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# Nuclear Data Validation and Re-evaluation by Means of Integral Experiments

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Integral experiments are a rich source of information with which a wide range of validation and comparison exercises can be made in the activation data field. Having access to a variety of sources allows the energy dependence of the analysis to be more complete. Materials samples were irradiated in a wide range of simulated fusion D-T neutron fields at JAERI FNS, Sergiev Posad SNEG-13, FZK Isochron-cyclotron and ENEA FNG and different responses such as neutron spectra, emission spectra, kerma, radioactivity and decay heat have been measured, allowing direct comparison with the code predictions. The results of the different validation exercises, when correlated with other sources of information, led to a set of corrective measures to be taken on some important reactions and these were implemented on the European Activation File EAF-99. These include cross section renormalisation, profile correction in a given energy range, threshold and Q-value correction, partial cross section adjustment and even new energy-dependent isomeric branching ratios.

***KEYWORDS: Nuclear data, evaluation, neutron cross section, excitation function, threshold reaction, energy range $10^{-5}$ eV to 20 MeV, branching ratio***

## I. Introduction

Integral experiments are a rich source of information with which a wide range of validation and comparison exercises can be made in the activation data field. The broad trends of the time dependent values of the calculated-to-experimental ratios represent the best estimates of the uncertainties that need to be applied to predictions made with activation codes.

Validation of code predictions by means of direct comparison with integral data measurements allows not only the calculational method to be certified but also generates the type of information that is necessary to indicate any inaccuracy or omission that may exist in the nuclear data base. Materials samples were irradiated in a wide range of simulated D-T neutron fields at JAERI FNS, Sergiev Posad SNEG-13, FZK Isochron-cyclotron and ENEA FNG and different responses such as neutron spectra, emission spectra, kerma, radioactivity and decay heat have been measured, allowing direct comparison with the code predictions.

The results of the different validation exercises, when correlated with other sources of information, led to a set of corrective measures to be taken on some important reactions and these were implemented on the European Activation File EAF-99 (1). These include cross section renormalisation, profile correction in a given energy range, threshold and Q-value correction, partial cross section adjustment and even new energy-dependent isomeric branching ratios. The fact that some of these benchmarks were performed by several research institutions using different activation codes and decay data has allowed clarification of the reasons for certain discrepancies. These have been found to be partly due to innaccurate decay data, partly to inaccurate cross section data, and partly to a variety of inadequate calculational processes.

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## II. Integral experiments

Four different experimental set-up have been used for this validation work. The experiments themselves have been carried out in the EU, RF and Japan during the past years in laboratories with different neutron sources, irradiation and measurements methods. Having access to a variety of sources and samples has allowed the analysis to be very broad.

### 1. ENEA FNG

In the Frascati Neutron Generator (FNG) a deuterium beam is accelerated and focussed on a fixed tritiated titanium target to produced nearly isotropic 14 MeV neutron through $T(d,n)^4He$ reactions. The ion beam, produced by a Doplasmatron, deflected by a 90° bending magnet, is accelerated by a three gap uniform gradient linac accelerator. The beam is focused by means of a magnetic triplet quadrupole onto the target situated nearly 2 metres away from the tube. The deuteron beam has an energy of 260 KeV and a current of 1 mA. The neutron intensity reaches $1.0\text{x}10^{11}$ n/s with a maximum of $1.0\text{x}10^{10}$ n/cm$^2$.s on the sample. The tritium content in the target is about 370 Gbq producing a target lifetime of around 10 hours.

### 2. FZK Isochron-cyclotron

The Karlsruhe Isochron-cyclotron produces a 19 MeV deuteron beam using constant radio frequency with a shaped magnetic field. The target is fixed, water cooled and made of beryllium. The deuteron current can be up to 15 μA leading to a neutron flux of $1.5\text{x}10^{10}$ n/cm$^2$.s on a sample of 1 cm$^2$. Higher values can be achieved with smaller sample sizes. The neutron energy, due to deuteron stripping can reache 22-23 MeV but with a very low yield above 19 MeV.

The neutron energy spectrum is continuous, white, and differs from that due to a pure 14 MeV generator in ways that broaden the energy base for validation.

**3. Sergiev Posad SNEG-13**

At Sergiev Posad a 300 KeV electrostatic accelerator delivers a beam of deuterons, up to 300 mA, on a large tritium bearing titanium rotating target. Two sample positions at angles of 4° and 73° with respect to the deuteron beam have been used. The neutron flux density in the sample reaches $1.0 \times 10^{13}$ n/s with highly peaked mean energies of 14.37 and 14.93 MeV respectively. Irradiation times in such an experiment have been routinely up to several days, allowing the build-up of long lived isotopes, and so increasing the count rates.

