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First high resolution measurement of neutron capture resonances in ^{176}Yb at the n_TOF CERN facility.

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Abstract. Several international agencies recommend the study of new routes and new facilities for producing radioisotopes with application to nuclear medicine. ^{177}Lu is a versatile radioisotope used for therapy and diagnosis (theranostics) of cancer with good success in neuroendocrine tumours that is being studied to be applied to a wider range of tumours. ^{177}Lu is produced in few nuclear reactors mainly by the neutron capture on ^{176}Lu. However, it could be produced at high-intensity accelerator-based neutron facilities. The energy of the neutrons in accelerator-based neutron facilities is higher than in thermal reactors. Thus, experimental data on the ^{176}Yb(n,γ) cross-section in the eV and keV region are mandatory to calculate accurately the production of ^{177}Yb, which beta decays to ^{177}Lu. At present, there are not experimental data available from thermal to 3 keV of the ^{176}Yb(n,γ) cross-section. In addition, there is no data in the resolved resonance region (RRR). This contribution shows the first results of the ^{176}Yb capture measurement performed at the n_TOF facility at CERN.

1 Introduction

Nuclear medicine has proven to be a much needed medical specialty in order to diagnose and treat several diseases, among them, cardiovascular diseases and cancer, the first and the second causes of mortality worldwide, respectively [1]. Diagnosis represents 90% of the procedures in nuclear medicine and the most used radioisotope is the ^{99}Mo/^{99m}Tc which is produced in nuclear reactors [2]. For this reason, the so-called "world technetium crisis" in 2009 was a first alarm to find out new procedures and facilities to produce radioisotopes for nuclear medicine [3,4]. In addition, in the last decades more than 3000 new radioactive isotopes have been discovered in different nuclear physics facilities. Many of them could have properties that make them useful for nuclear medicine and potentially they could have better properties for diagnosis and therapy than the conventional ones.

In this framework, several international agencies and committees recommend the study of new routes for producing radioisotopes with application to nuclear medicine [2,3,5,6]. This has been specially pushed in the last years with the development and the availability of high-intensity accelerators and new installations. In addition, these new installations can provide several radioisotopes quantities at regional level. CERN's MEDICIS at Isolde facility is an excellent example [7].

Besides the charged-particle facilities also accelerator-based neutron sources are being considered for radioisotope production. In this context, IFMIF-DONES (International Fusion Material Irradiation Facility - Demonstration Neutron Source) is one of the possible facilities to be used for this. It is an ESFRI (European Strategy Forum on Research Infrastructures), and the European city host is Granada (Spain) [8]. Its main objective is the irradiation of materials for fusion reactor technology. IFMIF-DONES has already considered the production of radioisotopes as one of the main complementary applications of the facility [9]. Indeed, the design of the building includes an experimental hall for such applications.

Furthermore, ^{177}Lu ($t_{1/2} \approx 6.65$ d) is one the of most important emergent radioisotopes [10]. It is used for theranostics (therapy and diagnosis), with good success in gastroenteropancreatic neuroendocrine tumours [11]. Currently, ^{177}Lu is under study for several other tumours with good results [12]. At present, ^{177}Lu is only produced in nuclear reactors through two production routes: the direct route, ^{176}Lu(n,γ)^{177,177m}Lu; and the indirect route, ^{176}Yb(n,γ)^{177}Yb ($t_{1/2} \approx 1.9$ h) →^{177}Lu + ^{177m}Lu [13].

At present, the most used production route is the direct route, ^{176}Lu(n,γ). Although the cross-section of the direct route is higher, several advantages in the indirect route have been pointed out:

i) The specific activity is four times higher [13]. About 100% of the theoretical specific activity can be achieved [14,15].

ii) The contaminants that remain in the final quantity of the material are much lower.

iii) The undesirable ^{177m}Lu ($t_{1/2} \approx 160$ d) is produced in the direct route, 0.05%, whereas in the indirect route it is less than 10^{-5} % [16]. ^{177m}Lu creates important problems, the urine of the patient must be treated as radioactive waste, and the patient receives an undesirable dose.

