EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Radiative capture on ^{242}Pu for MOX fuel reactors

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Abstract

The use of MOX fuel (mixed-oxide fuel made of UO_2 and PuO_2) in nuclear reactors allows substituting a large fraction of the enriched Uranium by Plutonium reprocessed from spent fuel. Indeed around 66% of the plutonium from spent fuel is made of ^{239}Pu and ^{241}Pu, which are fissile in thermal reactors. A typical reactor of this type uses a fuel with 7% reprocessed Pu and 93% depleted U, thus profiting from both the spent fuel and the remaining ^{238}U following the ^{235}U enrichment.

With the use of such new fuel compositions rich in Pu the better knowledge of the capture and fission cross sections of the Pu isotopes becomes very important. This is clearly stated in the recent OECD NEA's "High Priority Request List" and in the WPEC-26 "Uncertainty and target accuracy assessment for innovative systems using recent covariance data evaluations" report. In particular, a new series of cross section evaluations have been recently carried out jointly by the European (JEFF) and United States (ENDF) nuclear data agencies. As the new evaluations on ^{240}Pu and ^{241}Am have been already completed, ^{242}Pu is the next to be reevaluated, and the scarceness of capture data (only two TOF measurements from 1973 and 1976 are available and disagree with each other) calls for a new time-of flight capture cross section measurement. This will be the first measurement in 40 years and, with the use of more advanced techniques, shall provide a more reliable and accurate result.

We propose to measure the capture cross section of ^{242}Pu in the region from thermal up to at least 60 keV, aiming for a high energy limit of 500 keV. The experiment would make use of an array of 4 low neutron sensitivity C_6D_6 detectors and be carried out at the n_TOF EAR-1 (185 m flight path) measuring station. Compared to the current uncertainty of 35%, this measurement aims at an improved accuracy between 7% and 12% depending on the energy region.

Requested protons: 3.5×10^{18} protons on target.

1. INTRODUCTION AND MOTIVATION

The measurement of accurate capture and fission cross sections is essential for the design and operation of current and innovative nuclear systems such as Accelerator Driven Systems and Gen-IV reactors aimed at the reduction of the nuclear waste [1]. These advanced nuclear reactor concepts make use of fuel different from previous ones and thus the response to neutrons of different energies in these fuels must be investigated in detail. Already nowadays, but even more in the case of future reactors, the burned fuel can be recycled in order to separate the plutonium from the spent fuel. This plutonium contains 66% of ^{239}Pu and ^{241}Pu, which are fissile and can be thus combined with depleted uranium (^{238}U) to make what is known as MOX (mixed oxide) fuel. In this way the Pu from spent fuel and the depleted uranium, otherwise considered as waste, are used in a new reactor cycle, contributing in this way to the long-term sustainability of nuclear energy.

Currently, the use of MOX fuel has been established on an industrial scale in a number of countries. In Belgium, France, Germany, Japan and Switzerland, there are a considerable number of thermal power reactors using MOX fuel. However, the utilization of MOX fuel only in thermal power reactors will not ultimately resolve the issues related to accumulation of quantities of discharged spent fuel and separated plutonium requiring to be stored, but a much more efficient use of plutonium will ultimately be made in fast reactors, where multiple recycling is possible and has been demonstrated. See [2] for more details on the status and progress in MOX fuel technologies.

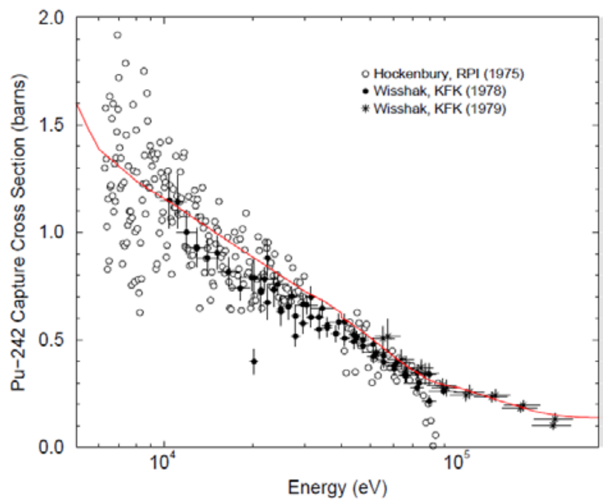

Figure 1. Measured (markers) and JEFF-3.1 evaluated (red-line) neutron capture cross section of ^{242}Pu in the region of interest for MOX fuel nuclear reactors.