**4. JAERI FNS**

14 MeV neutrons are generated by a 2 mA, 350 KeV deuteron beam impinging on a stationary tritium-bearing titanium target. The total neutron flux at the sample location, can be up to $3.0\ 10^{10}$ n/cm$^2$.s, the same order of magnitude as in the first wall of the JET tokamak when operating successfully with D-T plasma. Sample chamber positions at angles of 0° and 80° with respect to the deuteron beam could be used. Earlier experiments using a rotating target increased by an order of magnitude the neutron yield.

The major advantage of this experimental set-up lies in the development and implementation on site of new measurement techniques with high sensitivity and rapid response. The decay energy in an irradiated thin sample was measured in the Whole Energy Absorption Spectrometer (WEAS) which comprises two large bismuth-germanate BGO scintillators in a geometric arrangement which provides almost 100% detection efficiency for both β and γ-rays. Using the highly sensitive WEAS method, both β and γ-rays decay energies were measured at selected cooling times as early as one minute after the irradiation ended. The overall experimental uncertainty totals between 6 to 10%. The WEAS provides high sensitivity, less than 1 pW, which is valuable for measurement of some nuclides with long half-lives. It also has a wide dynamic range: measurements of up to a few μW have been achieved.

For both FNS and FNG experiments broader, more thermalised neutron spectra have been obtained when the target and sample areas have been surrounded by different types of structure. These mock-up assemblies are there to better simulate plant conditions and the resultant neutron spectra.

## III. Benchmarking

Much of the high energy nuclear data is based on calculations, relying on model codes to predict the excitation function. Although a certain number of measurements have been made and used, the calculations of activation characteristics require comparisons of such calculations with experimental measurement to be made.

Activation calculations are performed by computer codes which solve the large number of coupled differential equations (the Bateman equations) which govern the generation and decay chains for the many nuclides involved. They rely on a large volume of nuclear data, both neutron induced cross sections and radioactive decay data. These include energy-dependent partial reaction cross sections for all neutron reaction in a comprehensive range of nuclides, together with isomer ratios in the product nucleus, as well as all nuclear half-lives and decay branching ratios.

The European Activation System, EASY, has been used to perform these validation exercises. Two cross section data bases have been accessed using the 97 version of the FISPACT code [(2)]: EAF-97 [(3)] and FENDL/A-2.0 [(4)]. The decay data libraries used with the two cross sections libraries are also different: the EAF-97 decay data and the FENDL/D-2.0 data respectively. In order not to bias the experimental spectral data, the groupwise libraries used in the calculational scheme both correspond to a 175 Vitamin-J groups structure collapsed using a flat micro flux weighting function.

The results of the different validation exercises, combined with other sources of information, led to a set of corrective actions to be carried out on some important reactions. These were implemented in EAF-99. The fact that these benchmarks were performed by several research institutions using different activation codes and decay data has allowed clarification of the reasons for many discrepancies.

## IV. Nuclear data validation and re-evaluation

From such validation exercises much information can be extracted and conclusions drawn. However, the special features of each require caution to be applied when drawing or projecting conclusions from the results.

The general trend of the C/E or (C-E)/E ratios of a particular response indicate the correctness of the calculation. It is important to keep in mind the inevitable experimental uncertainty bound while assessing the agreement. The time dependence of a specific response indicates the consistency of half-life between measurement and calculation. The time dependence of the comparison has to be clearly established and it was not surprising to see some fluctuations in the degree of agreement with time. This is due to the fact that the set of predomi-

