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Measurement of the ^{197}Au(n,γ) stellar cross section at kT=30 keV by activation.

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The Maxwellian-averaged cross-section (MACS) of ^{197}Au(n,γ) at kT=30 keV is used in Nuclear Astrophysics as a reference for activation measurements relevant to the s-process. The traditionally adopted value, (582±9) mb, corresponds to an accurate activation measurement carried out by Ratynski & Käppeler in 1988. Recently, new measurements with the time-of-flight technique have reported values about 5 % higher than Ratynski&Käppeler. Moreover, the evaluations of different data base provide values from 613-618 mb. In our work we have measured the MACS of ^{197}Au(n,γ) at kT=30 keV by activation, at 3 MV Tandem Pelletron accelerator at CNA (Seville). We have found a value equal to (626±25) mb. We describe the experiment and we discuss the analysis, including MCNPX simulations and a correction proposed for flat samples, and a comparison with previous and recent measurements.

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1 Speaker

1. Introduction.

Neutron capture processes (s-process and r-process) are the main responsible for the nucleosynthesis of the main part of the heavy elements beyond iron [1]. In the helium-burning phase in red giants stars, neutrons are produced via (α,n) reactions. Neutrons are quickly thermalized through elastic scattering, and their velocities are represented by a Maxwell-Boltzmann distribution (or maxwellian in unit of energy). A Maxwellian- averaged cross-section (MACS) or stellar cross-section of the involved isotopes is used as a key parameter for modeling the stellar nucleosynthesis processes. This MACS can be calculated analytically at any kT if the differential neutron-capture cross-section, measured as a function of the energy (i.e. using time-of-flight technique) is known (Eq.1).

$$MACS \equiv \langle \sigma \rangle = \frac{2}{\sqrt{\pi}} \cdot \frac{\int_0^\infty \sigma(E) \cdot E \cdot e^{-\frac{E}{kT}} dE}{\int_0^\infty E \cdot e^{-\frac{E}{kT}} dE} \qquad (1)$$

However, as shown by Beer & Käppeler [2], MACS at kT≈25 keV can be measured almost directly for many isotopes using activation technique. In this method, a quasi-Maxwellian neutron spectrum is generated using the $^{7}Li(p,n)^{7}Be$ reaction. The best approach to a Maxwell spectrum is achieved when the proton energy is adjusted to E_p=1912 keV [3]. With the criterion of overlapping of areas the optimal kT was selected equal to 25 keV. After irradiating the sample, if the nucleus produced by the neutron capture reaction is radioactive (with a convenient half-life), the subsequent gamma activity measurement will allows us to obtain the experimental neutron capture cross section. This experimental cross section is then a quasi-Maxwellian averaged cross section, and this value must be corrected later, taking into account the different between the quasi-maxwellian spectrum and a true Maxwellian one. In addition to this, if the MACS at kT=30 keV is needed an extrapolation from 25 to 30 keV is necessary. For both, correction and extrapolation, the knowledge or assumption of the cross-section as a function of the energy is mandatory. This activation method has been extensively used for MACS measurements of many isotopes relevant to the s-process [4].

A temperature of $T=348 \cdot 10^6$ K, typical of helium-burning in red giants stars, corresponds to an energy of kT=30 keV. For activation measurements, the MACS of $^{197}Au(n,\gamma)$ at kT=30 keV is used as a reference [5]. Its traditionally adopted value was obtained by Ratynski & Käppeler, in an accurate activation measurement using a spherical segment gold sample [3]. They reported a value equal to (582±9) mb. Recently, new measurements of the $^{197}Au(n,\gamma)$ cross section with time-of-flight (TOF) technique at n_TOF facility at CERN reported a value of (611±22) mb [6]. The evaluations performed in different data base show values ranging from 613 to 618 mb [7]. In this work, we have measured the MACS of $^{197}Au(n,\gamma)$ at kT=30 keV by activation at the Centro Nacional de Aceleradores (CNA). A gold flat sample was used and a so-called "flat sample correction" is proposed and is used for the comparison with Ratynski & Käppeler.

2. Experimental procedure.

2.1 Experimental setup and activation.

The activation was carried out at the 3MV Tandem Pelletron accelerator at CNA (Seville). The proton energy was adjusted to E_p=1912 keV. The accelerator terminal and the 90° analyzing magnet were carefully calibrated before the experiment, using the 991.86 keV $^{27}Al(p,\gamma)^{28}Si$ and 2409 keV $^{24}Mg(p,p'\gamma)^{24}Mg$ resonances, and with the Rutherford backscattering technique for alpha beam of higher terminal voltages. The energy spread for proton beam at these energies was less than 1 keV.

