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Measurement of the ^{33}S(n,α) cross-section at n_TOF(CERN): Applications to BNCT

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ABSTRACT

Aim: The main purpose of this work is to present a new (n,α) cross-section measurement for a stable isotope of sulfur, ^{33}S, in order to solve existing discrepancies.

Background: ^{33}S has been studied as a cooperating target for Boron Neutron Capture Therapy (BNCT) because of its large (n,α) cross-section in the epithermal neutron energy range, the most suitable one for BNCT. Although the most important evaluated databases, such as ENDF, do not show any resonances in the cross-section, experimental measurements which provided data from 10 keV to 1 MeV showed that the lowest-lying and strongest resonance of ^{33}S(n,α) cross-section occurs at 13.5 keV. Nevertheless, the set of resonance parameters that describe such resonance shows important discrepancies (more than a factor of 2) between them.

Materials and methods: A new measurement of the ^{33}S(n,α)^{30}Si reaction cross-section was proposed to the ISOLDE and Neutron Time-of-Flight Experiments Committee of CERN. It was performed at n_TOF(CERN) in 2012 using MicroMegas detectors.

Results: In this work, we will present a brief overview of the experiment as well as preliminary results of the data analysis in the neutron energy range from thermal to 100 keV. These results will be taken into account to calculate the kerma-fluence factors corresponding to ^{33}S in addition to ^{10}B and those of a standard four-component ICRU tissue.

Conclusions: MCNP simulations of the deposited dose, including our experimental data, shows an important kerma rate enhancement at the surface of the tissue, mainly due to the presence of ^{33}S.

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1. Background

The most recent reactors and the proposed accelerator-based neutron sources produce epithermal neutron beams, in the keV range, which are considered the most suitable ones for Boron Neutron Capture Therapy (BNCT). A cooperative target to ^{10}B showing resonant capture in this range may enhance the dose for superficial tumors. For this purpose, ^{33}S, a stable isotope of sulfur which represents 0.75% of ^{nat}S, has been studied by Porras[1] because of its large (n,α) cross-section in the keV range, much greater than the (n,γ) one. Porras already showed an enhancement of absorbed dose due to the cooperating effect of ^{33}S and ^{10}B by means of Monte Carlo simulations.[2]

There are some discrepancies, more than a factor of 2, between experimental ^{33}S(n,α)^{30}Si cross-section measurements with respect to the resonance parameters available in the evaluated nuclear data files.[7] Only one dedicated measurement of ^{33}S(n,α) cross-section can be found in the literature[3] since the other measurements do not provide new specific data for BNCT.[4,5] Such measurement provided data from 10 keV to 1 MeV, thus the cross-section is unknown for neutron energies below 10 keV, the most important range for BNCT since neutrons are moderated by the tissue. Furthermore, there is only one measurement of the (n,tot) cross-section.[6] Both experiments showed that the lowest-lying and strongest resonance of ^{33}S(n,α) cross-section occurs at a neutron energy near 13.5 keV. The discrepancies between the resonance parameters obtained[3,5,6] amounted to a factor of 2, being the results of Wagemans et al.[3] the lowest ones. Moreover, the main evaluated nuclear data libraries,[7] such as ENDF/B-VII.1, JEFF-3.1.2, JENDL-4.0, ROSFOND-2010 and JEF-2.2, do not even describe the experimentally found resonant structure. Only EAF-2010 shows the resonances but the values are smaller than those reported by Wagemans et al.[3] Within this framework, a new measurement at the n_TOF facility at CERN was proposed to clarify the existing discrepancies.

2. Aim

Since Monte Carlo (MC) simulation is a standard tool employed to describe neutron transport and to study the dose delivered to tissue, preliminary results of the ongoing ^{33}S(n,α) cross-section data analysis from the last measurement at n_TOF (CERN) will be shown and taken into account to calculate the kerma-fluence factors as well as the kerma rate corresponding to ^{33}S in addition to ^{10}B for a standard four-component ICRU tissue.

3. Materials and methods

3.1. *^{33}S(n,α)^{30}Si measurement at n_TOF(CERN)*

The neutron time-of-flight facility (n_TOF) at CERN, based on a spallation neutron source, is mainly dedicated to measure neutron-induced cross sections, (n,γ) and (n,f), for nuclear technology, astrophysics and basic nuclear physics applying the time of flight technique.[8] In November 2011, a test of the set-up to measure (n,α) cross-section with MicroMegas detectors at n_TOF was carried out.[9] The success of the test led to the first (n,α) cross-section measurement, not related to monitoring purposes, at n_TOF[10] in 2012, preliminary results of which will be presented in this paper.

n_TOF is a neutron facility which provides a neutron beam of energy from thermal to 1 GeV and the energy resolution at 10–100 keV is $\Delta E/E \sim 10^{-3}$. These features let experimental data to be extended below 10 keV. On the other hand, a high energy resolution is required to solve the discrepancies in resonance parameters among evaluations and EXFOR[3,5] data. A fast ionization chamber based on 10 MicroMegas detectors[11] was used to collect the energy deposited by the emitted alpha particles from six ^{33}S samples, two blank samples to characterize the background and two ^{10}B$_4$C samples. The use of the ^{10}B(n,α)^{7}Li cross-section, a standard in the neutron energy range of interest, as a reference allows to extract the relative cross-section and avoid systematic uncertainties specific to