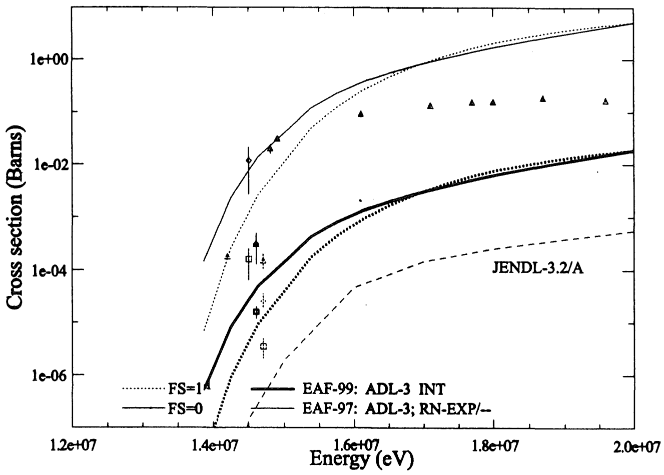


**Fig. 1** Ca-40(n,t): K-38 (7.6 m), K-38m(924.0 ms).
Experimental information on CaO sample indicates a gross overestimation of the production of K-38. The rather scattered EXFOR data is in the threshold range and JENDL-3.2/A does not contain partial channels. ADL-3 with corrected interpolation law is selected.

nant radionuclides evolves with time in direct relation to their half-lives.

For all these response it is evident that the set of dominant isotopes needs to be known. Experimental measurements rely generally on gamma energy line counts to detect the isotope types while the calculations rely on pathways analysis.

The results of all the different validation exercise [(5-13)], when correlated with other sources of information (i.e. EXFOR or new data measurements), led to a set of corrective measures to be taken on some important reactions. Corrective actions included cross section renormalisation, shape correction in a given energy range, threshold and Q-value correction, partial cross section adjustment and some new energy-dependent isomeric branching ratios. A few graphical examples are given below, with details in the captions.

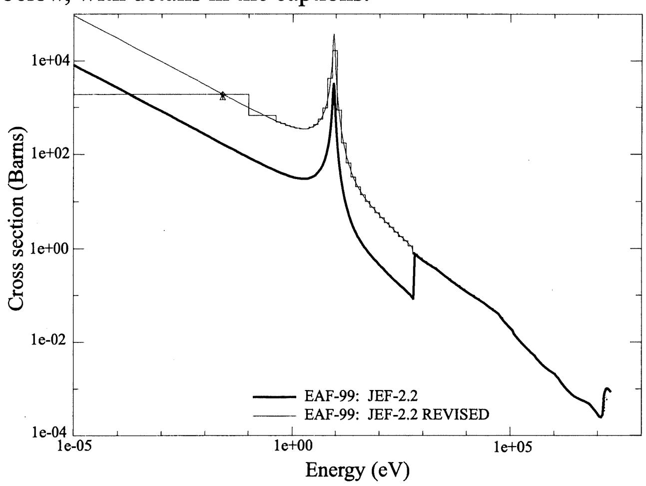


**Fig. 2** Co-58 (70.8 d) (n,g): Co-59.
High depletion rate of Co-58 in irradiated steels gave some reasons for concern. Little experimental data exist on this radioisotope but resonance integral information is available. Both (n,p) and (n,$\gamma$) depletion channels exist in the thermal range and the measured RI includes both. The (n,$\gamma$) excitation function has been renormalised in order to respect the original evaluator's splitting of 90% (n,p) and 10 % (n,$\gamma$) while the total agrees with the experimental RI.

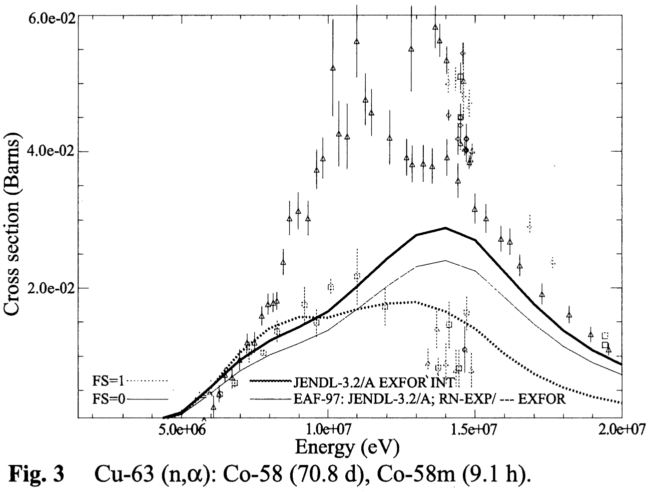


**Fig. 3** Cu-63 (n,$\alpha$): Co-58 (70.8 d), Co-58m (9.1 h).
A new measurement of the total cross section at 14.5 MeV suggest a value of 42 mb. Time dependent C/E of Cu sample indicates an underestimation after 6 days cooling. Only the ground excitation function has been renormalised in order to give a total of 42 mb.

