

# Quantifying Oxygen Content in Tungsten by Ion Beam Analysis

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**Abstract:** The quantitative analysis of  $^{16}\text{O}$  in near-surface regions of samples can be performed with  $^{16}\text{O}({}^3\text{He},\text{p})^{18}\text{F}$  reaction. The cross section for this nuclear reaction in  $\text{SiO}_2$  samples has been determined at a reaction angle  $135^\circ$  for  ${}^3\text{He}$  energies in the laboratory frame between 1800 keV and 6100 keV. A relation between the number of protons collected as a result of the nuclear reaction and the amount of oxygen in tungsten samples has been determined.

## I. INTRODUCTION

Tokamaks are one of the main devices to allow controlled fusion reactions with the aim of providing energy on a large scale. The reaction will be produced in a deuterium-tritium plasma and confined by means of strong magnetic fields, as no material is capable of resisting such high temperatures [1]. Oxygen plays a major role in the retention of hydrogen isotopes in the walls of the Tokamaks, mainly made of tungsten; the more fuel retention, the less probability of efficient fusion. The presence of oxygen is technically unavoidable, as the surfaces will be no matter what in contact with air, either during the manufacturing process, during maintenance of the machine or during exposure with the plasma itself. In this thesis, a method to quantify the oxygen in tungsten via nuclear reaction analysis (NRA) is presented [2].

The typical method to quantify elements in a sample is by means of Rutherford Backscattering (RBS). However, this procedure is not so favourable when it comes to measure light elements in heavy materials, just like tungsten, as the Rutherford cross section for heavy elements is so big that would impede the signal of the light elements from being noticeable. Therefore, an indirect method is needed as well as another target, more suitable for a proper oxygen quantification through RBS. The material selected is  $\text{SiO}_2$ , as it is a good candidate for both RBS and NRA: the goal is to get a precise quantity of oxygen out of the RBS profile in the case of the  $\text{SiO}_2$  and establish a relation between the number of protons collected -result of the nuclear reaction- and the quantity of oxygen. As an example, this relation is used to determine with precision the abundance of oxygen in tungsten samples.

The reaction selected for the NRA analysis is  $^{16}\text{O}({}^3\text{He},\text{p})^{18}\text{F}$ , being  $^{16}\text{O}$  the target, and  ${}^3\text{He}$  the projectile. The helium isotopes are accelerated at the Tandem Accelerator facility in Max Planck Institut für Plasmaphysik in Garching. For Nuclear Reaction Analysis, the differential cross section of the reaction at a precise angle must be known. However, very few studies have

previously measured the cross section for this particular reaction, and those who had, did in very specific conditions, unhelpful for our purposes. W. N. Lennard [3] measured the cross section for all reactions detected  $^{16}\text{O}({}^3\text{He},\text{p}_{0,1,2,3,4,5,6})^{18}\text{F}$  for a unique energy of 2390 keV and an angle of  $153.7^\circ$ . In his study the Q-values of every reaction were also determined. Heggie, Switkowski and Clark [4] determined the cross section by means of measuring the deexciting  $\gamma$ -rays for the first three levels in  $^{18}\text{F}$  (937, 1040 and 1080 keV). The bombarding energies of  ${}^3\text{He}$  lied in a range between 2600 keV and 4000 keV.

## II. EXPERIMENT

The cross section of the reaction  $^{16}\text{O}({}^3\text{He},\text{p})^{18}\text{F}$  was determined by the energy spectra of the protons coming out of the reaction at the certain angle of  $135^\circ$ .

The  $\text{SiO}_2$  targets consist of thermally grown  $\text{SiO}_2$  on Si 001 single crystal mirror polished wafers. Different thickness  $\text{SiO}_2$  samples were studied.

