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Study of nuclear reactions producing ^{36}Cl by micro-AMS

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Abstract. ^{36}Cl is one of several short to medium lived isotopes (as compared to the earth age) whose abundances at the earlier solar system may help to clarify its formation process. There are two generally accepted possible models for the production of this radionuclide: it originated from the ejecta of a nearby supernova (where ^{36}Cl was most probably produced in the s-process by neutron irradiation of ^{35}Cl) and/or it was produced by in-situ irradiation of nebular dust by energetic particles (mostly, p, α , ^3He -X-wind irradiation model).

The objective of the present work is to measure the cross section of the $^{37}\text{Cl}(p,d)^{36}\text{Cl}$ and $^{35}\text{Cl}(d,p)^{36}\text{Cl}$ nuclear reactions, by measuring the ^{36}Cl content of AgCl samples (previously bombarded with high energy protons and deuterons) with AMS, taking advantage of the very low detection limits of this technique for chlorine measurements.

For that, the micro-AMS system of the LFI/ITN laboratory had to be optimized for chlorine measurements, as to our knowledge this type of measurements had never been performed in such a system (AMS with micro-beam).

Here are presented the first results of these developments, namely the tests in terms of precision and reproducibility that were done by comparing AgCl blanks irradiated at the Portuguese National Reactor with standards produced by the dilution of the NIST SRM 4943 standard material.

1. Introduction

The motivation for measuring cross sections of nuclear reactions producing ^{36}Cl lies in the field of astrophysics that studies the origin and formation of the solar system. ^{36}Cl is one of the short-lived radionuclides (called SLR's for abbreviation), thus named because of their short half-lives (~ 1 Myrs), whose abundances in the early stages of the formation of the solar system is much higher than expected from the galactic background. These SLR's are ^7Be ($t_{1/2} = 53$ days), ^{10}Be ($t_{1/2} = 1.4$ Myrs), ^{26}Al ($t_{1/2} = 0.74$ Myrs), ^{36}Cl ($t_{1/2} = 0.3$ Myrs), ^{41}Ca ($t_{1/2} = 0.1$ Myrs), ^{53}Mn ($t_{1/2} = 3.5$ Myrs) and ^{60}Fe ($t_{1/2} = 1.5$ Myrs), and are known to have been present in the early stages of the solar system through the presence of their daughter isotopes in CAIS (calcium and aluminium rich inclusions) and chondrule present in chondrites, which are the most common form of meteorites found on earth [1]. These structures are thought to be amongst the oldest solid material present in the solar system, and they can provide important information about its formation.

The analysis of Cl-S systematics of sodalite in carbonaceous chondrites yielded a ratio of $^{36}\text{Cl}/^{35}\text{Cl}$ around 10^{-6} . Since sodalite is in principle a late-stage product of aqueous alteration, this ratio's initial value should have been higher, assuming that the ^{36}Cl supernova injection happened in the prestellar core stage, as it is supposed to have happened with the other SLRs. Following this principle, the expected $^{36}\text{Cl}/^{35}\text{Cl}$ ratio at the time of the SLRs injection has been estimated to be around 10^{-4} . This value is already too high for the estimated supernova injection of $^{36}\text{Cl}/^{35}\text{Cl} \sim 10^{-6}$. However, recent analysis of wadalite in the allende carbonaceous chondrite seems to make the SN injection of ^{36}Cl scenario even more implausible, yielding a ratio of $^{36}\text{Cl}/^{35}\text{Cl} \sim (1.72 \pm 0.25) \times 10^{-5}$, making the initial ratio even higher than 10^{-4} [2]. This seems to definitely invalidate the possibility of ^{36}Cl injection by a



supernova. Therefore the irradiation of nebular dust by energetic particles (mostly, p, α , ^3He -X-wind irradiation model) becomes a strong candidate to explain the abundance of the ^{36}Cl isotope.

It is in this context that the research group is currently working on measuring the $^{37}\text{Cl}(p,d)^{36}\text{Cl}$ and $^{35}\text{Cl}(d,p)^{36}\text{Cl}$ nuclear reactions, using the Micro-AMS system at LFI/ITN to measure the amount of ^{36}Cl produced in the AgCl irradiated samples, starting by the optimization of this system for ^{36}Cl measurements.

2. The Micro-AMS system at LFI/ITN

The Micro-AMS system present at LFI/ITN is the former AUSTRALIS system developed by Sie et al. [3] at CSIRO. It is based on a 3 MV tandem accelerator. Its main feature is the ion source, a HICONEX cesium sputter source that was adapted in order to produce a Cs^+ micro-beam that can be focused to very small diameter (30 μm), and a low-energy and high-energy bouncing system that allows quasi-simultaneous measurement of different isotopes.

