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Neutron Capture Measuremetns on Minor Actinides at the n_TOF Facility at CERN: Past, Present and Future

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The successful development of advanced nuclear systems for sustainable energy production and nuclear waste management depends on high quality nuclear data libraries. Recent sensitivity studies and reports [1-3] have identified the need for substantially improving the accuracy of neutron cross-section data for minor actinides. The n_TOF collaboration has initiated an ambitious experimental program for the measurement of neutron capture cross sections of minor actinides. Two experimental setups have been constructed for this purpose: a Total Absorption Calorimeter (TAC) [4] for measuring neutron capture cross-sections of low-mass and/or radioactive samples and a set of two low neutron sensitivity C_6D_6 detectors for the less radioactive materials.

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I. INTRODUCTION

According to the recent publications "Vision report" [5] and "Strategic Research Agenda" [6] of the European Technological Platform for Sustainable Nuclear Energy (SNETP), the sustainability of nuclear energy requires a combination of the present Light Water Reactors, future Advanced Fast systems (critical reactors or subcritical Accelerator Driven Systems) and waste minimization in closed fuel cycles using Partitioning and Transmutation technologies.

The successful development of advanced nuclear systems for sustainable energy production depends on high-level modeling capabilities for a reliable and cost effective design and safety assessment. High quality and accurate nuclear data are an essential component of such modeling capabilities, since they complete the necessary information about the nuclear reactions taking place in advanced reactors and the fuel cycle. The availability of all the required data, their quality of and the description of their uncertainties in the most recent databases and simulation tools need to be improved. The main challenges come from:

- Isotopes which have become relevant due to their increased presence in the fuels for the new generation of reactors. This applies in particular to the systems aiming at the Pu and Minor Actinides (MA) recycling: fast reactors and ADS, but also Gen III(+) and GenIV reactors with a high Pu load.

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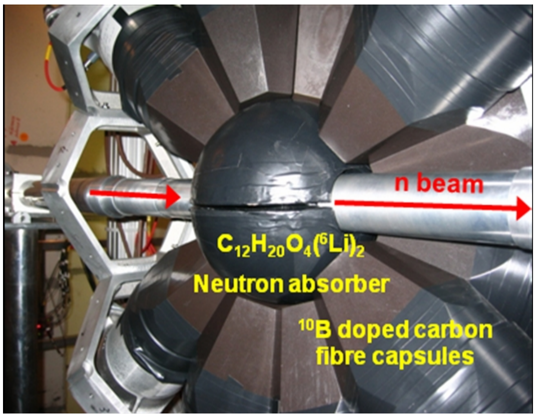

Fig. 1. (Color online) Photography of the open n_TOF Total Abosorption Calorimeter. Visible in the figure are the pentagonal and hexagonal crystals with carbon fibre capsules doped with ^{10}B, and the neutron absorber doped with ^6Li compound at the centre. The travelling sense of the neutron beam is marked with a red arrow.

- The proposal of fast systems, which has enhanced the relevance of the region from 1 eV to several MeV for all isotopes and, very especially, for high mass actinides.
- New fuel cycles with multi-recycling of actinides that could drive to new levels of accumulation of minor isotopes, thus generating additional risks and/or costs at several stages of the fuel cycle. One of the most important aspects is the propagation of the uncertainties as the number of irradiation cycles increases.
- New requirements on the level of precision. In the nuclear industry as everywhere else, the computer

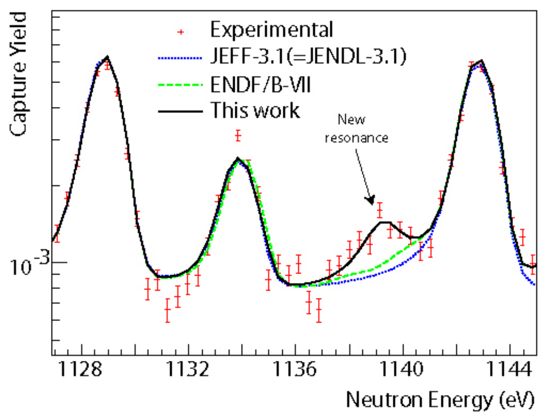

Fig. 2. (Color online) Neutron capture yield for ^{240}Pu. The n_TOF data (red points) are analyzed with the SAMMY code [12] (black line) and compared to the JEFF3.1 evaluated file. The high resolving power at the 185 m flight path has allowed identifying 6 new resonances in the keV region.

simulations should help minimizing (but not replacing) the need for small scale and costly experimental demonstrators. This new role of the simulation tools will require a higher level of precision that can only be achieved if the precision of initial basic nuclear data is previously improved.

- Better assessment of the uncertainties on the data and on the derived magnitudes. This requires a more realistic evaluation of the cross section uncertainties and their correlations between different energies and different channels. Only in this way the estimations from the simulations will profit from the enhanced basic data precision and from the constraints set by the integral experiments.

Recent sensitivity studies and reports [1-3] have identified the nuclear data for different scenarios. From the point of view of the design of innovative reactor concepts, both critical (GenIV) and sub-critical Accelerator Driven Systems, improved nuclear data on fast neutron actinide cross sections are necessary. There are strongly motivated requests for improvement of the nuclear data for ^{238}U (capture and inelastic) and the $^{238-242}$Pu isotopes (capture and fission). From the point of view of the advanced fuel cycles, capture and fission cross sections of Pu, Am and Cm isotopes need to be improved.