The extensive use of MOX fuels in fast reactors calls for a revision of the neutron cross sections that play a role in the neutronics of such reactors and are not known with enough accuracy yet. For the particular case of ^{242}Pu, the first attempts to measure its neutron capture cross sections were made in 1973 and 1976, when Portmans et al. [3] and Hockenbury et al. [4] used the time-of-flight technique for measurements at low (below 1.3 keV) and high (6-87 keV) neutron energies, respectively. A few years later, Wisshak and Kaeppeler [5,6] measured the cross section in the 10-90 and 50-250 keV energy intervals. The comparison of the results (see Figure 1) indicates an uncertainty of about 35% in the capture cross section value in the keV region.

In this context, the Nuclear Energy Agency recommends in its "High Priority Request List" [7] and its report WPEC-26 "Uncertainty and target accuracy assessment for innovative systems using recent covariance data evaluations" [8] that the capture cross section of ^{242}Pu should be measured with an accuracy of at least 7-12% in the neutron energy range between 500 eV and 500 keV (see Table 1). Furthermore, interpretations with JEFF-3.1 of the PROFIL and PROFIL-2 experiments carried out in the fast reactor PHENIX have shown an overestimation of about 14% of the ^{242}Pucapture cross section [9,10]. In addition to the direct measurement in the fast energy region (E_n>2 keV), accurate average radiation width and strength function are required to solve some ambiguous results obtained between optical model calculations and the statistical analysis of the *s*-wave resonances [11]. This calls as well for an accurate measurement of the resonance region (1 to 1000 eV) with enough resolution to determine accurately such average resonance parameters.

Table 1. Current and required accuracy for the capture cross section of ^{242}Pu for different nuclear reactor concepts, including as well the neutron energy range of interest [7,8].

	Neutron Energy range	Present accuracy (%)	Required accuracy (%)
SFR	2-500 keV	35	8
EFR	2-67 keV	35	25
GFR	2-183 keV	35	7-8
LFR	9-183 keV	35	11-12
ADMAB (ADS)	9-25 keV	35	10
PHENIX	0.5-2 keV	14	7

2. EXPERIMENTAL SET-UP

2.1 Detection system

At n_TOF [12] there are, as of today, two detection systems available for detecting γ-rays in neutron capture measurements: the Total Absorption Calorimeter TAC (a 4π BaF_2 array) and the Total Energy detectors (a pair of C_6D_6 scintillators). In this proposal we chose the C_6D_6 detectors not only because of their lower neutron sensitivity but mainly because they suffer significantly less from the so-called γ-flash, thus allowing measurements at higher neutron energies. This is important because the neutron energy range of interest in this measurement (see Table 1) reaches up to 500 keV, while the limit for the TAC measurements is usually ~20 keV.

One of the drawbacks of the C_6D_6 detectors with respect to the TAC is its lower detection efficiency, which we will maximize by using for the first time at n_TOF an array of four new C_6D_6 detectors [13], thus bringing the efficiency up to ~40%. The simplified sketch of the set-up is illustrated in Figure 1. However, since this new set-up is still to be tested, the calculations presented below correspond to a conservative efficiency value of 20%.

Another possibility would be to perform the measurements with the n_TOF Total Absorption Calorimeter (TAC). This has the advantage of a higher efficiency and better background discrimination capabilities with respect to the C_6D_6; however, the highest neutron energy that can be reached is limited to ~10 keV because of a very slow recovery from the γ-flash. If the modification in the electronics of the TAC crystals for improving this point are successful, and the background in the C6D6 is not really improved (see section 2.3 below), the TAC could also be consider for the experiment. However, the C_6D_6 is nowadays the best detector choice for this particular measurement.