The setup consisted of a copper backing that held the metallic lithium target and the gold sample. The dimensions of the backing were 3x3x0.8 cm^3 with a centered cylinder hole of 1 cm in diameter and 0.75 cm in height to place the thick lithium layer (of 380 μm thickness). The backing contains an internal cooling water circuit (centered toroid). The gold sample, 2.5cm diameter and 0.1mm thickness, was secured to the back surface of the backing. The copper backing between the lithium layer and the gold sample was 0.5 mm thickness. At the proton beam energy of the experiment, E_p=1912 keV, all the neutrons produced by the $^7Li(p,n)$ reaction are kinematically collimated into a forward cone of about 140^o opening angle. So, except for scattered neutrons at high angles, the entire neutron flux is completely impinging on the gold sample. Metallic lithium reacts both with nitrogen and oxygen at room temperature. Therefore, the lithium foil was always stored and manipulated in inert gas to avoid its degradation. Neutron irradiation time was 4 h 45 min. During the irradiation, neutron dosimetry and proton current on the target was recorded in order to correct possible beam instabilities.

2.2 Activity measurement.

The produced ^7Be and ^{198}Au have a half-life of 53,29 and 2,6947 days respectively. After the irradiation, a HPGe Ortec GMX gamma detector was placed at 28 mm distance from the backing, in order to measure the gamma-rays lines of 477,6 keV and 411,8 keV emitted in the decay of the beryllium and gold, respectively. Activity was measured during 6 h 30 min. Time between end of irradiation and beginning of activity measurement was 15 min.

3. Data analysis.

The cross section can be deduced if the number of activated nuclei *A*, the neutron fluence through the sample Φ, and the sample mass thickness *N* are known (eq. 2).

$$\sigma_{exp} = \frac{1}{N_{Au}} \cdot \frac{A_{Au}}{\Phi} \quad (2) \qquad \sigma_{exp} = \frac{1}{N_{Au}} \cdot \frac{C_{Au}}{C_{Be}} \cdot \frac{[f_d \cdot I_\gamma \cdot \varepsilon \cdot K_\gamma]_{Be}}{[f_d \cdot I_\gamma \cdot \varepsilon \cdot K_\gamma]_{Au}} \cdot \frac{1}{K_n} \cdot \frac{1}{K_f} \quad (3)$$

A, the number of activated nuclei, can be determined from the γ-ray spectra measured with the HPGe detector. In eq 3, *C* is the number of events registered by the detector, I_γ is the gamma intensity of the 477,6 keV and 411,8 keV lines, corresponding to 10,44% and 95,54% respectively; ε is the HPGe gamma detector efficiency (including energy efficiency and

geometric efficiency that takes into account the extended samples effects); K_γ is the correction due to the gamma absorption and scattering in materials, before the gammas reach the detector; K_n reflects the neutron scattering (meanly due to Cu backing); K_f is a correction due to the flat sample used in our experiment. It must be noticed that the Au sample thickness as seeing by neutrons passing through Au depends on the entering angle θ inside the sample. Ratynski & Käppeler [3] used a semi-spherical sample to appear equally thick to all neutrons. The factor f_d relates the number of the decay nuclei during the measurement time (t_m) with the total number of activated nucleus (during irradiation time, t_a). It also includes the decay nuclei during the waiting time (t_w), thus the time between the end of neutron irradiation and the beginning of activity measurement with the HPGe detector, and $\phi(t)$ is the neutron flux (eq. 4).

$$f_d = \frac{e^{-\lambda t_a}\cdot\left(\int_0^{t_a}\phi(\tau)\cdot e^{\lambda\tau}d\tau\right)}{\int_0^{t_a}\phi(\tau)\cdot d\tau}\cdot\left(e^{-\lambda t_w}\right)\cdot\left(1-e^{-\lambda t_m}\right) \qquad (4)$$

C_{Au} and C_{Be} were counted using Canberra GENIE2000 software (fig 1) connected to the acquisition system consisting of HPGe, a high voltage supply HVPS 9645 Canberra and digital signal processor DSP 9660 Canberra.

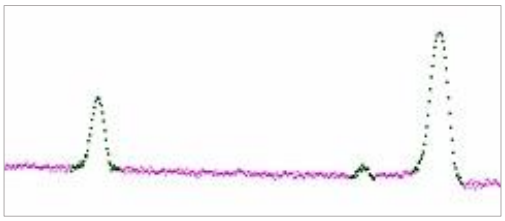

Figure 1: Au and Be gamma peaks (Genie2000).

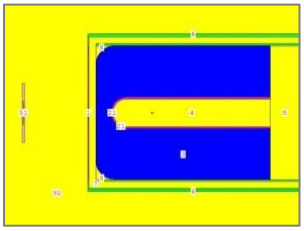

Figure 2: MCNPX simulation.