It was developed with the goal of using the capabilities of a SIMS system (spatial and in-depth analysis of bulk samples) connected to an AMS system that provides the possibility of resolving isobaric and molecular interferences, that in many cases spoil the detection limits of SIMS analysis.

3. Adaptation of the system to the measurement of ^{36}Cl

Measurement of AgCl samples is very common in conventional AMS but has never been tried with Micro-AMS, since its lower intensity primary beam implies much lower detection limits for ^{36}Cl , while in principle not bringing any important advantage. However, since it is possible to produce ^{36}Cl (through the astrophysically relevant $^{37}\text{Cl}(p,d)^{36}\text{Cl}$ and $^{35}\text{Cl}(d,p)^{36}\text{Cl}$ nuclear reactions) in enough quantity to bring the isotopic ratios present in the sample to the $^{36}\text{Cl}/^{35}\text{Cl} > 10^{-10}$ range, the micro-beam AMS technique becomes a valid technique to measure cross-sections while eventually providing some advantages as, for instance, direct measurement of the irradiated samples without further chemical manipulation, since with the micro-beam it is possible to measure on the exact beam spot of the irradiation in the sample.

The original AUSTRALIS system was not prepared for very low isotopic ratio measurements (below 10^{-5}), and so adaptations had to be made in order for it to be possible to measure the high intensity beams (^{35}Cl and ^{37}Cl) and very low intensity beams (^{36}Cl) quasi-simultaneously, which was done by installing off-axis faraday cups after the Low-energy and high-energy magnets. Another important step was to minimize sulfur contamination in the target chamber.

Sulfur contamination is a problem in conventional Chlorine AMS measurements, since the ^{36}S isotope is an isobaric interference of ^{36}Cl . Up until recently an effective isobar separation of ^{36}S and ^{36}Cl was impossible using 3 MV tandems. However, work developed in ETHZ Zurich and VERA in Vienna [4,5] in the last years has successfully achieved separation for ^{36}Cl and ^{36}S using a 3 MV tandem.

However, these separation procedures for 3 MV tandems were not tried in the case of this work because the current state of the ITN tandem does not permit it to operate much above 2,5 MV. But since the Micro-AMS uses a primary cesium micro-beam, which is on average three orders of magnitude less intense than the conventional AMS primary beam, this means that its detection limits fall by that amount or more when compared to AMS. Our current detection limit, measured by analyzing a blank AgCl sample (produced with sulfur removal chemical treatment [6]) and monitoring the detector background, yielded a background level below $^{36}\text{S}/^{35}\text{Cl} = 10^{-10}$.

It is in principle possible to keep the ^{36}S reaching the detector on the level of $^{36}\text{S}/^{35}\text{Cl} = 10^{-12}$, by keeping the sample chamber very clean and by taking special steps in the design of the sample holder, besides using the well-known chemical sample preparation methods for the standards and for the blank AgCl material to be used as blank material and irradiation material.

The contamination of ^{36}S present in the sample holder is a well-known problem in conventional AMS. The micro-beam used in Micro-AMS is an advantage in this case, because it is easier to assure

that the whole bombarded area is located strictly within the target limits, as opposed to conventional AMS where the large diameter beam, maximized for intensity, will easily impact the sample holder as well during operation. However, since there is no possibility to separate ^{36}S from ^{36}Cl in our system, it is important to assure that no sulfur contamination may be present anywhere close to the sample. This means that the chamber has to be as clean as possible, and the sample holder must be designed so as to minimize any contamination as well.

The original sample holder was covered with a sheet of tantalum. In each of the positions where the samples were to be mounted, a round disk of two centimeters in diameter of compressed AgBr was installed. The AgCl samples (also a compressed powder) were then mounted at the center of these disks. Using AgBr to cover the sample holder in the sample area was shown in the literature [7] to be the most efficient material in minimizing sulfur contamination in the chlorine beam.

Since cross-contamination of samples can happen easily, the maximum number of samples per run was set at 4. This way it was possible to keep the samples at a reasonable distance (> 3 cm) from each other, and still be able to have a blank sample, a standard, and two unknown samples in each run.

4. Measurements

The AgCl samples were irradiated in the RPI nuclear reactor at ITN using the neutron capture nuclear reaction $^{35}\text{Cl}(n,\gamma)^{36}\text{Cl}$. Neutron flux and irradiation times were set to produce $^{36}\text{Cl}/^{35}\text{Cl}$ isotopic ratios in the 10^{-6} to 10^{-9} range. The thermal neutron fluxes were controlled with gold monitor samples, by measuring the 411,8 keV gamma emission, emitted by the decay of the ^{198}Au ($t_{1/2}=2,695$ d) produced by the neutron capture reaction $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ with a HPGe detector, after each neutron irradiation.