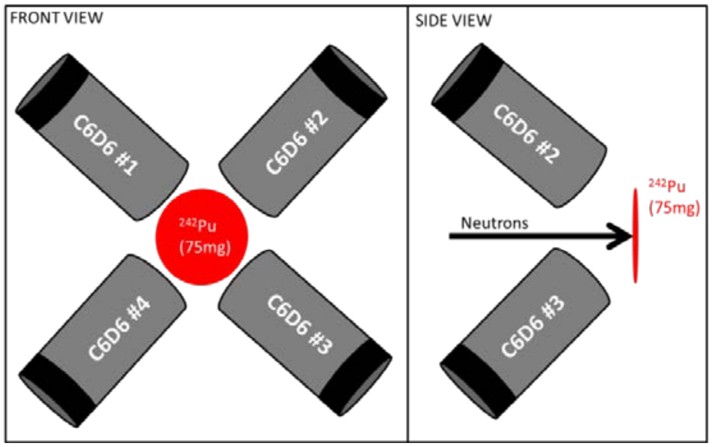

Figure 2. Sketch of the C_6D_6 array that will be used for measuring the capture cross section of ^{242}Pu. Four new C_6D_6 detectors, with reduced neutron sensitivity and enhanced safety, will be looking at the sample from a backward position.

2.2 Samples

As in many of the n_TOF experiments, the availability and/or the preparation of a suitable sample are crucial to the success of the measurement. In the case of ^{242}Pu, we will profit from our collaboration within the EC-FP7-ERINDA project [14] with the nELBE facility [15] in Dresden, Germany. In collaboration with the University of Mainz and using a total of 75 mg of high purity $^{242}PuO_2$, a series of eight thin ^{242}Pu samples 76 mm in diameter deposited on a 0.1 μm Ti backing is under preparation. The technique to be employed is molecular plating [16] through which surface densities of the order of a few mg/cm^2 can be achieved (the limit with evaporation techniques is ~500 μg/cm^2). These samples, intended for fission reaction measurements at nELBE, are well suited for measuring the capture cross section at n_TOF. Furthermore, we are planning to prepare, using the same technique and the additional ^{242}Pu material available at nELBE, a new set of samples with a total mass of 100 mg deposited in a diameter of 40 mm, which is even better suited for a capture measurement at n_TOF .

2.3 Improved background conditions

As shown below, the expected background level is of a similar magnitude than the capture contribution in the unresolved resonance region (above 1 keV). If measured with enough accuracy, this background level should still allow an accurate measurement of the capture cross section. However, it is expected that the modifications planned in the n_TOF EAR-1 beam line will result in a sizable reduction of this background level. These actions include the elimination of the aluminum (0.5 mm thick) vacuum window, and the reduction of the diameter of the evacuated beam line by a factor of two. The measurement proposed here will only take place after the background level determination during the commissioning of EAR-1 after these modifications are implemented (beginning of 2014).

3. OBJECTIVES AND BEAM TIME REQUEST

Considering the concluding remarks of the OCDE NEA "High Priority request List" and "Uncertainty and target accuracy assessment for innovative systems using recent covariance data evaluations", the measurement shall aim to determine the capture cross section of ^{242}Pu with an accuracy between 7% and 12% depending on the neutron energy range, being the neutron energy interval of interest from 500 eV to 183-500 keV (see Table 1).

