EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of the $^{35}\mathrm{Cl}(n,\gamma)$ cross section at n_TOF EAR1

[3/1/2018]

I. Porras[1], T. Wright[2], S. Bennett[2], F. Ogállar[1,3], J. Praena[1], M. Sabaté-Gilarte[3,4], P. Torres-Sánchez[1], F. Arias de Saavedra[1], C. Lederer-Woods[5], S.J. Lonsdale[5], R. Garg[5] , M. Dietz[5] and the n_TOF Collaboration.

[1]Universidad de Granada, Spain
[2]University of Manchester, UK
[3]European Organization for Nuclear Research, Switzerland
[4]Universidad de Sevilla, Spain
[5]University of Edinburgh, UK

Spokespersons:
Ignacio Porras (porras@ugr.es)
Tobias Wright (tobias.wright@manchester.ac.uk)
Technical coordinator:
Oliver Aberle (Oliver.Aberle@cern.ch)

Abstract

We propose to measure the $^{35}\mathrm{Cl}(n,\gamma)$ cross section for its importance in medical and nuclear fission applications. Firstly, chlorine is present in the brain therefore this reaction is important for the calculation of the radiation dose in Boron Neutron Capture Therapy of brain tumors. In addition to this, it is also present in the materials of fission reactors so is important for criticality calculations and predictions of the build up of the long-lived radionuclide $^{36}\mathrm{Cl}$. The experimental data available in the resonance region comes from a single capture measurement which covers the neutron energy range 0.1 to 500 keV. The behavior at low energies is fitted to the thermal value obtained from a different experiment with the assumption of a bound state near the neutron separation energy. The n_TOF facility allows a new measurement that can improve the knowledge of the cross section from thermal up to MeV neutron energies.

Requested protons: 2 x 10^{18} protons on target
Experimental Area: EAR1

Introduction and motivations

The accurate determination of the neutron capture cross section of ^{35}Cl is of dual interest, both in medical physics and in nuclear fission reactor studies. For the former, the $^{35}\mathrm{Cl}(n,\gamma)$ reaction is known to contribute noticeably to the radiation dose delivered to healthy tissue in Boron Neutron Capture Therapy of brain tumors.

Modern Boron Neutron Capture Therapy (BNCT) is facing a new era due to the most recent clinical results for brain tumors and head and neck cancers [1-3], performed up to now at research reactors, and the substantial change expected from the ongoing projects in accelerator based neutron sources (ABNS) for this therapy, that can be built in hospitals [4]. The BNCT community, by means of the working groups formed under the auspices of the IAEA and the International Society for Neutron Capture Therapy (ISNCT), is elaborating a new technical document with guidelines for the future practice of BNCT [5]. This includes a revision of the dose calculation system and treatment planning of BNCT.

A very important requirement for optimizing the therapy is the accurate determination of the dose delivered both to the tumor and to the healthy tissue. As BNCT is based on the selective uptake of boron from the malignant cells due to their enhanced metabolism (boron is delivered by means of a boronated aminoacid, boron-phenylalanine), the tumor dose is dominated by the energy released from the $^{10}\mathrm{B}(n,\alpha)$ reaction. However, the dose on healthy tissue comes from elastic scattering (mainly with hydrogen) and different neutron induced reactions with the materials present in tissue.

The dose delivered to healthy tissue is the limiting factor in a radiotherapy treatment. In BNCT, due to the complicated dosimetry of neutrons, which depends crucially on the isotope composition of the tissue, with different dose components delivered by diverse secondary particles, the treatment is planned with a safe margin, i.e. the equivalent dose delivered to healthy tissue is well below the maximum tolerated dose in conventional radiotherapy. Improvements in the determination of the delivered radiation dose, which requires accurate neutron cross sections and more precise relative biological effectiveness factors, should lead to a more precise dose planning and potentially a better therapeutic outcome.