These properties have a direct impact on the quality of the diagnosis and the therapy. The higher specific activity allows a much better tumour uptake; thus, the dose deliver to tumour for the same activity is much higher in case of the indirect route, and in addition, the quality of imaging of the tumour is much better [15,16].

In order to investigate the feasibility of producing ^{177}Lu in neutron irradiation facilities by the indirect route, it is crucial to have accurate nuclear data of the reaction ^{176}Yb(n,γ). However, at present there is data only from one measurement by Wisshak et al [17], from 8 to 100 keV energy range. None of the resonances of the reaction were measured in this experiment. In addition, discrepancies are found with other experiments carried out by means of integrated cross-section in this energy range.

There are several important facts that indicate a high-resolution measurement is needed. The most relevant are:

i) There is no data from thermal to 3 keV.

ii) There is no high-resolution measurement above 3keV of the ^{176}Yb(n,γ) cross-section. However, resonances have been detected in transmission experiments [18].

Regarding nuclear data libraries, ENDF/B-VIII.0 [19] and JEFF-3.3 [20] use different upper limits for the resolved resonance region (RRR), 5 and 50 keV, respectively.

Therefore, the measurement presented here will provide, for the first time, high resolution data of the ^{176}Yb(n,γ) cross section from thermal up to 100 keV, covering the full range of the RRR.

2 Experiment at the n_TOF (CERN) facility

The neutron capture cross section of ^{176}Yb has been measured by means of the Time-of-Flight technique at the neutron Time-of-Flight (n_TOF) facility at CERN. At n_TOF, pulsed neutron beams are produced by spallation of proton beams from CERN PS accelerator into a massive lead target. Experiments are performed in two Experimental AReas (EAR): EAR1 has very high resolution in energy due to its 185 m flighpath, whereas EAR2, which is only 20 m away from the target, has worse resolution but much higher flux.

In the CERN's Second Long Shutdown (2019-2020), the facility has gone through a major upgrade, consisting of the installation of a new spallation target designed to fully optimise the features of both beam lines [23]. In 2021, a full commissioning campaign was carried out in order to characterize the neutron flux of both EARs with the new target [25,26].

Aiming at the highest possible in resolution, the experiment was performed in EAR1. The ^{176}Yb(n,γ) reaction was measured by employing four custom-made C6D6 liquid scintillation detectors, optimized for an extremely low neutron sensitivy. The yield of the reaction is determined by applying the Total Energy Detection (TED) [24,27,28].

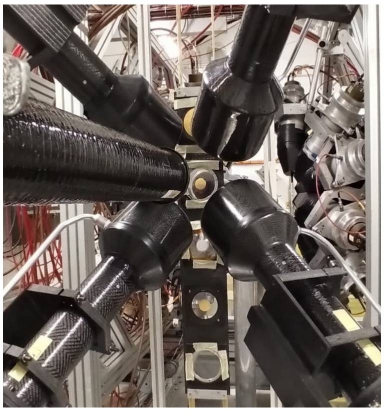

Fig. 1. Experimental Set-Up with four custom C_6D_6 scintillation detectors, and the $^{176}Yb_2O_3$ enriched sample in position.

These detectors were positioned at 125° to the beam position to reduce the in-beam gamma ray background and minimize the effects of the primary radiation angular distribution [29], Fig. 1 shows the experimental set-up. In addition, a SiMon detector, cosisting in an array of four silicon detectors facing a thin enriched lithium fluoride foil, was used to monitor the neutron flux during the experiment.

The sample used for the experiment consisted in a $^{176}Yb_2O_3$ enriched to 99.43% purity, with a weight of 1.5976 g. The powder was pressed into a quartz capsule with internal diamater and thickness of 19 and 2 mm, respectively. The sample and the quartz capsule are shown in Fig. 2.

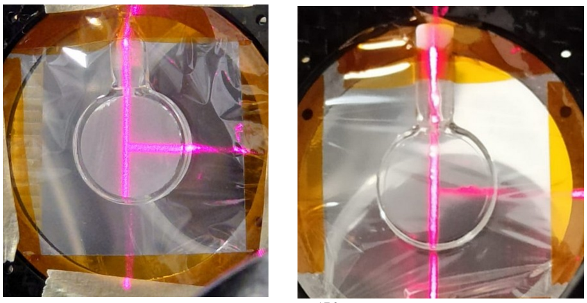

Fig. 2. The left image shows the $^{176}Yb_2O_3$ enriched sample put in place, and the right image shows the quartz capsule put in place.

