AMS measurement of the reaction $^{35}Cl(n,g)^{36}Cl$ and its relevance to astrophysics and nuclear technology

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Abstract

^{36}Cl is a long-lived radionuclide ($t_{1/2}$ = 301000 a), which is dominantly produced via the reaction $^{35}Cl(n,\gamma)^{36}Cl$. The seed nuclei of this reaction, the stable ^{35}Cl, acts as a neutron poison in the nucleosynthesis processes during later burning phases of stars. This makes the reaction important for astrophysical calculations, aiming to reproduce the abundances of elements. Due to the long half-life of ^{36}Cl, the cross section and the production rate of the above reaction are also important for nuclear technology and nuclear waste management.

The two main goals of this work are: (i) the production of an independent $^{36}Cl/^{35}Cl$ standard for accelerator mass spectrometry (AMS); and (ii) the determination of the Maxwellian averaged cross section (MACS) of $^{35}Cl(n,\gamma)^{36}Cl$ at 25 keV using AMS.

Approaching the first goal, NaCl pellets were irradiated at the TRIGA Mark II reactor at the ATI in Vienna and at the Budapest research reactor. The neutron flux was monitored via the reference cross section of $^{197}Au(n,\gamma)^{198}Au$ (gold foils attached to and gold powder homogenously mixed into the pellets) and determined by activity measurements on the foils and the pellets. With this data we calculated a $^{36}Cl/^{35}Cl$ ratio for the irradiated samples.

The AMS measurements on these samples were performed at VERA (Vienna Environmental Research Accelerator). To determine the neutron capture cross section of ^{35}Cl, AMS measurements were performed on two samples, which were irradiated with neutrons of a Maxwell-Boltzmann energy distribution of 25 keV at the Forschungszentrum Karlsruhe. A preliminary mean value for the cross section is deduced by combining the AMS data and the neutron-fluence. The MACS will be calculated by weighting the mean value for the cross section with a Maxwell-Boltzmann energy distribution of 25 keV.

Theory

The cross section σ is defined as a measure for the probability that a specified reaction between a projectile and a target takes place. It is measured in units of area and it is the physical proportionality factor which connects the number of target nuclei and the fluence Φ with the number of produced nuclei (e.g. $N(^{36}Cl)=N(^{35}Cl)\Phi_n\sigma_{(n,\gamma)}$). In the case of neutron capture the cross section in nonresonant energy regions is proportional to the inverse velocity of the projectile. If the sum of the energies of the projectile and the target matches the energy of an excited state of the product nucleus resonances occur and the cross section can be orders of magnitude higher (see Fig.1).

In stellar environments neutrons thermalize very fast and their energies are Maxwell-Boltzmann distributed (see Fig.2) according to the temperature of the star. For the calculation of the reaction rates it is necessary to know the averaged cross section for these spectra. In the case of MB-distributions this is the MACS.

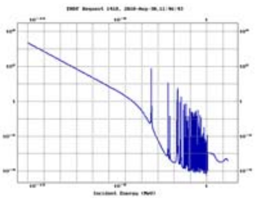

Fig.1: Energy dependent cross section of neutron capture on ^{35}Cl [1]

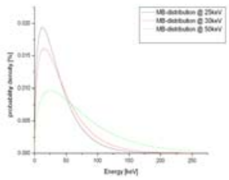

Fig.2.: Maxwell-Boltzmann disributions at different Energies

Proceedings

1.) Irradiations:

Neutron Irradiations of the NaCl samples were performed in 3 different facilities.

ATI, TRIGA II: Two samples (ATI2, ATI3) were irradiated for 30s with a thermal n-flux of ~$10^9 cm^{-2} s^{-1}$. The epithermic n-flux could not be neglected so these samples were used as test samples.

BRR, PGAA: Three more samples (BUD1-3) were irradiated with cold neutrons (~5meV) at the **P**rompt **G**amma-ray **A**ctivation **A**nalysis beamline of the **B**udapest **R**esearch **R**eactor. Here the neutrons of a certain energy are guided by total reflexion from the reactor to the target. So the neutron energies are very sharp but the flux is low ($10^7 cm^{-2} s^{-1}$).