Most of the measurements were performed on irradiated samples with isotopic ratios in the 10^{-6} and 10^{-7} range. The high content of ^{36}Cl in this samples made it easier to tune the system. The standards were prepared to have an isotopic ratio of $^{36}\text{Cl}/^{35}\text{Cl}= 1 \times 10^{-6}$, by diluting the NIST SRM 4943 standard. Due to the lower intensity of cesium primary beam, the memory effect was not important and it was monitored with a blank sample after each measurement of sample or standard.

5. Results

Figure 1 shows a measurement performed on an irradiated sample. Figure 2 shows three different normalized measurements on the same sample (each of the measurements was succeeded by a standard measurement which was used for normalization); the first two in the same day but with somewhat different beam tunings and the last, two days after the first two. The highest error bar is in the 4% precision range.

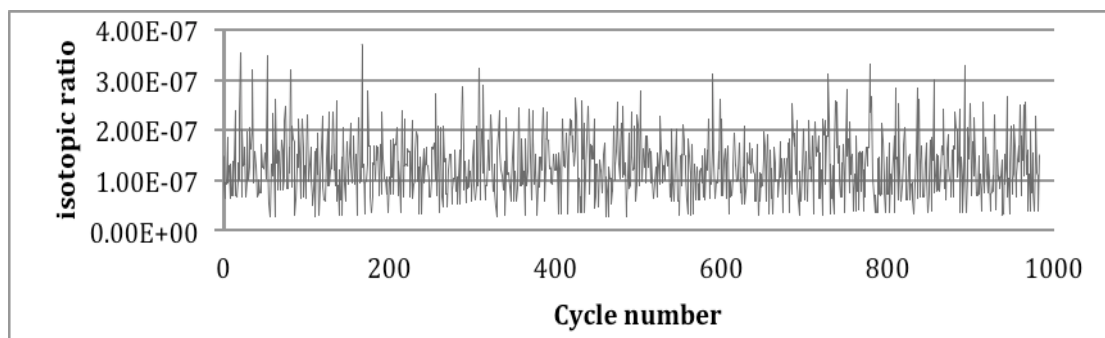


Figure 1: $^{36}\text{Cl}/^{35}\text{Cl}$ isotopic ratio measured in an irradiated sample vs. cycle number. Each of the cycles represents a 100 ms measurement time for each of the three chlorine isotopes.

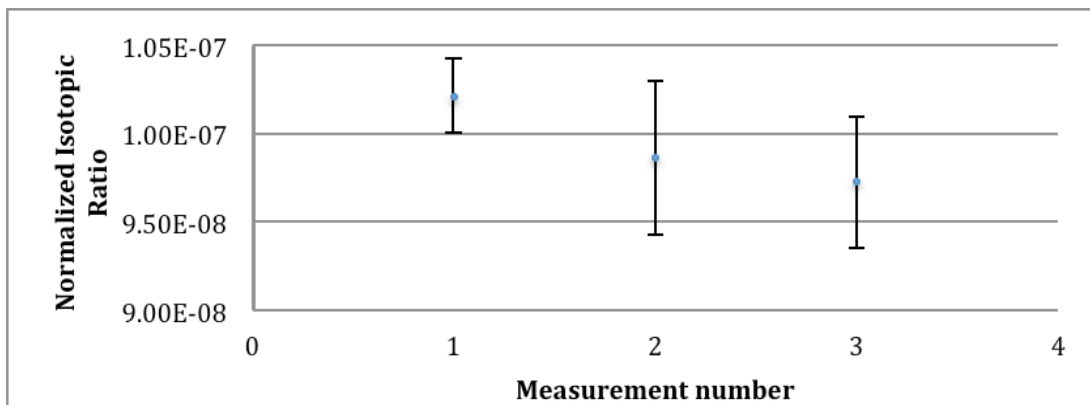


Figure 2: Isotopic ratios of an irradiated sample normalized for the diluted standard. Measurement number 1 and 2 were performed on the same day. Measurement number 3 was performed two days later.

6. Conclusions

The results so far show on the one hand a good agreement in terms of the ^{36}Cl content of the samples between the irradiated samples and the diluted standards and a good reproducibility of the measurements. This validates both the neutron irradiations as a valuable process for the production of standards and the chemical procedures used for the production of standards and blank material. However, the precision of the measurements is still not as good as expected. This is a problem as the objective of the work is to measure samples with less ^{36}Cl than the ones measured so far, which means an even lower precision, as less ^{36}Cl means less counts per second in the particle detector.

In order to achieve a better precision, steps are being taken to increase the primary beam intensity as well as decreasing the measurement cycle time, especially for the ^{35}Cl and ^{37}Cl beams, by connecting the Faraday cups to a faster current integrator.

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