The measurement was carried out at the end of the 2021 campaign at n_TOF. In addition to the $^{176}Yb_2O_3$ sample, additional ancillary measurements were performed: a measurement on a gold sample, whose capture cross section is accurately known, was performed for absolute yield normalization by applying the saturated resonance technique [30]; an empty ^{176}Yb quartz capsule was used to measure its contribution to the background, and lead and carbon samples were empoyed to estimate the background due to scattering of neutrons and in beam gamma rays in the ^{176}Yb sample.

In addition, calibration runs with radioctive sources were perfomed on a weekly basis to ensure an accurate amplitude-to-energy calibration throughout all the measurement.

The total number of counts as a function of neutron energy with the ytterbium sample are shown in Fig. 3, for one detector. Also, beam-related background to the quartz capsule (Dummy Sample), beam-related background (Empty) and no-beam-related background (BeamOFF) are also shown.

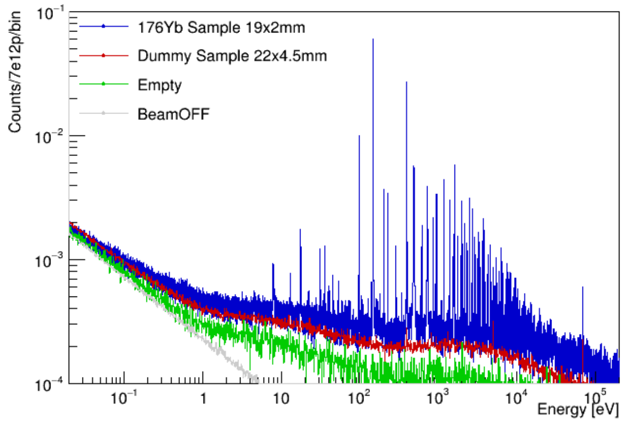

Fig. 3. Total counting rate per pulse of ^{176}Yb (5000 bins/decade) and contribution of the different background components (500 bins/decade). Background contribution of scattered neutrons and in-beam γ-rays not included yet.

As previously mentioned, in order to determine correctly the capture yield with C6D6 detectors it is necessary to apply the TED technique. For this, two conditions must be fulfilled: i) no more than one γ-ray is detected per capture cascade, and ii) the detection efficiency is proportional to the γ-ray energy.

Fulfilling these two conditions, we obtain that the efficiency is proportional to the energy of the cascade, and therefore, independent of the de-excitation path. It means:

$$\varepsilon_C=\alpha E_C=\alpha(S_n+E_n), \quad (1)$$

where where S_n is the neutron separation energy of the resulting nucleus and E_n is the energy of the incident neutron.

The first condition can be easily achieved by using small volume and low Z gamma ray detectors, like the C6D6 detectors used for this measurement. In order to fulfil the second condition we applied the Pulse Height Weighting Technique (PHWT), which has been validated to perform measurements at n_TOF [27].

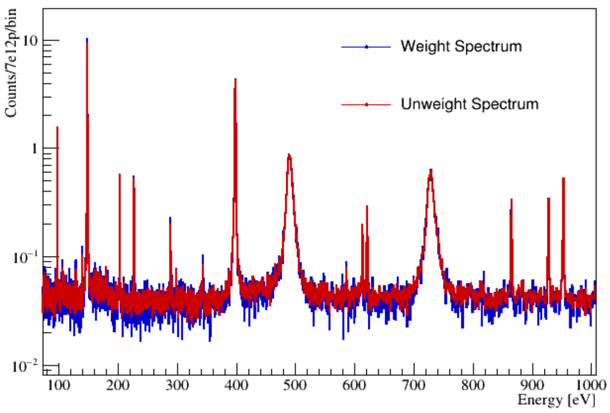

Fig. 4. The blue line represents the weighted counts for the Ytterbium sample, the red line represent the unweighted counts for the Ytterbium sample. The unweighted count has been normalized to the weighted counts at the first ytterbium resonance.

