Alpha-Spectroscopy to Determine $^{144}\text{Sm}(\alpha,\gamma)$ Activation Product

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October 2019

Abstract.

The purpose of this article is to describe the method used to determine the amount of $^{148}\text{Gd}$ produced in the $(\alpha,\gamma)$ activation of enriched $^{144}\text{Sm}$ samples. Advanced simulations are fitted to data to help constrain systematic uncertainties. The simulations are complimented with RBS measurements of the samples.

1. Introduction: $^{144}\text{Sm}(\alpha,\gamma)^{148}\text{Gd}$ Reaction

The study of isotopic abundance link two scientific fields together. These are the fields of nucleosynthesis and radiogenic isotope geology. The former’s goal is to explain the chemical evolution of the universe. Where as the latter uses isotopic abundances as a dating tool. The change in isotopic abundance as a function of time due to unstable isotopes define the age of an object.

The radiogenic dating technique relies on isotopes with half-lives in the range of 0.5 mega years to $10^{12}$ years. One of the weaknesses of this technique is the reliability of measured the half-lives. The half-lives have to be measured in the laboratory, which require samples to be made. The samples should have enough material to give a measurable signal rate‡. When an isotope has a half-life much shorter than the age of the universe (13.8 giga years), the material has to be made as the isotope will have no natural abundance.

Another weakness of the dating system is the reliance on accurate cross sections data to predict the isotopic ratios at formation. All of the alpha decaying isotopes used for dating are $p$-process isotopes. Most of these isotopes are created when material from the $s$ and $r$-process are broken down into lighter elements through $(\gamma,n)$ reactions, and subsequent $(\gamma,p)$ or $(\gamma,\alpha)$ processes (see [1] for more details). This topic is closely

‡ In this context a measurable signal rate should be higher than the background, and high enough to give a reasonable statistical uncertainty.
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related to $p$-process nucleosynthesis as the $p$-process calculation determine the material abundance at formation.

One example of a promising isotope that is plagued by these issues is $^{146}$Sm [2]. It is unknown if the issue with this isotope is due to the incorrectly measured half-life [3], or the initial abundance prediction from the $p$-process [4]. As $^{144}$Sm and $^{146}$Sm are pure $p$-process isotopes, their abundance at formation is sensitive to the $^{148}$Gd($\gamma,\alpha$)$^{144}$Sm process. For clarity a cut-out of the relevant region of the chart of nuclei has been added, see the left side of Figure 1. There is also a table denoting the energy and half-lives of the relevant alpha decaying isotopes given in Table 1.

To help to determine the source of the inconsistency a remeasurement of the inverse reaction $^{144}$Sm($\alpha,\gamma$)$^{148}$Gd cross-section was carried out. Six samples were made and five were activated. The samples were then measured with an ultra-low background ionisation chamber. The chamber was specially designed and built to measure low signal rates and has a background in the region of interest ($E_\alpha\approx4\text{MeV})$ of around 0.8 counts per day (c.p.d.) [5]. The design and operation of this alpha detector will be discussed, as well as some preliminary result on the determination of the geometric efficiency of the samples. For more information on the sample production and a more thorough discussion on the nuclear astrophysics implications see [6].

| Isotope | Alpha Energy [MeV] | Half-life [yr] |
|---------|--------------------|---------------|
| $^{148}$Gd | 3.182690(24) | 71.1(12) |
| $^{147}$Sm | 2.248(1) | 1.060(11) $\times 10^{11}$ |
| $^{150}$Gd | 2.726(9) | 1.79(8) $\times 10^{6}$ |
| $^{146}$Sm | 2.460(3) | 6.8(7) $\times 10^{7}$ |
| $^{232}$Th | 4.0123(14) | 1.40(1) $\times 10^{10}$ |
| $^{238}$U | 4.198(3) | 4.468(6) $\times 10^{9}$ |

2. Detector Design

To measure alpha decay rates in the order of 10 c.p.d. the design of the twin Frisch grid ionisation chamber (TF-GIC) was utilised. The traditional design for the device was used to measure fission fragments using the so-called double kinetic energy technique (see [9], [10]). The sample would be on the centre of the cylindrical chamber and two identical anodes would be placed on the top and bottom of the TF-GIC. This would allow detection of angular correlations if the sample was thin enough.

