389	0.97391	0.97387
	error	±0.00029	±0.00021	±0.00020
	Δk/k (pcm)		2	–2
Zr-90	k_{src}	0.97349	0.97521	0.97572
	error	±0.00020	±0.00020	±0.00021
	Δk/k (pcm)		177	**229**
All isotopes	k_{src}	0.97389	0.97615	0.97454
	error	±0.00029	±0.00022	±0.00024
	Δk/k (pcm)		**232**	**67**

No major differences between ENDF/B-VI.8 and JENDL-3.3 were observed comparing the capture and fission reaction rates of ^{235}U as a function of neutron energy, except for a slightly higher capture rate in the 10 – 300 keV region in the case of JENDL (Figure 3). On the other hand, a slight discrepancy in the MeV region can be observed comparing the elastic (n,ne) and the inelastic (n,ni) scattering reaction rates (Figure 4): JENDL-3.3 predicts more elastic and less inelastic scattering. This divergence shifts the spectrum of the neutrons, producing more moderation in the case of ENDF/B-VI.8, increasing the resonance escape probability and effectively increasing k_{src}.

The analyses using different ND libraries for all isotopes show contrasting results. JEFF-3.0 gives discrepancies with ENDF/B-VI.8 (232 pcm) well above the statistical error, whereas JENDL-3.3 is in good agreement with the latter. As Table 2 shows, this change is mainly due to the lower capture rates in ^{90}Zr using JEFF-3.0 and JENDL-3.3, and highlights the need for single-isotope analysis in order to avoid compensation effects which would otherwise hide uncertainties for particular isotopes.