In this technique, each count detected is weighted according to its amplitude by a factor, given by a Weighting Function (WF). To calculate the WF it is necessary to obtain the response function of the C6D6 detectors to monoenergetic g rays in the energy range of interest, which is accomplished by employing detailed MC simulations of the full detection setup [31].

Fig 4 shows a comparison between the weighted and the unweighted counts per pulse spectra measured with the ^{176}Yb sample, in the neutron energy range between 100 and 1000 eV. No background subtraction has been applied. The non-weighted counts has been normalized with respect to the weighted counts, at the first resonance (~98 eV, according to resonances reported in ENDF).

3 Results

After applying the PWHT, the experimental yield Y_{exp} can be expressed as:

$$Y_{exp}(E_n)=f_{Norm}\cdot f_{Corr.}\cdot\{C_w(E_n)-B_w(E_n)\}/\{\Phi_n(E_n)\cdot(S_n+E_n)\} \quad (2)$$

where C_w are the weighted sample counts and B_w are the weighted background counts, (S_n+E_n) is the full energy of the capture γ-ray cascade, Φ_n is the neutron fluence intersecting the sample, f_{Norm} is a normalization factor and f_{Corr} is a correction factor. As mentioned above, the evaluation of the flux is ongoing, therefore a preliminary "flux" has been employed for the present work. Fig. 5 shows the ^{176}Yb(n, γ) reaction yield in the RRR interval from 0.1 to 1 keV.

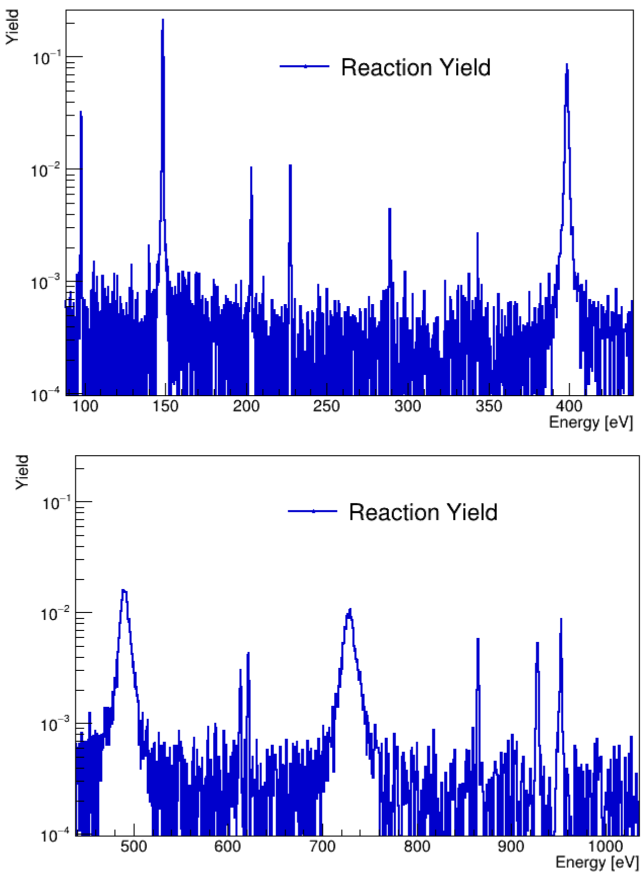

Fig. 5. The top plot shows the preliminary yield of the reaction from 100 eV to 450 eV, where the first ytterbium resonances are found. The bottom plot shows the preliminary yield of the reaction from 450 eV to 1 keV.

4 Conclusions

^{176}Yb capture cross section is required for an in-depth study of ^{177}Lu production, via the indirect route, in future high-intensity accelerators as IFMIF-DONES. The resonances region is of great interest for this purpose, with the corresponding set-up it will be possible to maximize the production in that energy region. However, the available experimental data only covers the neutron energies at thermal and in the range from ~5 to ~200 keV, respetively.

The present work will allow for a high-resolution measurement of the cross section from thermal up to several keV, covering the full range of the RRR. Finally, an in-depth study we will be able to extend our analysis into the URR.

In summary, a high-resolution measurement of the ^{176}Yb(n,g) cross section was succesfully performed at n_TOF EAR1 in 2021. The first preliminary experimental results presented here show that an accurate analysis of the resonace reigion will be possible.

