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Measurements of neutron capture cross-sections for ADS-related studies

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Abstract

Capture cross-sections on several isotopes relevant to accelerator driven systems for energy production and nuclear waste transmutation, and to stellar nucleosynthesis can be studied at the innovative neutron time of flight facility (n_TOF) at CERN. The experimental apparatus is based on a low-mass Si-based flux monitor and a set of C_6D_6 liquid scintillator detectors. The accurate reconstruction of the cross-sections relies on the pulse height weighting function technique. The set-up used in the measurements is here described. The first results on reference isotopes, Au, Ag and Fe, used to verify the accuracy of the method are presented.
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1. Introduction

The experimental determination of neutron cross-sections has always been of fundamental importance in several field of fundamental and applied Nuclear Physics. In the past few years, a pressing request for accurate neutron data has raised, due to new ideas in the field of nuclear technology. Among the concepts being investigated, accelerator driven systems (ADS) could represent a promising solution to the problem of safer energy production and to nuclear waste incineration. For the design and development of ADS, the accurate knowledge of cross-sections on several radioactive isotopes, mainly actinides and long-lived fission fragment, are urgently needed. At present, in fact, standard neutron cross-section databases cannot be considered a reliable basis for ADS-related studies, as they are lacunary and show large discrepancies in many important cases. Available data are based on few experiments, often not in agreement and generally each dedicated to specific energy domains.

Many of the required neutron induced reactions on actinides and long-lived fission fragments can be studied at the innovative neutron time-of-flight facility n_TOF, recently set-in-operation at CERN [1]. The unprecedented high instantaneous neutron flux, combined with the low duty cycle, the high resolution and low background of the n_TOF neutron beam allows one to collect capture cross-section data with good accuracy on several isotopes, including many radioactive ones, with an excellent signal to background ratio even for samples of low mass or high specific radioactivity [2].

Capture measurements planned by the n_TOF Collaboration include the most important isotopes involved in the Th-cycle for energy production, as well as minor actinides and long-lived fission fragments relevant to nuclear waste incineration. Taking advantage of the innovative features of the n_TOF facility, measurements relevant to Nuclear Astrophysics are also planned at n_TOF. Open questions in stellar nucleosynthesis, such as those involving branching isotopes in s-processes, can be consistently addressed at the new facility.

2. ADS-related capture cross-sections

While conventional nuclear power reactors are based on the fission process of ^{235}U and ^{239}Pu, an alternative fuel cycle based on the use of thorium has recently being gaining attention from the scientific and industrial community, since it would result in a much lower production of higher mass actinides, thus reducing significantly the build-up of transuranic isotopes, in particular Pu and Cm, responsible for a large part of the nuclear waste radiotoxicity. In the concept of the energy amplifier [3], the lower neutron multiplicity inherent to the Th-fuel cycle would be compensated by externally supplied neutrons, produced by means of a high-intensity accelerator.

In the Th-based fuel cycle, ^{233}U is the fissile isotope, produced by neutron capture of ^{232}Th followed by β-decays. The renewed interest in the thorium-based nuclear fuel cycle has made clear that the existing knowledge of reaction cross-sections for many of the relevant isotopes, such as ^{232}Th, ^{231}Pa, ^{233,234,236}U, is still rather poor. For example, discrepancies up to 40% in the experimental and up to 30% in the evaluated capture data are still present for ^{232}Th. These uncertainties are too high for a reliable design which should guarantee the safe operation of a Th-cycle based ADS. For a hybrid system with proton injection of constant intensity, a 10% uncertainty on the average capture cross section of ^{232}Th induces a 30% uncertainty on the required proton current for a system operating at the level of $k_{eff} \sim 0.97$ [4].

The high radioactivity stemming from the decay products of the various isotopes involved in the Th-cycle has very much hindered the accurate measurement of their capture cross-sections in the past. A considerably better situation is present at the n_TOF facility. The extremely high instantaneous neutron flux, up to three orders of magnitude higher relative to other facilities, is very favourable for studies of capture reactions on radioactive targets, since it significantly reduces the contribution of the background related to the natural radioactivity of the sample.