This design was adapted by placing the anode in the center of the TF-GIC and the cathodes on the top and bottom, effectively splitting the chamber into two sub chambers.
In this design the upper chamber acts as a veto for the lower chamber. This technique drastically reduces noise from micro-phonics as well as any cosmogenic backgrounds. Though it must be stated that the contribution from cosmogenic background is very little as cosmic rays rarely deposit enough energy to overcome the lower energy threshold. If they do however deposit larger amounts of energy, it is simple to remove this background using cuts.

The Frisch grid technique was developed in 1944 [11]. The energy of each event is determined by detecting the number of ionised electrons produced inside of the TF-GIC. This detection uses the induced current on the anode to do so. This current is induced when the ionised electrons move between the cathode and the anode. The amount of induced current is sensitive to the path that the ionised electrons take. If two events occur with the same energy from the same starting position but one has a different angle, the induced current will be different for both of them. This smears out the energy resolution of the chamber. The Frisch grid design inserts an intermediary grid between the anode and the cathode. This divides the chamber into an interaction region and a detection region. This effectively means that the anode is only sensitive to the electrons traveling between the grid and the anode, which reduces this smearing affect and improves the energy resolution. A schematic of the TF-GIC can be seen in the right side of Figure 1.

Ultra pure materials were used to further reduce the background expected in the


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The main body of the chamber is electro polished stainless steel, which helps in the removal of surface contaminations. The copper used to form the anode and grids were electroformed to remove contamination. The sample holder is a single 30 cm silicon wafer. This type of wafer was chosen as silicon has to be very pure to form crystals with such a large diameter. The silicon was shown not to act as an insulator in the TF-GIC, keeping the gas from becoming quenched [12].

The counting gas used in the detector is P10. This is a mixture of 10% methane and 90% argon. The temperature and pressure is measured throughout the run. The chamber is flushed with P10 before the start of each run. During this process the chamber is evacuated to approximately 0.1 mbar, and then refilled to 1 bar or P10 several times. Each time the chamber is filled and evacuated more contaminants such as oxygen and $^{222}$Rn are removed. An effective run can last as long as 40 days. For further description of the chamber design and calibration see [5].

Every event pulse shape can be recorded due to the low rate achieved in the TF-GIC. This means that a rigorous off line analysis can be performed. The chamber uses three channels of a pre-amp and ADC to record the signal. The low noise pre-amp amplifies the signal to a detectible voltage while the ADC digitises the signal which is then recorded onto a 7.3 TB hard drive. To optimise the storage each event is recorded in ROOT files. This is helpful as the analysis is also based in ROOT.

3. Data Taking

Six sample were made via the evaporation of enriched samarium. The contribution of $^{144}$Sm was enriched to 88%. These were then activated using an alpha beam with an energy range below 13 MeV. The total charge collected was less than 3.2 mC per sample. After activation the six samples were brought to the TU-Dresden for measurement. The samples were stored in a special evacuated container to reduce the rate of oxidation. They were only exposed to air during the loading and unloading of the TF-GIC.

The spectra of all of the runs can be seen on the left side of Figure 2. As can be seen the $^{147}$Sm line is roughly the same for all of the samples. The combined measured counting time of the samples was 111 d. The detector is able to measure continuously for long periods of time without interruption. The right side of Figure 2 shows the relative statistical uncertainty as a function of time. The cutoff was decided to be at 4% which is roughly 400 counts in the region from 2.5 MeV to 3.5 MeV.