The proton detector placed at  $135^\circ$  has a parabolic slit with a width of 3 mm and a height of 17 mm. Since it is located about 37 mm away from the sample, this results in a solid angle of 30.26 msr. It has a depletion depth of 2000  $\mu\text{m}$  and nickel and Mylar foils are installed in front of it to stop energetic backscattered  ${}^3\text{He}$ . These foils consist of 5- $\mu\text{m}$ -thick Ni and 12- $\mu\text{m}$ -thick Mylar layers, both coated with 10 nm Au. The RBS detector is positioned at  $165^\circ$ : the aperture slit is 5 x 1  $\text{mm}^2$  with rounded corners, resulting in a slit area of 4.8  $\text{mm}^2$ . The aperture - sample distance is 64.1 mm, resulting in a solid angle of 1.11 msr. The depletion depth for this detector is measured to be about 500  $\mu\text{m}$ .

Apart from the protons resulting from the several above-mentioned nuclear reactions related to oxygen, some others can reach the detectors and give additional signal. Those are mainly the protons of the reactions with silicon  $^{28}\text{Si}({}^3\text{He},\text{p}_{0,1,2,3,4})^{30}\text{P}$  [5], together with, to a lesser extent, protons coming from reactions with impurities in the sample, basically carbon:  $^{12}\text{C}({}^3\text{He},\text{p})^{14}\text{N}$  and  $^{13}\text{C}({}^3\text{He},\text{p}_{0,1,2})^{15}\text{N}$ .

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### III. RESULTS

#### A. Accurate Charge Measurement

The accuracy of the current measurement is known to be within 3% for the given setup. In order to reduce this even further, a 100-Å-thick Au layer was deposited onto the SiO<sub>2</sub>, by means of vacuum evaporation method. Plotting the integral under the Au peak for the RBS experiment and comparing it with the theoretical RBS cross section allows to derive the actual accumulated charge and do the subsequent corrections for the future cross section plots. The deviation has been observed to grow with the energy.

#### B. 680-nm-thick SiO<sub>2</sub> sample

The first sample studied is a 680-nm-thick layer of SiO<sub>2</sub> over bulk Si, and on top of it, the above mentioned Au layer. As for the measurement protocol, the energy step is 50 keV and a total amount of 5 μC of charge is collected for every data point. Before the first measurement on the studied sample, a test on millimetric paper is performed in order to check the position and dimensions of the beam spot. A visible 1x1 mm<sup>2</sup> squared signal was captured for a 3 μC accumulated charge.

In Fig. 1, the proton spectrum for an incident <sup>3</sup>He energy of 2400 keV is plotted, showing clearly four peaks corresponding to the different sources of protons. Except for the p<sub>0</sub> peak, all of them more or less superimpose, so extracting the cross section turns to be impossible unless only the p<sub>0</sub> peak is chosen. Therefore, the cross section will be calculated for the <sup>16</sup>O(<sup>3</sup>He,p<sub>0</sub>)<sup>18</sup>F reaction, which is, in this energy, practically noise free. At low energies, however, the background noise is rising due to technical issues of the detector.

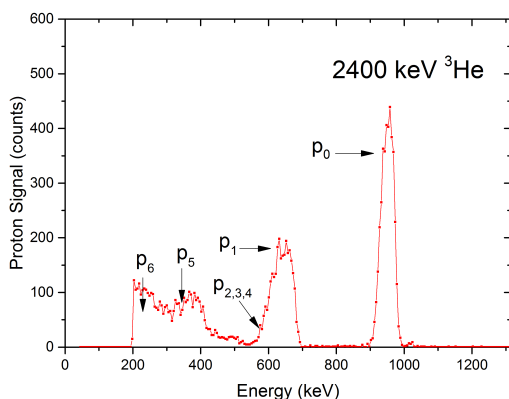


FIG. 1: Proton energy spectrum at an <sup>3</sup>He incident energy of 2400 keV with the clear contribution of different proton sources. Very little noise under the peak is recorded.