Similar arguments apply to the problem of nuclear waste transmutation. For a reliable design of systems for waste incineration, the knowledge of neutron capture cross-sections for many actinides and long-lived fission fragments is of fundamental importance [5]. However, experimental data on such isotopes are still scarce, or present large uncertainties, mainly because the natural radioactivity of the species prevents in most cases an accurate determination of the capture cross-section. A high-priority list of isotopes for which neutron data are needed include several isotopes of Np, Pu, Am and Cm, as well as the many abundantly produced long-lived fission products: ^{79}Se, ^{99}Tc, ^{107}Pd, ^{129}I, ^{135}Cs, ^{151}Sm, etc.... The large background suppression, relative to capture events, that can be achieved at n_TOF thanks to the very low repetition rate of the pulsed neutron beam, will allow one to collect with good accuracy data for many of the actinides and long-lived fission products involved in nuclear waste incineration.

The design of ADS requires capture cross-section data also for structural elements, in particular those constituting the spallation target and the cooling material. Among the various possibilities, a convenient choice would be to substitute the solid target technology with a heavy-liquid metal, in particular Pb–Bi eutectic, acting both as spallation target and for convective cooling [6]. The thermal properties of a liquid target (the boiling point of liquid metals is in excess of 1700 °C for Pb or Pb–Bi) and their high thermal inertia make this choice very attractive, as it would defer or prevent core cooling problems even in the case of complete loss of heat removal. The measurements of the small, resonance-dominated cross-sections for several Pb isotopes and for Bi can be performed at the CERN n_TOF facility, thanks to its excellent energy resolution and low background.

3. The experimental set-up and analysis technique

In the first phase of the n_TOF project, neutron capture measurements are being carried with an array of C_6D_6 (deuterated benzene) liquid scintillator cells. Compared to other γ-ray detectors, they have the advantage of being among the least sensitive to scattered neutrons. Two types of C_6D_6 detectors are presently being used at n_TOF. In order to reduce the neutron sensitivity, the commercially available Bicron BC-537 cylindrical cell, 10 cm diameter and 7 cm thickness, has been modified by substituting the quartz window with a boron-free one and by removing part of the expansion volume. In addition to the Bicron detectors, new C_6D_6 cylindrical cells 12.7 cm diameter by 7.8 cm thickness have been specifically designed and constructed for n_TOF [7]. For these detectors, the use of a carbon fibre container, and a minimization of the support material has led to a very low neutron sensitivity, allowing one to perform measurements of isotopes with a large scattering to capture ratio.

In the first measurement campaign, the detectors were positioned at 90° with respect to the neutron beam direction, at a distance of 4.5 cm from the samples. A remotely controlled sample changer with up to 10 positions, operated in vacuum, was constructed in order to frequently perform periodic measurements of the background (empty sample) and of reference isotopes, that is isotopes with well-known capture cross-sections (such as Au), necessary for normalization purposes. To minimize the background induced by neutrons scattered by the sample and captured in the surrounding material, the sample changer has been constructed in carbon fibre [2].

The quantity determined in a neutron capture experiment is the capture yield, i.e. the fraction of neutrons incident on a sample and undergoing the (n,γ) interaction. The capture yield $Y(E)$ is linked to the capture and total cross-sections by the following relation:

$$Y(E) = (1 - \mathrm{e}^{-n\sigma_T(E)}) \cdot \frac{\sigma_\gamma(E)}{\sigma_T(E)}, \quad (1)$$

where σ_T and σ_γ are the total and capture cross-sections, respectively and n is the number of atoms/barn of the sample.

The experimental determination of the capture yield requires the simultaneous measurement of the neutron fluence for normalization. To this end, a low-mass flux monitoring system has been constructed and is currently being used at n_TOF. It consists on a thin mylar foil with ^{6}Li (or ^{6}LiF) deposit, placed in the beam, viewed by an array of silicon detectors, 300 μm thick and 6×4 cm^2 surface, placed outside the beam, for the detection of tritons and alpha particles emitted in the ^{6}Li(n,α) reaction. The small amount of material in the beam ensures a low background from scattered neutrons. Furthermore, to minimize the γ-ray background caused by capture of scattered neutrons in the support structure, the vacuum chamber hosting the flux monitor has been constructed in carbon fibre. Fig. 1 shows a picture of the monitor flux mounted on the neutron beam line. The thickness of the ^{6}Li deposit was chosen as a trade-off between the need of a high-count rate and that of a clear identification of tritons and α particles in the Si-detectors, which is affected by the energy loss within the ^{6}Li layer. The optimal thickness of the deposit was studied by means of detailed simulations of the device, performed with GEANT4, in which the resolution of the Si-detectors was also taken into account. According to the results of the simulations, a pure ^{6}Li deposit of 200 μg/cm^2, or equivalently of 500 μg/cm^2 of ^{6}LiF, was chosen. The efficiency of the device have been estimated by simulations and experimentally determined at n_TOF by comparison against a calibrated fission chamber.