All the BNCT clinical trials performed have used Monte Carlo based treatment planning systems, the most popular being NCTPlan and SERA, which make use of evaluated cross section data of ENDF/B-V and ENDF/B-VI. As new planning systems are being under development for BNCT, it is important to update the cross sections and to provide new accurate data.

Although it has received very little attention due to it being considered a minor component of human tissue, chlorine has an important role in the dose delivered by low energy neutrons, especially for brain tissue for which it is 0.3% by weight. We have studied its role, using recent evaluated data, by means of an in-house Monte Carlo simulation code validated through comparisons with MCNP. In Figure 1 the most important contributions to the dose in a human head phantom are displayed. We have found that the capture reaction on ^{35}Cl accounts roughly for 11% of the total dose for energies in the range 100 eV-10 keV, those used in BNCT.

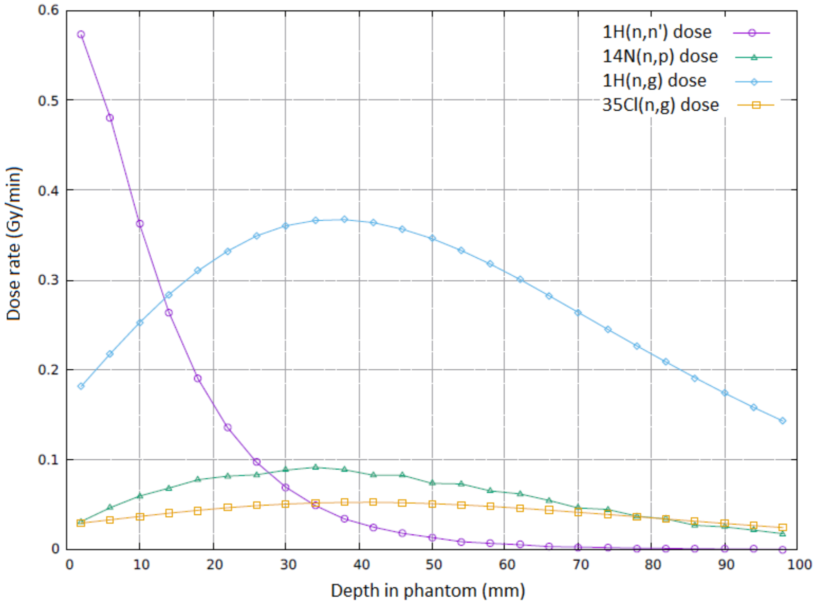

Figure 1. Depth distribution of the main components of the dose in brain tissue from a neutron beam of 10 keV, obtained from a Monte Carlo simulation code using the ENDF/VII.1 data for the cross sections. A Snyder head model phantom has been used. Similar contributions are found for other energies in the epithermal range.

As the neutrons are thermalized inside the body, the full range of energies from thermal to tens of keV is important for transport calculations and photon dose calculations in BNCT. The $^{35}\mathrm{Cl}(n,p)$ reaction has also been the subject of a recent measurement at n_TOF (EAR-2) [6] motivated by its contribution to the charged particle dose in BNCT.

In addition, the $^{35}\mathrm{Cl}(n,\gamma)$ reaction is of interest for nuclear reactors. ^{35}Cl is present in small amounts (2-5 ppm in weight) in fuel cladding, the amount dependant on the reactor and also as an impurity (<2 ppm in weight) in graphite, which has been used as a moderator in many nuclear reactor designs. By means of this reaction, during the reactor lifetime, the radionuclide ^{36}Cl is produced which has a long half-life of ~300,000 years [7]. Many nuclear fuel cycles are now going towards the so called ‘closed fuel cycle/once through fuel cycle’ where the nuclear waste is buried underground in a repository. As an example, in the UK, this repository would likely be surrounded by clay and therefore in the long term future, it may end up with pockets of waste saturated with water containing any soluble long lived radionuclides. Chlorine is water-soluble therefore poses a possible future contamination problem, especially if for example future generations mined these areas for drinking water. In order to accurately predict the amount of ^{36}Cl in irradiated fuel, the $^{35}\mathrm{Cl}(n,\gamma)$ cross section must be known. Now that countries such as the UK are planning their waste repositories, it is very timely that a measurement of the $^{35}\mathrm{Cl}(n,\gamma)$ cross section in the resonance region is performed in order to give confidence to the current data and improve its accuracy.