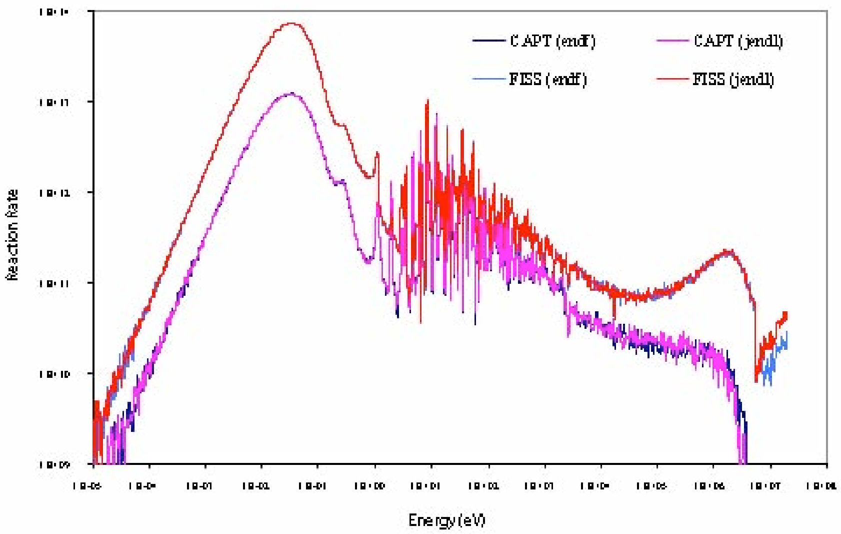

Fig.3 U-235 capture and fission rates using ENDF/B-VI.8 and JENDL-3.3

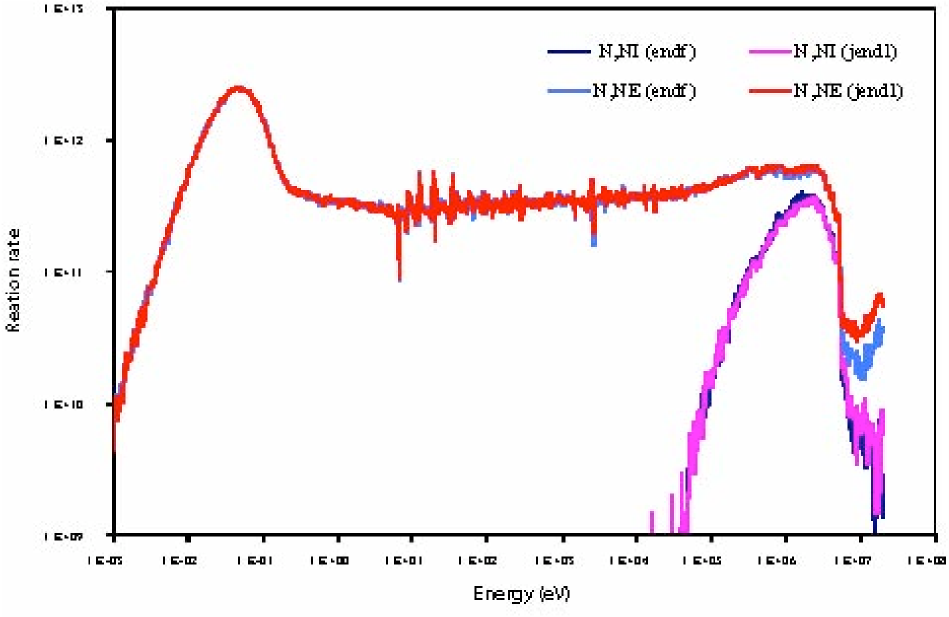

Fig.4 U-235 (n,ne) and (n,ni) reaction rates using ENDF/B-VI.8 and JENDL-3.3

The sensitivity study to nuclear data was also applied to the most important isotopes for waste transmutation. Table 3 reports the main discrepancies in reaction rates for these isotopes and for the main reaction channels.

Table 3 Main discrepancies in the reaction rates for several isotopes

Isotope	Reaction	Discrepancy	Library	Isotope	Reaction	Discrepancy	Library
Zr-90	Capture	-15.1%	JENDL	Zr-90	Elastic scat	-4.7%	JEFF
Tc-99	Capture	+3.4%	JENDL	Tc-99	Elastic scat.	+24.5%	JEFF
I-129	Capture	-7.5%	JENDL	I-129	Inelastic scat.	+12.4%	JEFF
U-233	Inelastic scat.	+33%	Both	U-235	Inelastic scat.	-7%	JENDL
Pu-238	Fission	+11%	JEFF	Pu-238	Inelastic scat.	+38%	JENDL
Pu-240	Elastic scat.	+9%	JENDL	Pu-240	Inelastic scat.	+18%	JEFF
Pu-241	Elastic scat.	+13%	JENDL	Pu-241	Inelastic scat.	-56%	Both
Pu-242	Fission	+5%	JEFF	Pu-242	Inelastic scat.	+21%	JENDL
Am-241	Fission	+9%	JENDL	Am-241	Inelastic scat.	-19%	JENDL
Am-243	Fission	+5%	JEFF	Am-243	Elastic scat.	-27%	JEFF
Cm-244	Capture	+12%	JENDL	Cm-244	Inelastic scat.	-39%	JENDL

These results show discrepancies in the neutron absorption reactions (capture, fission and n,xn) of approximately 10% for several important isotopes (i.e. ^{232}Th, ^{238}Pu, ^{241}Am), which could have an adverse effect on the neutron multiplication of the system. Major differences are also observed in elastic and inelastic scattering reactions for many of the isotopes studied as illustrated in Figures 5.