The next steps towards the determination of the capture yield will include the calculation of several experimental correction factors, and an in-depth study of the capture g-ray cascades of ^{176}Yb+n. In parallel, the evaluation of the neutron flux of EAR1 is currently ongoing [25].

Finally, in the last step of the analysis the capture cross section will be extracted from the experimental yield by employing the R-Matrix bayesian code SAMMY [32].

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References

1. https://www.euro.who.int/en/health-topics/noncommunicable-diseases/cancer.
2. Nuc. Phys. Euro. Coll. Com.: nuclear physics for medicine. ISBN: 978-2-36873-008-9
3. Richard Van Noorden; Radioisotopes: The medical testing crisis; Nature vol. 504, pag. 202–204 (2013).
4. Paula Gold; Europe's isotope shortage will continue into 2009; Nature vol. 508, (2008).
5. NuPECc. Long Range Plan 2017 Perspectives in Nuclear Physics
6. https://ec.europa.eu/euratom/observatory_en.html
7. https://medicis.cern/
8. http://www.roadmap2018.esfri.eu/projects-and-landmarks/browse-the-catalogue/ifmif-dones/
9. J. Praena et al., EPJ Web of Conf., **239**, 23001 (2020)
10. Licia Uccelli, Petra Martini, et al; **24**, 640, (2019)
11. K. Kim et al., Nucl. Med. Mol. Imag., **52(3)**: 208–215, 2018
12. J. Zhang et al., Clin. Nucl. Med., January **45**, Issue 1, p e48-e50, 2020
13. A. Dash et al., Nuc. Med. and mol. 1Ima., **49(2)**: 85–107, (2015)
14. R. Henkelmann; Lu-177 production with a focus on radiation in the KBA at FRM II, SAAGAS 24, TUM Garching; Thursday, (2013)
15. E. A. M. Ruigrok et al., Eur. J. Nucl. Med. Mol. Imaging; (2020 Oct 23)
16. K. M. Ferreira et al., EPJ Web of Conf., **146**, 08002, (2017)
17. K.Wisshak et al., Phy. Rev., C **61 6** p.065801 (2000)
18. H. I. Liou et al., Phys. Rev. C **7**, 823, (1 February1973)
19. D. A. Brown et al., ENDF/B-VIII.0, Nucl. Data Sheets, **148**, pages 1-142, (2018),
20. A. J. M. Plompen et al., JEFF-3.3, Eur. Phys. J. A **56**:181, (2020)
21. C. Guerrero et al., Eur. Phys. Jour. A **49**, 27 (2013)
22. C. Weiß et al., Nucl. Inst. Methods A **799**, 90 (2015)
23. Raffaele Esposito and Marco Calviani; Jour. Neut. Res. **22** 221–231 (2020)
24. R. L. Macklin and J. H. Gibbons, Phys. Rev. 159, 1007 (1967)
25. M. Bacak et al., *Characterisation of the n_TOF/CERN 185m beam-line after the facility's major upgrades* (2022), ND2022 Conference, *EPJ Web of Conferences*
26. J. A. Pavón et al., *Characterisation of the n_TOF 20 m beam line at CERN with the new spallation target* (2022), ND2022 Conference, *EPJ Web of Conferences*
27. U. Abbondanno, G. Aerts et al., Nucl. Ins. and Meth, in Phys. Res. A **521** 454–467 (2004)
28. P. F. Mastinu et al.; New C_6D_6 detectors: reduced neutron sensitivity and improved safety, (The n_TOF Collaboration), CERN-n TOF-PUB-2013-002 (2013)
29. C. Domingo-Pardo et al., phys. rev. C **74**, 055802 (2006).
30. R. L. Macklin, J. Halperin and J. H. Gibbons, Nucl. Ins. and Meth, 164, 213-214 (1979).
31. P. Žugec et al., Nucl. Inst. Methods A **760**, 57 (2014)
32. N.M. Larsson. Updated User's Guide for SAMMY: Multilevel R-matrix Fits to Neutron Data Using Bayes Equations. Technical report, ORNL, 2006. (50, 130)