Finally, ^{35}Cl has an impact on criticality safety computations on nuclear reactors. This has been the main motivation for previous measurements [8].

Current knowledge of the $^{35}\mathrm{Cl}(\mathrm{n},\gamma)$ cross section

The most recent evaluation of the ^{35}Cl total and partial cross sections is based on the *R*-matrix analysis of Sayer et al. [8], which ranges up to 1.2 MeV. However, there is only one measurement of the Cl-nat (n,γ) capture reaction (Guber et al [9]) which covers some of the resonance region in the range 0.1 to 500 keV. The quality of the data available in the resonance region limits the accuracy of the cross section to ~10%. This has led to discrepancies in the cross section evaluations across the major libraries (see attached figures).

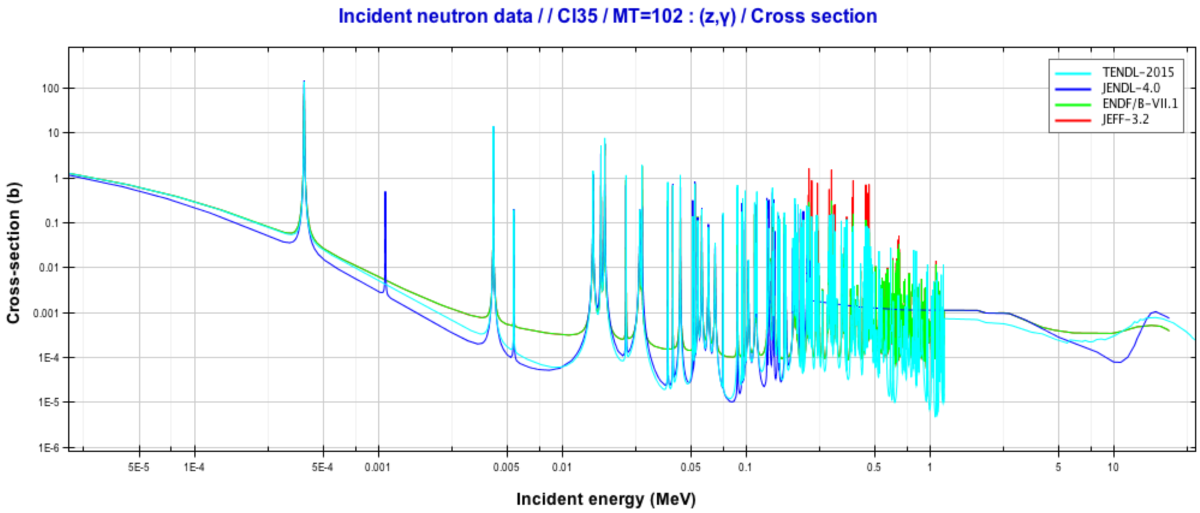

Figure 2. Comparison of evaluated data for the reaction $^{35}\mathrm{Cl}(n,\gamma)$ from the major nuclear data libraries.

In addition to this, there is one low energy measurement by Kashukeev [10], between 0.022 and 1 keV. The cross section below the first resonance has been obtained assuming a bound state in ^{36}Cl below the neutron separation energy for which there is empirical evidence [11]. The parameters for this state have been fixed in order to match the thermal cross section value, obtained from a different measurement [12].