Mass thickness N_{Au} was obtained with accurate measurements of mass (0.3820 g) and surface (206,12 mm^2) of the gold sample, cutting the gold sample after irradiation for a more precise measurement of the activated region. The curve of the gamma energy efficiency of the HPGe was first experimentally calculated using calibrated point sources. Then, a series of MCNPX simulations for those point sources and the HPGe detector were carried out, adjusting the geometry and dead layer thickness of the germanium crystal from its nominal value provided by the manufacturer to a value that fits better with the experimental point sources efficiency. This allowed getting a more reliable modelling of the HPGe [8] for a subsequent full MCNPX simulation (fig 2) to calculate ε and K_γ.

For the K_n calculation, a Fortran code [9] was used to generate the energy-angle distribution of the neutrons for the ^{7}Li(p,n) reaction, as the input for MCNPX. The code and the MCNPX simulations for the proton energy (1912 keV) involved in this experiment were tested with experimental data, see details in [10]. For K_f calculation, the angle-energy distribution of the neutron spectrum $\phi(\theta,E)$ and the energy dependence of the ^{197}Au(n,γ) cross section must be taken into account. We propose this analytical calculation for K_f (eq. 5):

$$K_f = \frac{\iint \sigma(E)\cdot\frac{1}{\cos\theta}\cdot\phi(\theta,E)\,dEd\theta}{\iint \sigma(E)\cdot\phi(\theta,E)\,dEd\theta} \qquad (5)$$

Finally, in order to obtain the MACS, we need to correct our experimental cross-section (σ_{exp}) due to the difference between a quasi-maxwellian neutron spectrum and a true maxwellian neutron spectrum at kT=25 keV (eq. 6 and 7), so-called K_{max} factor.

$$MACS = \frac{2}{\sqrt{\pi}} \cdot K_{max} \cdot \sigma_{exp} \quad (6) \qquad K_{max} = \frac{\int_0^\infty \sigma(E)\cdot E\cdot e^{-\frac{E}{kT}} dE \Big/ \int_0^\infty E\cdot e^{-\frac{E}{kT}} dE}{\int_0^\infty \sigma(E)\cdot \phi(E) dE \Big/ \int_0^\infty \phi(E) dE} \quad (7)$$

To perform the calculation of K_{max} and K_f we needed the experimental neutron spectrum and the energy-dependence of the cross-section. For the neutron spectrum, few experimental measurements are available [6,11,12]. For the gold cross-section we used ENDF/B-VII.1. For kT=25 keV, we find this correction to be K_{max25}= 0,955. Taken into account that energy dependence of the cross-section, we can extrapolate a MACS calculated for kT=25 to MACS at kT=30 keV, resulting $MACS_{(kT=30)}$=0,901*$MACS_{(kT=25)}$.

We resume the experimental data and corrections in table 1.

	Value	Uncert.(%)
C_{Au}	32950	0,7
C_{Be}	332000	0,3
f_{dAu}	0,06718	0,01
f_{dBe}	0,00353	0,01
$K_{\gamma Au}$	0,9801	0,3
$K_{\gamma Be}$	0,9286	0,3

	Value	Uncert.(%)
$\varepsilon_{Be}/\varepsilon_{Au}$	0,834	1,8
N_{au}	5,67E-4 at/b	1,5
K_f	1,263	2
K_n	0,977	0,3
K_{max25}	0,955	1,5
Total uncert.	≈ 4%	

Table 1.Experimental data and corrections

4.Results and conclusions

We obtain for the MACS (at kT=30) a value of (626±25) mb. This value is in good agreement with n_TOF differential measurement (611±22) mb, the ENDF/B-VII.1 evaluation value (614 mb), a very recent TOF measurement performed with a truncated neutron spectrum from 3.5 to 84 keV at GELINA 613 mb [13], and it is higher than the reference Ratynski & Käppeler activation measurement [5]. Our experimental cross section at kT=25 keV, σ_{exp}= (645±25) mb, is also consistent with the result provided in recent activation measurements, (616±17) mb [12].

Comparing with Ratynski & Käppeler experiment [5], apart from the flat sample correction, which in principle was not necessary for the R&K measurement due to the spherical Au sample (although the neutron source wasn't perfectly punctual), we found an important difference for the estimated neutron scattering term K_n. For our setup, with 0.5 mm thick Cu backing between Li and Au, with MCNPX we checked that the distortion in the spectrum is not significant, and we found a total of 2,3% neutron loss due to scattering. However, Ratynski &

Käppeler indicates only 1,8% neutron loss at the Au sample for a 1 mm thick Cu backing. We performed a MCNPX simulation of the Ratynski & Käppeler setup and the calculation indicates more than 5% neutron loss for a 1 mm thick Cu backing. Therefore, one of the possible factors contributing to the difference between Ratynski& Käppeler measurement and recent ones could be an underestimation of this neutron loss.

New measurements are on-going at CNA to confirm this result, including measurements with an improved method of generation of maxwellian neutron spectra as showed in [9]. With a controlled proton energy-shaped beam it is possible to generate a true maxwellian spectrum at different kT from 30 to 60 keV [14].

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