II. MEASUREMENTS AT N_TOF

The n_TOF facility [7] was built for providing the nuclear data community an intense spallation neutron source where to measure the cross sections of highly radioactive samples. It operated successfully during the period from 2000 to 2004. In 2004, a segmented Total Absortpion Calorimeter (TAC) consisting in 40 BaF_2 [4] modules was installed and used successfully for the neutron capture cross section measurements of ^{197}Au (as a reference cross section) [8], ^{233}U [9], ^{234}U [10], ^{237}Np, ^{240}Pu, and ^{243}Am in the range between 0.1 eV and 5 keV [11,13]. All the listed measurements were part of the European Commission 5$^{\text{th}}$ Framework Programme (FP) project NTOF-ND-ADS and the analysis was finalized as part of the 6$^{\text{th}}$ FP project IP-EUROTRANS. The overall uncertainty in the cross section obtained with the TAC technique ranges from 3% to 5%, assuming a good characterization of the sample is provided.

As an example of the quality of the data, Fig. 2 shows the experimental yield around 1.1 keV for the ^{240}Pu(n,γ) measurement. The data have allowed performing the resonance analysis up to 2 keV. The cross section was obtained from the combined analysis with the transmission data by Kolar *et al.* [15]. The TAC has been used as well for measuring the first TOF data set in the entire resolved resonance region for the ^{243}Am(n,γ) reaction. Despite the 90 MBq activity of the 6 mg ^{243}Am sample, it has been possible to obtain the capture yield (see Fig. 3) with a good signal to background ratio and to perform the resonance analysis in the resolved resonance region up to 300 keV with the SAMMY code [12]. The n_TOF data have confirmed the overall energy dependence of the transmission data by Simpson *et al.* [16] used for the ENDF-BVII evaluation, have allowed to identify new resonances and to improve the accuracy of the resonance parameters due to a combined capture and transmission analysis.

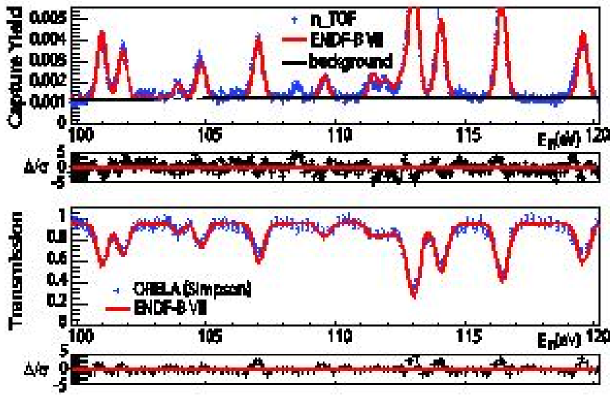

Fig. 3. (Color online) Top: comparison between the n_TOF capture data (blue points) and the ENDF-BVII evaluated file [14]. The data were normalized to the transmission data by Simpson [16]. Bottom: transmission data by Simpson compared to the transmission yield calculated from the ENDF-BVII evaluated file.

III. PRESENT AND FUTURE MEASUREMENTS AT N_TOF

After a three year long stop, the n_TOF facility restarted its operation in 2009 with a new lead target and an improved cooling circuit de-coupled from the neutron moderator circuit, thus allowing a greater flexibility on the characteristics of the neutron beam due to the possi-

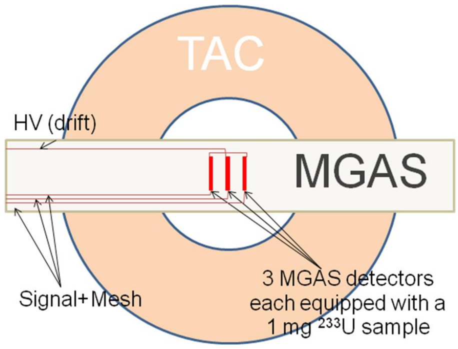

Fig. 4. (Color online) Scheme of the experimental setup to be used for the measurement of the ^{233}U(n,γ) and (n, f) cross sections.

ble use of various moderators like borated water, for the suppression of in-beam gamma rays in ^{1}H(n,γ) reactions, or heavy water, for producing a fast spectrum. In addition, the n_TOF experimental area has been upgraded in 2010 for fulfilling more strict radioprotection and safety rules and has obtained a Work Sector of Type A certification at CERN. This offers an enormous flexibility on the mass, shape and containment of the radioactive materials that can be manipulated and measured at n_TOF, which will not longer need to be sealed in massive ISO-certified containers.

The future plans for (n,γ) cross section measurements with the TAC at n_TOF aim at improving the quality of the data and at performing more challenging experiments:

- The signal to noise ratio of in the capture setup with the TAC has been improved by reducing the dead material and the use of neutron absorber and shielding [17].

- The (n,γ) measurements of the same sample will be performed with two different techniques: TAC and C_6D_6 [18] total energy detectors. The combined analysis of the two data sets will allow better control of the systematic uncertainties associated to each particular technique. This will be the case of the ^{241}Am(n,γ) measurement to be performed in summer 2010 and the ^{238}U(n,γ) in 2011. Both measurements are part of the experimental program of the European Commission 7$^{\text{th}}$ FP project ANDES.

- Perform (n,γ) measurements of more radioactive isotopes and/or samples with reduced masses $\leq$ 1 mg. This will be achieved by combining the use of the Class A experimental area, the use of low gamma ray background moderator, an efficient sample design and, eventually, the exploitation of a future 20 m long vertical flight path, now in planning phase.

- Perform (n,γ) of fissile materials and, in a second phase, simultaneous (n,γ) and (n, f) cross section measurements which will provide accurate fission to capture ratios. This will be the case of a pilot experiment on the ^{235}U(n,γ) with the TAC and a micromegas (MGAS) [19] detector, used as a veto for the electromagnetic cascades associated to the fission process. A more ambitious experiment on the ^{233}U(n,γ) cross section will proceed next. The scheme of the experimental setup is shown in Fig. 4.

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