The low energy behaviour may have an important impact in BNCT dose calculations for healthy tissue. As a test, we have computed, by means of a Monte Carlo simulation, the total absorbed dose in brain from different estimations of the cross section at low energies, as shown in Figure 3. The upper and lower bounds have been obtained by assuming a 1/v behaviour of the cross section, in the first case assuming the highest measurement of the thermal value (44 b) and in the second case from the minimum value of the cross section before the resonance. It is expected that the real cross section values lie between both bounds. However, the lower bound takes a value at the thermal point (6.7 b) which is 4 times smaller than the smallest reported measurement (30 b) and seems unrealistic. For this reason we have tested other estimations of the cross section with non 1/v behaviour due to the presence of a bound state. Assuming a negative energy resonance at -75 eV (as reported in [11]) or -180 eV (as reported in [8]), we can match the two different thermal values reported respectively (estimations 1 and 2 in Figure 3). With all these cross section values, we have performed a Monte Carlo calculation of the dose delivered in healthy tissue in a BNCT treatment of a brain tumor. For a reference therapeutic neutron flux (10^{10} n s^{-1} cm^{-2}) in the epithermal range (up to 10 keV) important differences are found. For example, for 100 eV neutrons, the total dose obtained from the different estimations range between 3.766 and 4.183 mGy/s. If we remove the (rather unrealistic) lower bound, the variations are within 3.966 and 4.186 mGy/s, still above the maximum 5% uncertainty recommended by the ICRU [13].

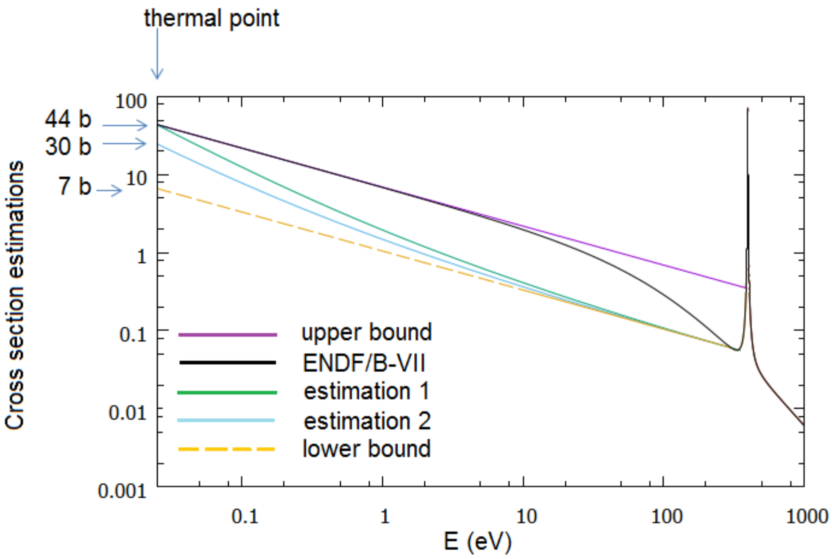

Figure 3. Different cross sections for the radiative capture of ^{35}Cl used as a test for the determination of the incidence in the total dose delivered in normal brain.

Therefore, the interest of the new measurement is two-fold: (i) in the low energy range (from thermal to the first resonance, at 397 eV) in order to test the assumptions of the current evaluation and provide reliable values of the cross section for BNCT dosimetry, and (ii) to expand the data of the resonance region which is important for criticality calculations in fission reactors and predicting amounts of ^{36}Cl present in nuclear waste. In addition to this, checking the values of the only measurement performed in the range (1 keV – 500 keV), and providing high resolution data in this important region for the safety calculations aforementioned is also an argument in favour of this measurement, which will compliment the data obtained from the (n,p) measurements recently performed at n_TOF.

Proposed set up and counting rate estimation

The sample will be a solid disc of pure Na^{35}Cl, made by pressing the salt that can be found in the market at 99% enrichment. This will avoid the need to correct for the contribution from the ^{37}Cl isotope, which was present in the previous measurements [9,10], both performed with natural chlorine. The setup proposed in EAR-1 consists of four C_6D_6 detectors, with low sensitivity to neutrons.