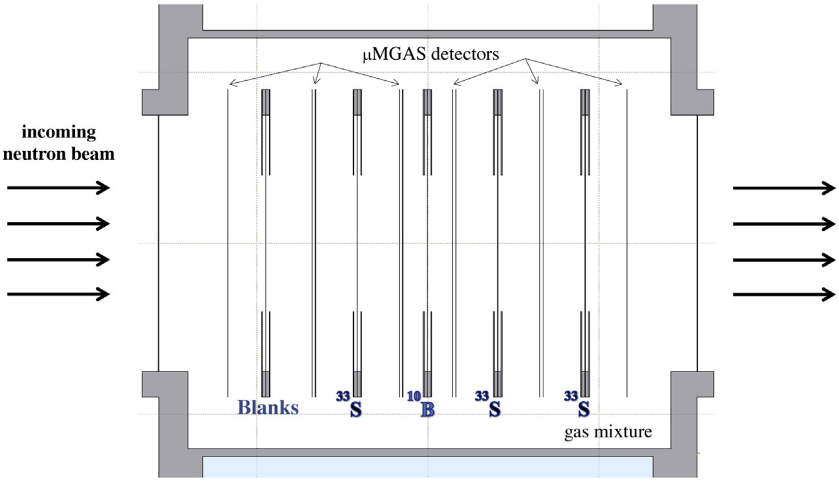

Fig. 1 – Experimental set-up used to measure the ^{33}S(n,α) cross-section at n_TOF(CERN). Several MicroMegas detectors can run in parallel intercepting the beam because they are transparent to neutrons.

the facility. All samples were coated at CERN. A new manufacturing method was developed to make samples more stable; ^{33}S was evaporated onto Cu–Ti–Cr film. Moreover, this method solves the mass loss in previous experiments due to sublimation.[3] Another advantage of the present set-up is the possibility to study the reaction anisotropy since all samples were mounted in a back-to-back configuration (see Fig. 1).

3.2. ^{33}S *as a cooperating target in BNCT*

Following previous works of the authors,[1,2,12] a set of MC simulations with MCNP code[13] of the neutron transport in tissue was carried out to evaluate the kerma rate (which approximates the absorbed dose rate) as a function of depth. A circular neutron beam with 10 cm in diameter, 13.5 keV in energy and a flux of 10^{10} cm^{-2} s^{-1} was propagated along the central axis of a cylindrical phantom of 10 cm in height and 10 cm in radius filled with a standard ICRU 4-component tissue (10.12% H, 11.10% C, 2.60% N and 76.18% O). This material has the same density as water and it can mimic normal human tissues. Relevant processes for the estimation of the kerma-fluence factors are elastic scattering of neutrons on ^{1}H, ^{12}C, ^{14}N and ^{16}O and ^{14}N(n,p), ^{10}B(n,α) and ^{33}S(n,α) reactions. Since radiative capture is comparatively negligible in the keV neutron energy range of interest and inelastic scattering becomes important for neutron energies above 1 MeV, these processes are not taken into account in the simulation. Moreover, electrons produced in the ionization are not considered because the simulation is focused on the local radiation effect, i.e., inside the tumor cells. The cross-sections needed to evaluate those factors are taken from ENDF/B-VII.1[7] data base for each process except for ^{33}S for which experimental cross-section data from Wagemans et al.,[3] the most conservative ones, and from this work are considered.

4. Results

Fig. 2 shows a preliminary ^{33}S(n,α) cross-section in the neutron energy range from 100 eV to 100 keV. Data below 10 keV are presented for the first time. It is confirmed that resonances start at around 13.5 keV with the strongest one. The ongoing analysis is preliminary and its results must still be corrected by several experimental factors which include uncertainties. A comparison between available experimental data[3,5] and this work shows an increase of 30% in height (with similar width) for the first resonance (13.5 keV), although the second one is around 40% (24 keV) smaller in this work. Nevertheless, we have to calculate the resonance parameters to solve the existing discrepancy.

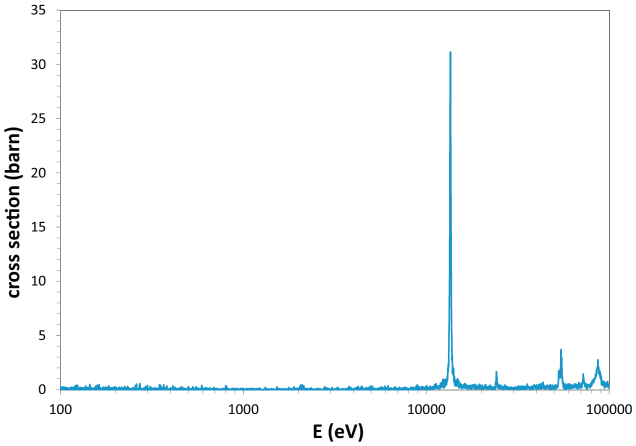

Fig. 2 – Preliminary ^{33}S(n,α) cross-section in the neutron energy range from 100 eV to 100 keV measured at n_TOF(CERN) in 2012. Results are shown without error bars because the analysis error is not finished.