Due to the small solid angle coverage and the low intrinsic efficiency of the C_6D_6 detectors, which result in an overall efficiency of ~10%, only one γ-ray per event is detected from the de-excitation cascade following neutron capture. For an accurate cross-section determination, the efficiency of the set-up has to be made independent on the

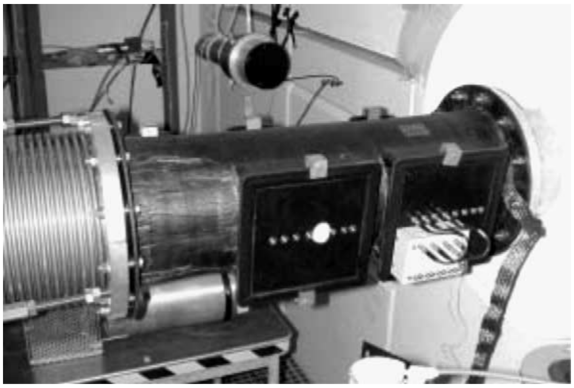

Fig. 1. A photo of the low-mass flux monitor mounted on the n_TOF neutron beam line. The carbon fibre vacuum chamber hosts a ^{6}Li-foil and a set of Si detectors.

details of the de-excitation cascade, in particular on the γ-ray multiplicity. To this end, a pulse height weighting function technique (PHWF) can be used [8]. It consists in suitably modifying by software the detector response so that the efficiency ε_γ is proportional to the photon energy E_γ ($\varepsilon_\gamma = kE_\gamma$). Under these condition the efficiency for detecting a cascade becomes proportional to the known cascade energy E_c and independent of the actual cascade path ($\varepsilon_c \approx \Sigma_j \varepsilon_{\gamma j} = kE_c$). The proportionality of the efficiency with the γ-ray energy is achieved by modifying the detector energy response distribution $R(E)$ with a pulse height (deposited energy) dependent weighting factor $W(E)$, applied to the recorded spectrum. The weighting factor is determined by minimizing the following expression for different γ-rays energies in the range of interest (up to 10 MeV):

$$\chi^2 = \sum_i \left(\frac{\sum_j R_j^i W_j - E_{\gamma i}}{\sigma_i} \right)^2, \qquad (2)$$

where R_j represents the binned $R(E)$ energy distribution. The detector response is simulated for each sample under investigation with the code GEANT (versions 3.21 and 4), in which a detailed software replica of the C_6D_6 and other elements of the experimental apparatus have been implemented. The energy resolution, measured with γ-ray sources (^{137}Cs, ^{60}Co and Pu/C) is also included in the simulation.

A validation of the weighting function technique, and in particular of the accuracy of the simulations of the detector response, has been performed by measuring isotopes with well-known capture cross-sections. In particular, data for ^{nat}Fe, ^{nat}Ag and ^{197}Au samples have been analyzed. The calculated weighting functions for the three samples, as a function of the energy deposited in the C_6D_6 is shown in the upper panel of Fig. 2.

After applying the weighting functions and normalizing for the measured neutron fluence, the experimentally determined capture yield is obtained. In the lower panel of Fig. 2, the ratio between measured and predicted yield for the major resonance of the three samples (4.9 eV for ^{197}Au, 5.2 eV for ^{109}Ag and 1.15 keV for ^{56}Fe) is shown as a function of the capture energy. The predicted yield was calculated from the tabulated cross-sections extracted from