The efficiency of this setup is of the order of 15%. With this assumption, and with the current data available for the capture cross section, we have made an estimation of the counting rate which is shown in Figure 4. The background has been estimated from previous measurements in the same area. With 2x10^{18} protons, very good statistics for the low energy region as well as for the resonances is found. We have enough statistics so we can use part of these protons for background measurements and normalization with an Au sample.

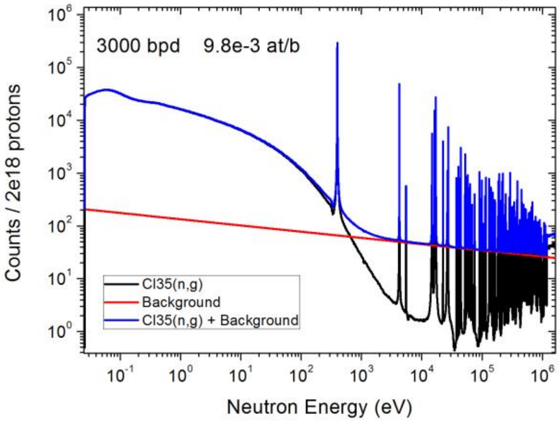

Figure 4. Counting rate estimation of the measurement at EAR-1 with a sample of 9.8×10^{-3} atoms per barn of ^{35}Cl, displayed with 3000 bins per decade and with the assumption of 15% efficiency.

Summary of requested protons:

2×10^{18} protons (1.5×10^{18} for measurement and 0.5×10^{18} for background and normalization)

References:

[1] R.F. Barth, M.G.H. Vicente, O.K. Harling et al., Current status of boron neutron capture therapy of high grade gliomas and recurrent head and neck cancer, *Radiation Oncology* **7,** 146 (2012).

[2] Nuclear Physics European Collaboration Committee (NUPECC), Nuclear Physics for Medicine, ESF Report: http://www.nupecc.org/pub/npmed2014.pdf

[3] International Atomic Energy Agency (IAEA), press release: https://www.iaea.org/newscenter/news/boron-neutron-capture-therapy-back-in-limelight-after-successful-trials

[4] A.J. Kreiner, J. Bergueiro, D. Cartelli et al., Present status of Accelerator-Based BNCT. *Rep Pract Oncol Radiother*. **21**, 95-101 (2016).

[5] ISNCT forum: http://www.bnct.it/forum

[6] J. Praena et al., The ^{14}N(n,p)^{14}C and ^{35}Cl(n,p)^{35}S reactions at n_TOF-EAR2: dosimetry in BNCT and astrophysics, CERN-INTC-2017-039; INTC-P-510.

[7] R. Mills, Z. Riaz, A. Banford, Nuclear Data issues in the calculation of ^{14}C and ^{36}Cl in irradiated graphite, ENC 2012 Conference proceedings.

[8] R. O. Sayer, K. H. Guber, L. C. Leal et al., R-matrix analysis of Cl neutron cross sections up to 1.2 MeV, *Physical Review* **C 73**, 044603 (2006)

[9] K. H. Guber, R. O. Sayer, T. E. Valentine et al., New Maxwellian averaged neutron capture cross sections for ^{35,37}Cl, *Physical Review* **C 65**, 058801 (2002).

[10] N. T. Kashukeev, Yu. P. Popov, and F. L. Shapiro, *J. Nucl. Energy* A/B **14**, 76 (1961).

[11] C. T. Hibdon and C. O. Muehlhausk, Negative Energy Neutron Resonance in Cl^{35}, *Physical Review* 79, 44 (1950).

[12] G.L. Molnár, Zs. Révay and T. Belgya, Accurate absolute intensities for the ^{35}Cl(n,γ) reaction gamma-ray standard, *Nucl. Instrum. Meth. Phys. Res. B*, **213,** 32 (2004).

[13] International Commission on Radiation Units and Measurements (ICRU), Dose Specification for Reporting External Beam Therapy, ICRU Report 29 (1978).