KIT: The samples FZK 35Cla and FZK 35Clb were irradiated with a quasi Maxwellian neutron spectrum shown in Fig.3 at the **K**arlsruhe **I**nstitute of **Technology**. The neutrons are generated in the reaction $^7Li(p,n)^7Be$ by bombarding a Li or LiF target with 1912keV protons.

In all three cases Au-foils were used as fluence monitors.

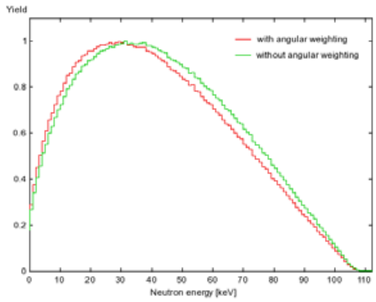

Fig.3: Neutron spectrum of the irradiation at FZK generated with the Monte Carlo simulation PINO [2] http://141.2.245.217/pino/

2.) Activity measurements:

The activity measurements on the Au-foils were carried out with a high purity Ge-diode. Gold was used as fluence monitor because the thermal cross section σ_{Au} of the reaction $^{197}Au(n,g)^{198}Au$ and the decay constant of ^{198}Au is very well known. The fluence was calculated by:

$$\Phi = K_\mu \frac{\lambda t_m}{(1-e^{-\lambda t_m})} \frac{T_B}{(1-e^{-\lambda T_B})} e^{\lambda T} \frac{A * mol_{Au}}{\sigma_{Au} N_L}$$

The first three factors are correction terms for the photon absorption, the measurement time t_m, the irradiation time T_B and the waiting time T. We calculated the $^{36}Cl/^{35}Cl$ ratio with the well known thermal n-capture cross section on ^{35}Cl.

$$\frac{^{36}Cl}{^{35}Cl} = \Phi \sigma_{Cl}$$

3.) AMS measurement:

Accelerator mass spectrometry is a sensitive technique to measure low isotopic ratios. The rare isotopes of interest are separated with different electromagnetic filters, an accelerator and different detectors.

For sulphur removal the NaCl samples were undergoing chemical pretreatment, where AgCl powder was produced. This was pressed in Cu-targetholders with an AgBr backing.

Our AMS measurements were performed at the 3MV tandem accelerator VERA (Fig.4). In order to get sufficient energy for the separation of 36Cl from its stable isobar ^{36}S, the measurements were performed with terminal voltages between +3.0 and +3.3MV and terminal foil stripping to the 7+ charge state. The identification of ^{36}Cl and ^{36}S is achieved with a split-anode ionization chamber and a silicon strip detector by the different energy loss of the two isotopes in a counting gas. The ^{35}Cl and ^{37}Cl currents were measured with Faraday cups.

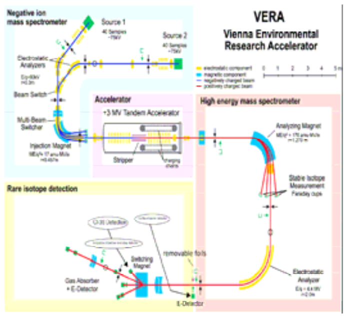

Fig.4: Schema of VERA based on a figure of L. Michlmayr

The evaluation of the data was done by setting gates for the two energy loss signals and the residual energy. Sample spectra recorded on a standard and a blank material are shown below.

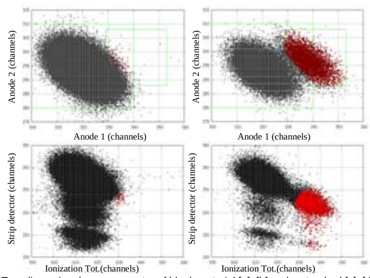

Fig.5: Two dimensional energy spectra of blank material [a], [b] and a standard [c], [d]. The red dots are counted as ^{36}Cl, all other registered events are marked by black dots.