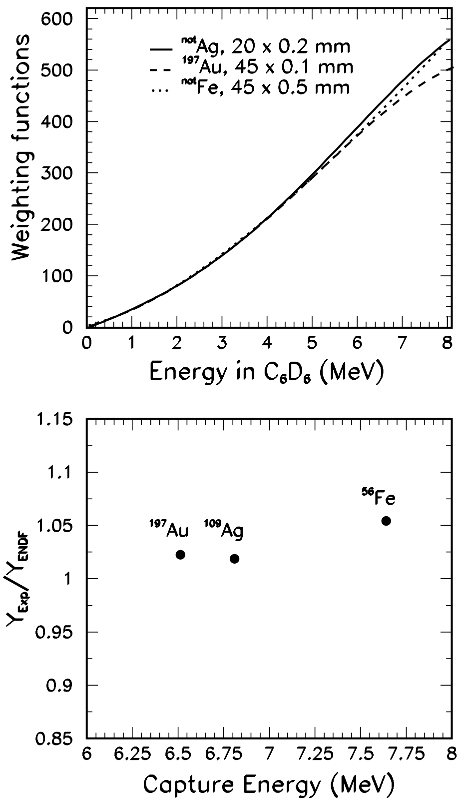

Fig. 2. Upper panel: the calculated weighting functions for three samples measured at n_TOF with Bicron C_6D_6 detectors, versus the energy deposited in the detectors. The weighting functions were obtained by simulating the response of the detectors to γ-rays of 10 different energies, from 1 to 10 MeV. Lower panel: the ratio of experimental capture yields, obtained after application of the weighting functions, to the predicted ones, are shown for three samples as a function of the respective capture energy. The yields refer to the major resonances of the three samples, and were obtained by integration over the resonance area.

the ENDF/B-VI database. An agreement with the prediction within 5% is observed for all three cases. Part of the difference can be most probably attributed to a systematic uncertainty in the flux measurement. When normalized to Au, the other two samples show a relative uncertainty within 2%. This result demonstrates the reliability of the pulse height weighting function technique and of the simulations of the detector response, thus providing confidence on the accuracy of the capture cross-sections measurements at n_TOF.

Fig. 3 shows the capture yield as a function of the neutron energy for a sample of Au, 0.1 mm thick, measured with the Bicron C_6D_6 cell. The figure demonstrates the important features of the n_TOF facility, which are of great advantage in capture cross-section measurements: the wide energy range of the neutron beam, its high resolution and the low background. Taking advantage of these features, a vast experimental program on ADS-related capture studies has recently started at the n_TOF facility.

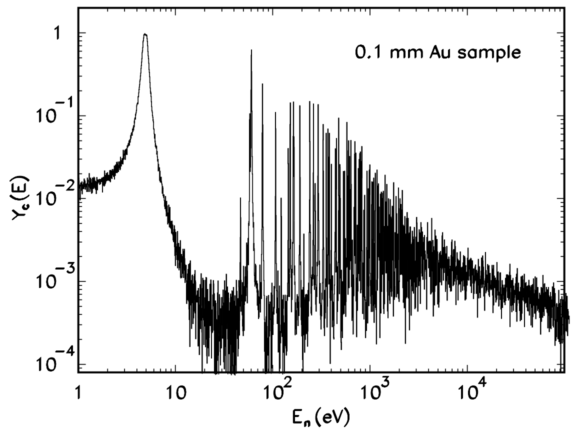

Fig. 3. The capture yield as a function of the neutron energy for a 0.1 mm thick Au sample, measured at n_TOF for 3000 neutron bunches. The pulse height weighting function technique was applied in the analysis. The main features of the n_TOF facility, that is the high flux (10^5 n/bunch/cm^2) in a wide energy range, the high resolution ($\Delta E/E < 10^{-3}$) and low background, can be appreciated in the figure.

4. Conclusions

Simulations, tests and preliminary experimental results have confirmed that high accuracy neutron cross-sections data related to the development of ADS for safer energy production and nuclear waste incineration can be measured at the new n_TOF facility at CERN. The innovative features of the facility, in particular its very high instantaneous neutron flux, high resolution and low background, will allow one to improve the quality of existing data, to extend the resolved resonance region and, most importantly, to measure isotopes for which data are still missing or largely incomplete. This applies, in particular, to many radioactive samples involved in the Th-fuel cycle or to actinides and long-lived fission products that constitute the nuclear waste.

The experimental apparatus, currently based on a low-mass flux monitoring system and a set of C_6D_6 γ-ray detectors, combined with the pulse height weighting function technique, will be used in the first phase of the n_TOF project for the measurement of capture cross-sections. A validation of the apparatus and of the analysis method has been obtained by performing reference measurements on Au, Ag and Fe samples. The results testify of the high accuracy that can be achieved on neutron cross-section data at the n_TOF facility.

Acknowledgements

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