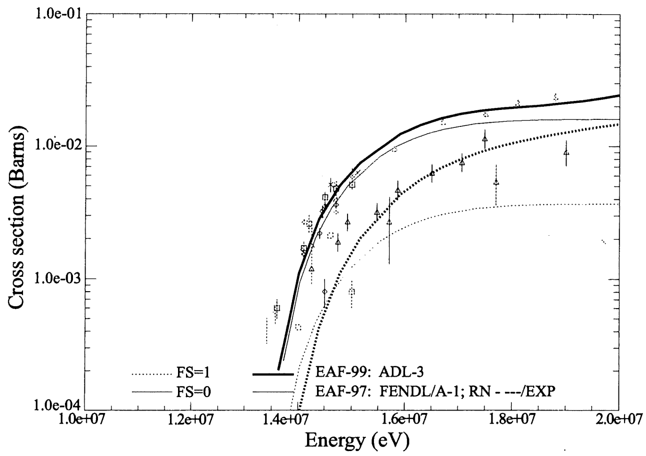


**Fig. 4** K-39 (n,2n): K-38 (7.6 m), K-38m (924.0 ms).
Experimental information from $K_2CO_3$ indicates an underestimation of the metastable production. EXFOR data corroborates such a finding and the excellent branching ration of ADL-3 better fits the experimental information.

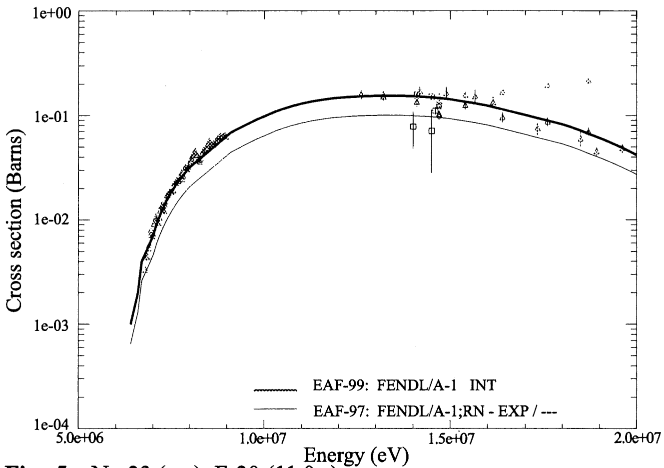


**Fig. 5** Na-23 (n,a): F-20 (11.0 s).
Experimental information on $Na_2CO_3$ indicates a 20% underestimation of F-20 that is perfectly corroborated by the cross section measurement contained in EXFOR.

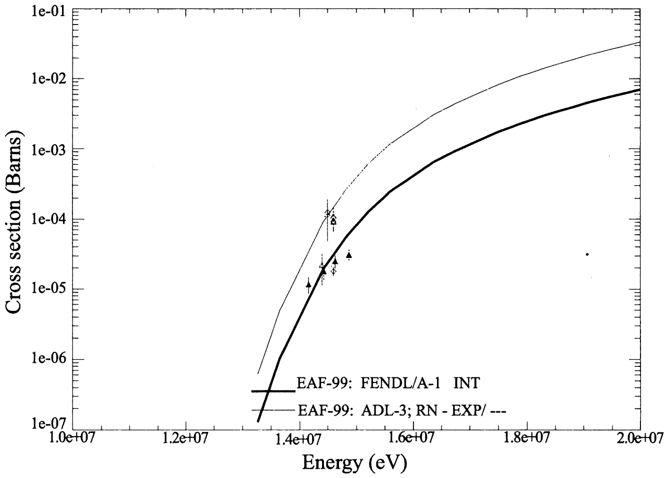


**Fig. 6** Ni-58 (n,t): Co-56 (77.3 d).
Measurement indicates that the calculated prediction overestimates the production of Co-56. This is better satisfied by the ADL-3 cross section. Note that the experimental data are close to the threshold.