As shown in figures 2 and 3, as the energy of the incident beam increases, more background signal is detected. These protons are the result of the reaction of <sup>3</sup>He with silicon.

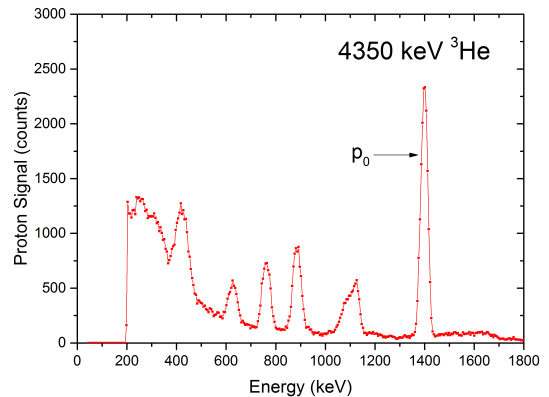


FIG. 2: Proton energy spectrum at an <sup>3</sup>He incident energy of 4350 keV with the different proton signals. Evident background signal under the p<sub>2</sub> peak is visible.

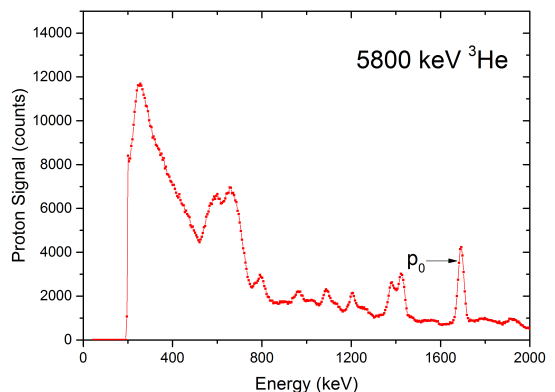


FIG. 3: Proton energy spectrum at an <sup>3</sup>He incident energy of 5800 keV with the different proton signals. Here the integral of the peak is the same order of the background integral.

The fact that the number of detected protons is proportional to the cross section is used to get the relative shape of the cross section. This is done by plotting, for the p<sub>0</sub> peak, the integral of counts versus the mean energy; however, the signal belonging to silicon reactions must be removed from the integral. This is done by subtracting a trapeze under the examined peak. The conversion from number of protons to cross section units is achieved by using equation 1, where  $r$  is the number of protons,  $\Phi$  is the flux of particles through the detector, experimentally known, and  $\rho$  is the density of oxygen atoms in the sample.

$$r = \Phi\rho\sigma \quad (1)$$

Apart from the NRA analysis, the backscattered  $^3\text{He}$  was detected in the  $165^\circ$  detector. By analyzing the data with SIMNRA program [6], which incorporates the Backscattering Rutherford cross section, one can fit the whole profile to a single curve. Doing this, the exact quantity of each element is extracted, and in particular, the oxygen in the sample is accurately quantified. In Fig. 4 a typical RBS profile is shown, with all the elements identified and distinguished by noticeable edges. The position of the narrow peak of gold, recorded for all the measured energies, is used for the channel-energy calibration, whereas the number of counts gives the uncertainty for the beam current measurements.

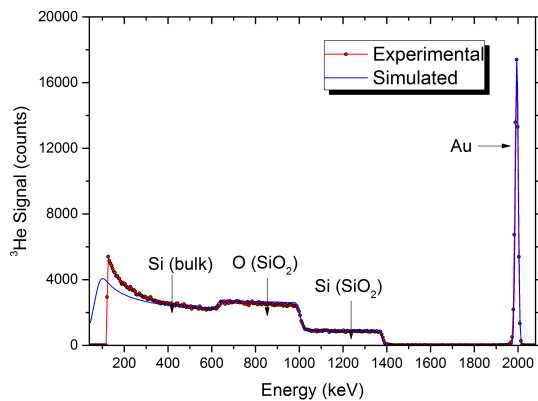


FIG. 4: Experimental (black circles) and fitted Rutherford Backscattering profile (red line), for an incident  $^3\text{He}$  energy of 2150 keV and a longer exposure measurement ( $20 \mu\text{C}$ ). The elements present in the sample are identified according to the different shapes.