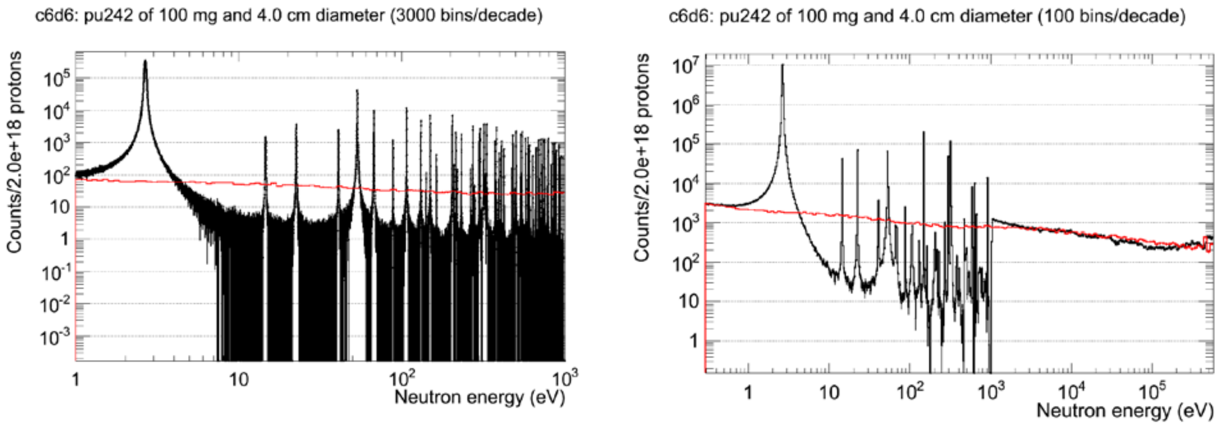

Figure 3. Expected counting rate for the ^{242}Pu(n,γ) reaction under neutron irradiation with a total intensity of 2×10^{18} protons on target. Top: 3000 bins/decade, needed to study the resolved resonance region. Bottom: 100 bins/decade, needed for the study of the smooth Unresolved Resonance Region. The red line indicates the expected background level, considering the data available from 2012, that is before the improvements in the neutron beam line that shall reduce the background significantly (see text for details).

The beam time request (see Table 2) has been calculated attending to the minimum statistics needed to achieve the accuracies mentioned above. We have assumed a conservative value of 20% for the detection efficiency, a ^{242}Pu mass of 100 mg over a 40 mm diameter sample, the detailed characteristics of the n_TOF neutron beam, and the latest ENDF evaluated cross section values [17]. The calculation of the expected counting rate indicates that a minimum of 2×10^{18} protons on target are needed to register more than 1000 counts at the peak of the resonances (in the resolved resonance region below 1 keV). With this number of protons one also expects a minimum of 200 counts per bin in the Unresolved Resonance Region (16 to 600 keV). This will provide a point-wise data set with a statistical uncertainty of ~7%, which will dominate over the systematic uncertainty, expected to be around 5%.

In addition to the measurement of the ^{242}Pu sample, a series of other measurements will be performed in order to estimate the background and to validate the methodology. The validation of the measurement at high neutron energy will be achieved by measuring a gold (^{197}Au) sample with 1000 mg and 45 mm in diameter which, after irradiation with 4×10^{17} protons, will provide data with less than 2% statistical uncertainty in the unresolved resonance region. The capture cross section of ^{197}Au is considered standard above 150 keV and thus the comparison of our results with the evaluated cross section will demonstrate the quality of the measuring technique or indicate the existence of systematic errors in case that the results are not satisfactory, which can be then tackled and solved.

Table 2. Number of protons requested for the measurement of the ^{242}Pu(n,γ) cross section and auxiliary samples.

Sample	Purpose	Protons
^{242}Pu (100 mg, 40 mm diam.)	Capture Cross Section measurement	2.0×10^{18}
^{197}Au (1000 mg, 40 mm diam.)	Validation of the σ(n,γ) measurement	0.4×10^{18}
Sample-out/Dummy	Sample-independent background	0.8×10^{18}
^{nat}Pb/^{12}C	Sample-dependent background	0.3×10^{18}
Beam-off	Room background	-
TOTAL		**3.5×10^{18}**

As in previous C_6D_6 measurements at n_TOF, the determination of the different types of background (sample dependent/independent) for both ^{242}Pu and ^{197}Au will be determined by performing sample-out, dummy, ^{nat}Pb and ^{12}C measurements using a total of 0.8x10^{18} protons.

4. CONCLUSIONS

We propose to measure the capture cross section of ^{242}Pu, which is one of the main components of MOX fuel in current and future reactors. The proposal follows the request from the nuclear reactor design community and the Nuclear Energy Agency recommendations. The measurement would be performed at n_TOF EAR-1 on a $^{242}PuO_2$ sample of 100 mg and 40 mm in diameter making use of an array of C_6D_6 detectors. The measurements aims at an overall uncertainty of ~7-12% in the neutron energy region all the way up to 500 keV.

A total of 3.5x10^{18} protons are requested for measuring the ^{242}Pu capture cross section and the associated background and validation measurements.

Summary of requested protons: 3.5x10^{18} protons on target.

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