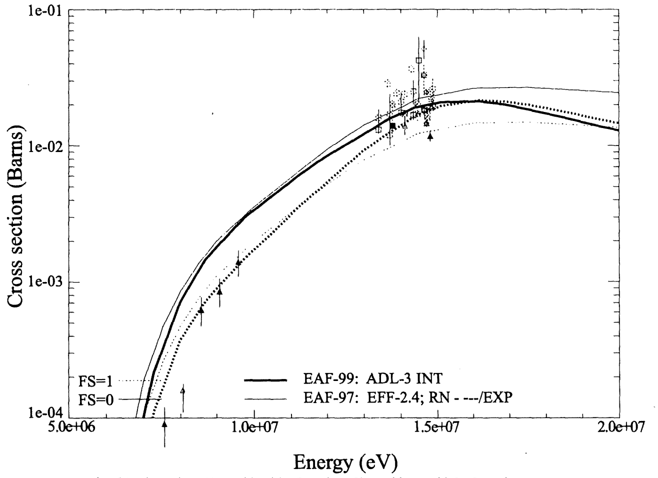


**Fig. 7** Ni-62 (n,p): Co-62 (1.5 m), Co-62m (13.9 m).
The partial channels of ADL-3 better fit the integral measurement. Note the energy dependence of the branching ratio and the rather scattered EXFOR data

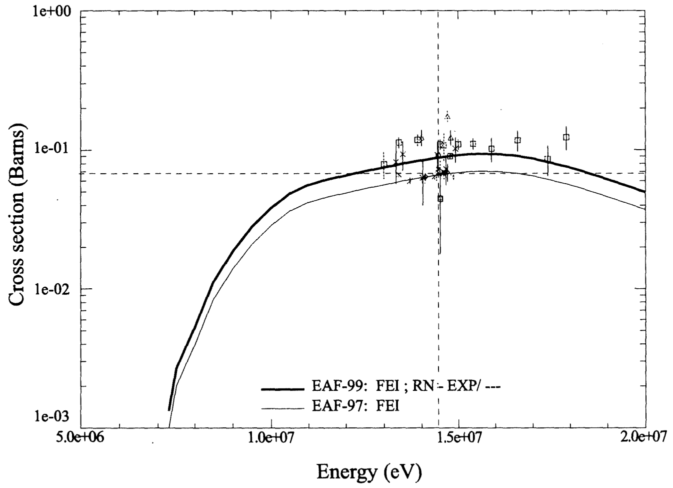


**Fig. 8** Si-30 (n,α): Mg-27 (9.45 m).
A new cross section measurement at 92 mb corroborates the 20% underestimation predicted by calculations on a $SiO_2$ sample.

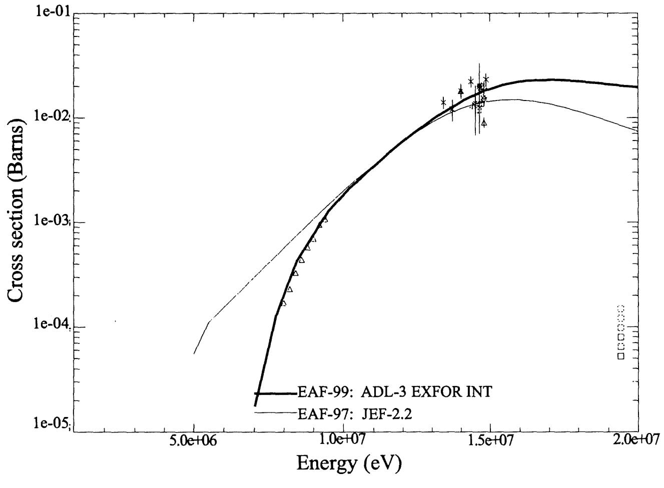


**Fig. 9** Sr-88 (n,p): Rb-88 (17.8 m).
A perfect excitation function shape correction suggested by experimental information on $SrCO_3$ and the EXFOR data.

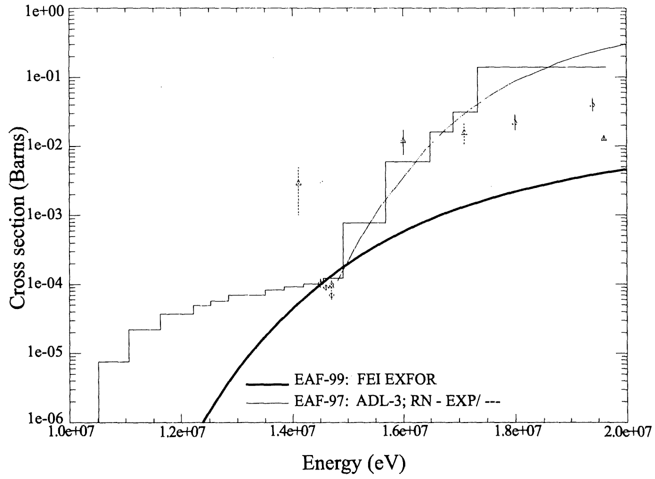


**Fig. 10** V-51 (n,nα): Sc-47 (3.34 d).
Gross overestimation of Sc-47 production was predicted in V samples. The reason were many: wrong interpolation law, renormalisation and threshold. Obviously some of the EXFOR data are inaccurate.