At the start of each measurement a signal generator was used to determine the offset of the ADC as well as the contributions of the electronic components to the energy smearing. The pulse shape generator is set to mimic a signal pulse. The pulses are then injected into the ADC during the run. The frequency is set to 100 Hz for approximately 10 min, to get good statistics while causing little dead time. The most active sample has a rate of around 20 mBq. As explained in section 2, the detector only uses three channels of the pre-amp and ADC. The signal from the pulse shape generator is recorded on all four channels simultaneously. This makes it possible to retrieve all of
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Figure 2. left: A histogram showing the rate of each sample, the events in the range 3.5 MeV to 5 MeV comes from natural thorium and uranium contamination. The peaks above this are from 222Rn contamination. right: A graph showing the relative statistical uncertainty as a function of time. The red line showing the 4% cutoff (400 counts). Sample 7 is the exception to this, as the rate was very low.

the pulses from the signal generator without compromising the data.

Sample 4 was measured as the first sample and again as the last (referred to as “sample 4 test”). This was to test if the adherence of the material to the substrate was stable over a long period of time after being handled multiple times. There was 55 d between the measurements. The comparison plots can be seen in the left side of Figure 3. The spectra are mostly the same apart from a small change in the shape of the 147Sm peak. This could be due to a change in the size of the oxide layer. The test did however show that non of the material was lost during the measurements, and that there was no contamination between the samples.

Figure 3. left: The comparison of the two runs of sample 4. The subfigure shows the significance of the deviation of each bin, where $s = (N_1 - N_2)/\sqrt{N_1^2 + N_2^2}$. right: Comparison of the signal from the samples. Sample 5 is shown in red as it was not activated and gives an idea of the contamination in the signal region.

Sample 5 was never activated, but was kept in the same conditions as the other samples. This means that if sample 5 shows a $^{148}$Gd peak, there was definitely cross
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contamination. The sample also shows the possible limit of this activation technique. Sample 5 shows a count rate of around 21 counts during a measuring time of 36 d within the $^{148}$Gd signal region. This rate is more than an order of magnitude lower than the rate of the sample with the lowest $^{148}$Gd activity. The source of this contamination is mainly from the uranium and thorium chain contamination at higher energies. As the uranium and thorium contamination is homogeneously spread through the aluminium backing, a flat spectrum is expected from 5 MeV to zero. This can be suppressed by using cleaner sample backing materials. The level of contamination can be seen if the right side of Figure 3.

4. Analysis

The aim of this analysis is to determine the number of $^{148}$Gd atoms present in the samples. This value is determined using the half-life and the true rate. The true rate is determined by measuring the rate in the TF-GIC and accounting for the efficiencies. There are three efficiencies that have to be taken into account. The first is the detector efficiency. This is derived by comparing the measured rate to that of a calibration source. The next efficiency is the data cut efficiency. This is determined by applying the data quality cuts to signal and noise pulses.

The last efficiency is the geometric efficiency. Here the term geometric efficiency denotes the ratio of the total events that occur in a sample to the number of events that deposit enough energy to be detected. This factor is heavily influenced by the thickness of the sample; as self absorption dominates the alpha spectrum. As the chamber only measures the upper surface of a sample, the maximum geometric efficiency is 50% for an extremely thin sample. The geometric efficiency is determined using the Monte Carlo method.

As mentioned in [6], the RBS showed a layer of oxide on the samples. This does have some impact on the determination of the geometric efficiency and number of counts. For this reason an analysis method was developed to effectively determine the size of the oxide layer during the measurement. This could only be done due to the contamination of $^{147}$Sm in the sample. Even though the samples were made using enriched $^{144}$Sm (88% abundance), a small contamination of $^{147}$Sm remained (3.8% abundance). The $^{144}$Sm should follow the same distribution as the $^{147}$Sm in the sample. This contamination of $^{147}$Sm caused the largest part of the signal coming from the samples. A technique was developed to model the geometry and then fit the data with the Monte Carlo approach. This was done using the GEANT4 [13] simulation package.