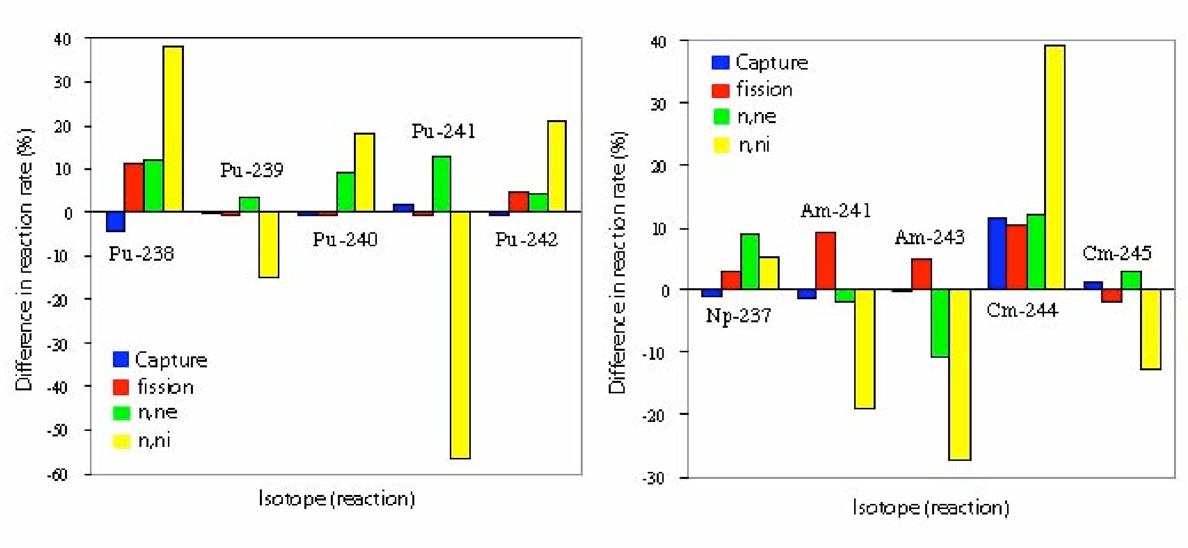

Fig.5 Reaction rate discrepancies for the Pu isotopes (left) and the Minor Actinides (right)

These discrepancies have a direct effect on the neutron energy spectrum (Figure 6) and consequently on k_{src}. The results for ^{90}Zr highlight the importance of the sometimes neglected need for accurate cross section data for structural materials.

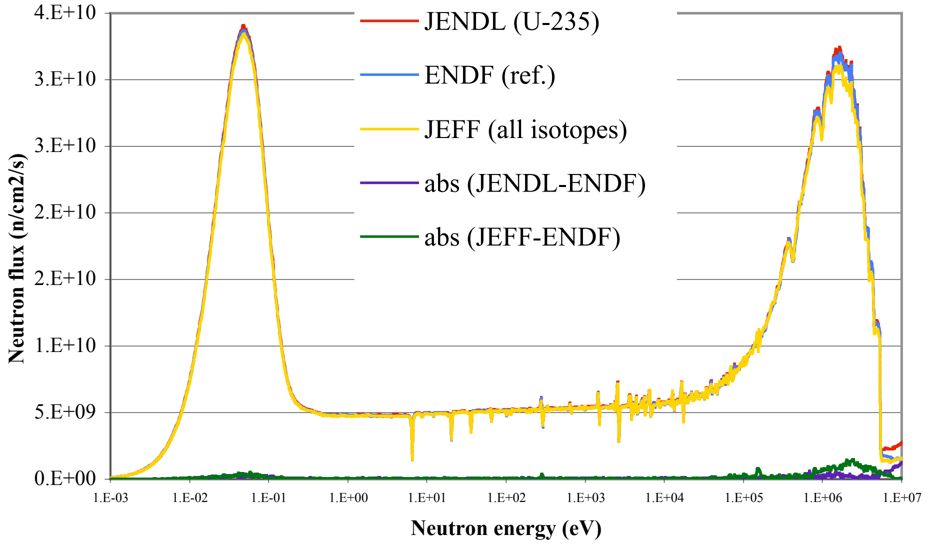

Fig.6 Neutron energy spectra in the TRADE fuel core for different ND libraries

4. Conclusions

The effect of the ND has been found to be of major importance for a reliable design of an ADS, especially in terms of safety. Discrepancies appear larger in fast systems, even though large uncertainties were found at thermal energies for isotopes such as ^{241}Pu and ^{244}Cm.

Discrepancies in the ND for coolant and structural materials, such as ^{90}Zr in the case of TRADE, and ^{207}Pb and ^{209}Bi in the case of a fast ADS, also have an important effect on the neutron multiplication of the system, which should not be neglected.

Large uncertainties in the neutron balance for isotopes such ^{237}Np, ^{238}Pu and ^{241}Am might have large effects on k_{src}, depending of the configuration of the fuel. Moreover, even small differences in the neutron balance for ^{233}U and ^{232}Th introduce large changes in the neutron multiplication. There is also a general need to improve the scattering cross sections, and inelastic in particular, for most of the isotopes; these uncertainties perturb the spectrum, therefore affecting the transmutation rate and the reactor behavior.

No major improvements with the new library releases were found, with the exception of capture in ^{237}Np and ^{240}Pu, and fission in ^{240}Pu. This demonstrates the need for the research carried out in experiments such as n_TOF7), where cross section measurements for many of these isotopes are being taken for a large energy range. This should greatly reduce uncertainties in the calculations of the different reactor parameters.

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