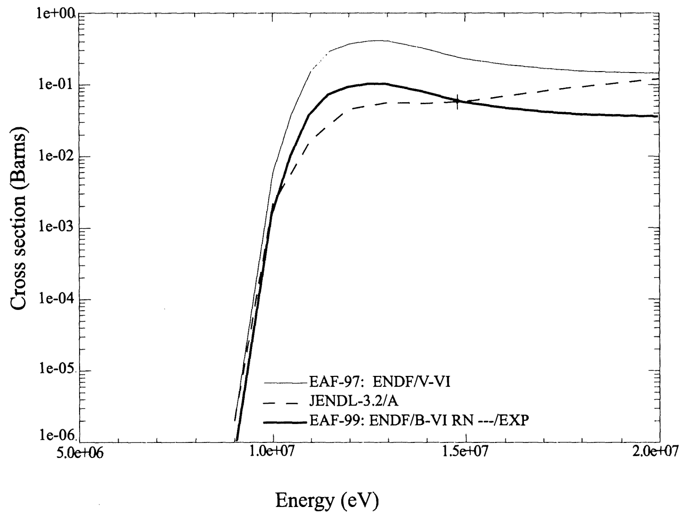


**Fig. 11** V-51 (n,np): Ti-50 (stable).
A very interesting case where only the emitted charged particle could be measured so includes the (n,p) channel. The total (n,xp) measured cross section of 91 mb and a (n,p) of 31 mb would lead to an (n,np) of 60 mb. Such a value is corroborated by kerma and proton emission experimental data on V-51.

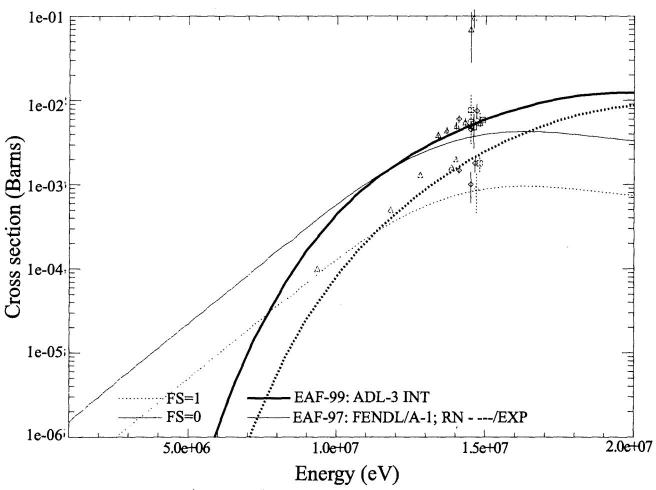


**Fig. 12** Y-89 (n,α): Rb-86 (18.6 d), Rb-86m (1.0 m).
Experimental information on $Y_2O_3$ indicates an underestimation of the production of the metastable. Threshold problems have been detected as well. The ADL-3 data better agree with the experimental information and the ground partial channel measurements.

Around 30 cross sections have been adjusted, in EAF-99, on the basis of the integral validation exercises. In addition, very importantly, 130 cross sections have been validated. One other advantage is that the uncertainty file of EAF-99 has been updated using these findings.

In the Figures the dashed line (Final State = 1) corresponds to the first isomeric state while the continuous line (Final State = 0) relates to the ground state.

## V. Conclusions

The neutron-induced cross section data included in comprehensive activation files such as EAF-99 come from sources having varying degrees of quality. The processing code SYMPAL enables all the necessary modifications to be performed in a well defined and quality assured manner. This leads to the creation of an activation file that can claim a much higher degree of quality than the original sources. The validation of the processed data, in various energy ranges, allows the correctness of the data treatments to be certified at the processing stage.

In EAF-99, for the first time, results of integral experiments have been used to adjust data. Validation of activation code predictions, and thereby of cross section and decay data, has been performed by means of direct comparison with measurements of sample material under fusion-relevant neutron spectra. Integral experiments are a rich source of information with which a wide range of validation and comparison exercises can be made. The broad trends of the time dependent values of the calculated-to-experimental ratios represent the best estimates of the uncertainties that should be applied to predictions made with activation codes.


### Acknowledgement

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