The samples were modelled using a symmetrical gaussian shape, this can be seen in the left side of Figure 4. The model was also simplified in the following way. The ionisation is more sensitive to the density than the composition of the material, this is due to the ratio of ($Z/A$) being mostly constant. Instead of simulating the oxide layer in the Monte Carlo, pure samarium can be simulated and the density can be changed. Changing the density is also equivalent to changing the thickness of the layer. This
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Figure 4. left: A graph showing the position of each simulated event for a 100 μm geometry, the colour corresponds to the energy deposited in the detector. The simulation is consistent with TRIM calculations [14]. right: Histogram showing the simulated spectra broken into top and bottom layers.

greatly simplifies the simulation, as only one sample geometry has to be simulated and the oxide layer can be mimicked later in the analysis by applying a cut. Histograms of the Monte Carlo simulations can be seen in the right side of Figure 4.

The data was fit with Monte Carlo simulations in two ways. The first was to transform a high statistics spectrum into a spline function using the TSpline class from ROOT. This was then given to the fitting algorithm. The fitter was able to smear the spline function and adjust the energy calibration simultaneously. This essentially turned the Monte Carlo template into a continuous function. This can be seen in the left side of Figure 5. To determine the thickness of the oxide layer the simulated thickness was varied over a wide range. The $\chi^2$ was determined for each simulated thickness. The minimum and one $\sigma$ uncertainty bands can then be calculated. The Spline fitting method is helpful here as it gives a much smoother $\chi^2$ distribution, shown in the right side of Figure 5. This method is also able to determine the energy resolution and calibration in a more consistent manner.

Figure 5. left: Spectrum of sample 2 fitted with the spline method. The reduced $\chi^2$ is 1.65 which is acceptable at this level. right: A graph showing the distribution of $\chi^2$ as a function of thicknesses.
To determine the number of counts, the TFractionFitter [15] package is used. This fitting method takes into account that the Monte Carlo sample is finite. The weighting in the Monte Carlo templates were kept at unity. This is important for this fitting method as it can give false results otherwise. The optimum Monte Carlo templates were defined using the spline fitting method, and were then fed into the fraction fitting method to determined the number of counts of $^{148}\text{Gd}$, $^{147}\text{Sm}$ and background. A flat distribution from is used here to simulate the background. This is consistent with thorium or uranium that is homogeneously embedded into the aluminium backing. The left side of Figure 6 shows the results of the TFractionFitter algorithm.

**Figure 6.** *left* A histogram showing the results of the fraction fitting. A flat background is added, and the fitting range is increased to constrain this contribution. *right* A histogram showing the simulated $^{148}\text{Gd}$ efficiencies. The variation here comes from the variation of the RBS parameters shown in [6], as well as the statistical fluctuation of the Monte Carlo model.

Due to the simplification made in the simulation, the geometric efficiency is easy to extract. The geometric efficiency of the oxide layer does not have to be calculated separately, as the whole sample is simulated using the same material. The simulation is projected into an energy spectrum, which is then smeared with the energy resolution found from the spline fitting. The events above the lower energy threshold are then counted. This number is then divided by the number of simulated events. A trick is used to determine the systematic uncertainty on this value. Firstly, all of the parameters in the simulation are fixed. The simulation is run numerous times. This gives a gaussian distribution which is due to the statistical fluctuation of the Monte Carlo. This process is then repeated but the parameters are varied. The increase in the width of the geometric efficiency distributions is now due to the variation of the parameters. This distribution can be seen in the right side of Figure 6. The preliminary geometric efficiency was found to be $46.77(20)\%$. 
5. Conclusion & Outlook

Thus far it has been shown that the Monte Carlo simulation can be simplified to mimic the oxide layer on the sample. These results are only preliminary, and a detailed uncertainty analysis is being performed on the chosen method. This method will then be applied to all of the samples. Preliminary results and discussion can be found in [6]. A long term goal of remeasuring the half-life of $^{146}$Sm is also underway.

Acknowledgments

We would like to thank K.O. Zell and A. Blazhev for the target production and V. Foteinou and H.W. Becker for the assistance during the RBS measurements. This project was supported by the Deutsche Forschungsgemeinschaft under the contract ZI 510/8-1 and the ULDETIS project within the UoC Excellence Initiative institutional strategy.

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