Fig. 3 shows the kerma-fluence factors calculated[14–17] and Fig. 4 illustrates the results of the simulations for four different cases to estimate the effect of ^{33}S as a cooperating target for BNCT: normal tissue (ICRU 4-component material); tissue with a typical concentration of ^{10}B in treatment (20 μg/g) and a mixture of 20 μg/g of ^{10}B and 10 mg/g of ^{33}S considering data

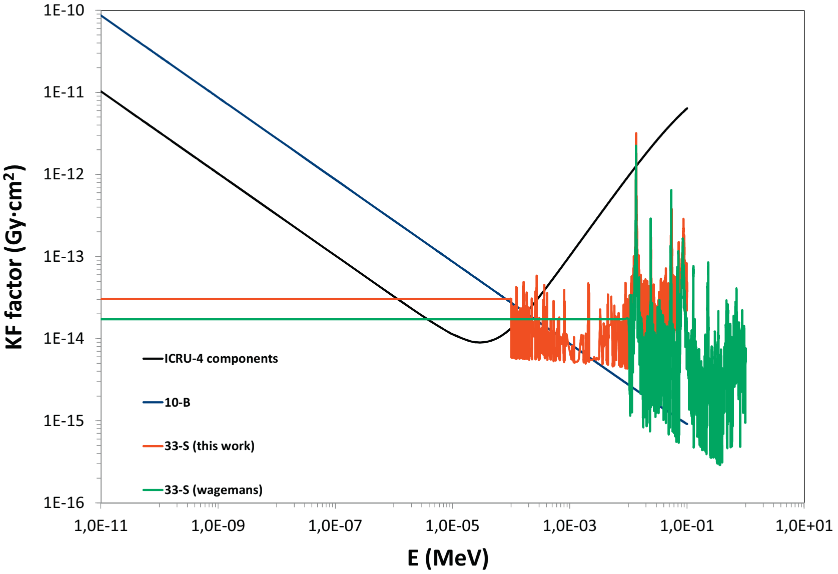

Fig. 3 – Kerma-fluence factors calculated[14–17] for ICRU-4 component material (ENDF/B-VII.1), ^{10}B (ENDF/B-VII.1) and ^{33}S (Wagemans et al.[3] and this work).

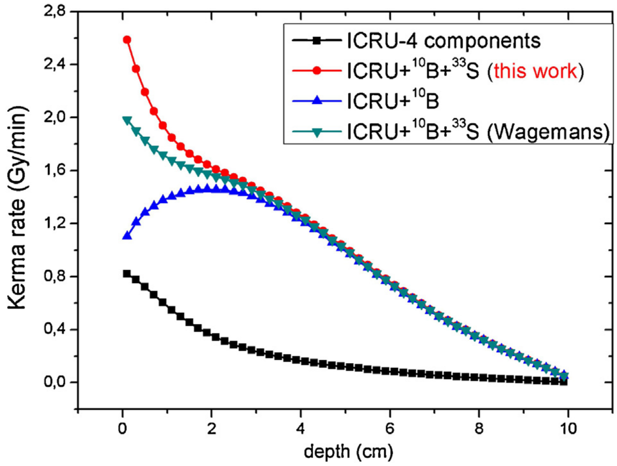

Fig. 4 – Kerma rate as a function of the depth for ICRU-4 component tissue with different concentrations of ^{33}S and ^{10}B: normal tissue (black squares), tumor with 20 μg/g of ^{10}B (blue triangles), tumor with 10 mg/g of ^{33}S and 20 μg/g of ^{10}B using experimental data from Wagemans et al.[3] (green inverted triangles) and from this work (red circles). A superficial improvement of the kerma rate due to the presence of ^{33}S in tissue is shown.

from Wagemans et al.[3] and from this work, all these cases representing tumor. The concentration of sulfur was chosen as that in previous works,[1,2,12] A cooperative effect is observed as well as a superficial improvement, from 0 to 2 cm, is found on the kerma rate mainly due to the presence of ^{33}S in tissue in addition to an enhancement of 10% compared with Wagemans et al.[3] data.

5. Conclusions

A new experimental measurement of ^{33}S(n,α)^{30}Si cross-section was performed at n_TOF(CERN). Although the data analysis is ongoing, we found higher cross-section values at 13.5 keV, the first and strongest resonance, in comparison with the existing data available. This preliminary result was included in Monte Carlo simulations (MCNP) to study the effect of ^{33}S as cooperative capturer for BNCT in ICRU 4-component tissue considering 13.5 keV neutron energy beam. An important kerma rate improvement at the surface of the tissue (0–2 cm) was found, mainly due to the presence of ^{33}S. An enhancement of the 10% has been found respect to the Wagemans et al.[3] data. Finally, it is remarkable that we showed for the first time data below 10 keV neutron energy for ^{33}S(n,α) cross-section.

Conflict of interest

None declared.

Financial disclosure

None declared.

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