Results

The raw $^{36}Cl/^{35}Cl$ values were corrected for the sulphur induced background and the blank value and then normalized to a standard.

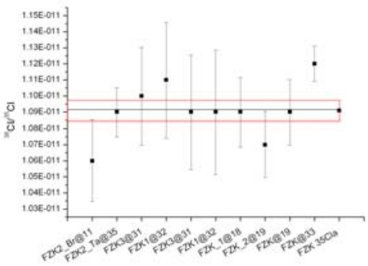

Fig.6: Comparison of different measurements of the $^{36}Cl/^{35}Cl$ ratio on the same sample. It is clearly shown that nearly all values are within the uncertainty (red line) of the averaged value (black lines).

	36Cl/35Cl activity	err [%]	36Cl/35Cl AMS	err [%]	AMS/activity
BUD1	7.82E-12	1.22	7.84E-12	0.84	1.003E+00
BUD2	1.05E-11	1.22	1.07E-11	0.68	1.016E+00
BUD3	9.57E-12	1.21	9.88E-12	1.07	1.033E+00
ATI2	5.25E-12	1.50	4.77E-12	0.50	9.092E-01
ATI3	5.47E-12	1.49	4.99E-12	0.57	9.118E-01

Tab.1: Results for the 36Cl/35Cl ratios from the activity and the AMS measurements. The vlaues for the BUD samples are in good agreement while the values for the ATI show discrepancies of 9%. This discrepancies are mainly caused by epithermic neutrons during the irradiation at the reactor.

First, the experimental cross section for the FZK samples was calculated. To get the MACS for 35Cl we normalized the experimental cross section and weighted it with a factor which considers the difference between a true Maxwell-Boltzmann-spectrum and the FZK neutron spectrum.

$$\sigma_{MACS} = \frac{2}{\sqrt{\pi}} \frac{\langle \sigma_{MB} \rangle}{\langle \sigma_{SACS} \rangle} \sigma_{exp}$$

	36Cl/35Cl	err [%]	Φ [cm^{-2}]	err [%]	σ [cm^2] exp	err [%]
FZK 35Cla	1.09E-11	0.58	1.22E+15	5.50	8.94E-27	5.53
FZK 35Clb	8.90E-12	0.62	1.27E+15	5.50	7.01E-27	5.53

Tab.2: Results of the AMS measurements on the samples irradiated at the KIT.

	σ [cm^2] MACS	err	err [%]
FZK 35Cla	1.06E-26	5.86E-28	5.53
FZK 35Clb	8.84E-27	4.89E-28	5.53
Sayer [3]	1.10E-26	3.30E-28	3.00
Bao [4]	1.17E-26	-	-

Tab.3: Values for the MACS of 35Cl measured in this work and values from Sayer et all[3] and Bao et all[4]. The two values from this work show a discrepancy of 20% the reason is not clear yet.

Conclusions:

- The absulute 36Cl/35Cl value for the BUD and ATI samples from the activity measurements allows us to use these samples as independent standards for AMS mesurements.
- The MACS calculated from the measurement on FZK 35Cla is within the uncertainty of the value from Macklin. The MACS measured on FZK 35 Clb is 20-32% smaller than the other values. This would decrease the estimated stellar production of ^{36}Cl dramatically. To clarify this discrepancy between the two measured values, more AMS measurements on samples irradiated at KIT should be performed.

References:

[1] http://www-nds.iaea.org/exfor/servlet/E4sMakeE4; ENDF/B-VII database

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[3] K. H. Guber, R. O. Sayer, T. E. Valentine, L. C. Leal, R. R. Spencer, J. A. Harvey, P. E. Koehler and T. Rauscher; New Maxwellian averaged neutron capture cross section for ^{35,37}Cl Phys. Rev. C **65**, 058801, 2002

[4] Z. Y. Bao, H. Beer, F. Käppeler, F. Voss, K. Wisshak and T. Rauscher; Neutron cross sections for nucleosynthesis studies; At. Nucl. Data Tables 75, 1 (2000)