The shape of the cross section for the  $^{16}\text{O}(^3\text{He,p})^{18}\text{F}$  reaction is shown in Fig. 5. A previous rectification in the energy had to be done in order to describe better the energy of the helium atoms when reacting with oxygen: the energy loss in the gold film and half the energy loss in the  $\text{SiO}_2$  layers was subtracted from the incident energy, being about 100 keV for high beam energies and up to 200 keV for the lowest incident energies. The statistical uncertainty of individual data points is the square root of the count integral of the signal peak and the background signal. The total uncertainty will be the quadratic sum of all of them as they are physically independent.

Several observations on the shape of the cross section are worth mentioning: first, the unexpected contour suggests interesting nuclear resonances in this range of energies either in the oxygen atom or in the fluorine one. This opens a wide new field of deep investigation, which unfortunately is way far from the purpose of this thesis. On the other hand, the width of the peaks is about the

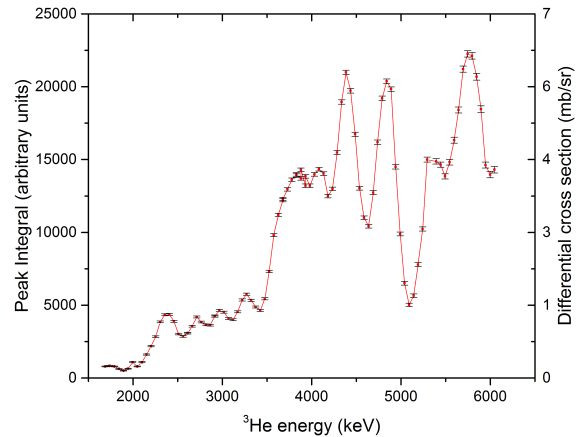


FIG. 5:  $p_0$  proton peak integrals and cross section for  $^{16}\text{O}(^3\text{He,p})^{18}\text{F}$  reaction, for a scattering angle of  $135^\circ$  measured with a 680-nm-thick  $\text{SiO}_2$  layer on Si 001 sample.

order of magnitude of the previously mentioned stopping power of  $^3\text{He}$  in the sample, i.e. the energy loss that the incident helium undergo by going through the different layers of material. This fact, characteristic of bulk materials, could alter the cross section shape and make these peaks different or even non existing. Therefore, this cross section measurement is non reliable and the same experiment needs to be carried out for a thinner  $\text{SiO}_2$  sample.

### C. 91-nm-thick $\text{SiO}_2$ sample

In order to minimize the stopping, a thinner sample is studied. For this new experiment, some other considerations besides the sample thickness must be taken into account. On the one hand, it is preferable to decrease the energy step so that the profile is better defined. On the other hand, a higher amount of charge needs to be collected in order to achieve acceptable statistical error.

Additionally, another source of error in the proton peak integration was observed: the background signal subtraction, which rises with the energy, presents a non-uniform behaviour with some notable peaks. When a peak of the silicon signal turns out to be in the same position as the oxygen related peak, then the trapeze subtraction yields a high error, up to 30% (Fig. 6). To overcome this obstacle, another sample was prepared: pure silicon with 10 nm of Au on top [7].

Thus, the new parameters for the measurements will be the following: 91-nm-thick  $\text{SiO}_2$  layer on bulk Si and 10 nm Au on top; energy step of 25 keV and accumulated charge of  $10 \mu\text{C}$ .

The cross section for the thinner sample is plotted together with the 680-nm-thick cross section to allow a straightforward comparison (Fig. 7). Just like in the first sample, the energy loss corrections were done, im-

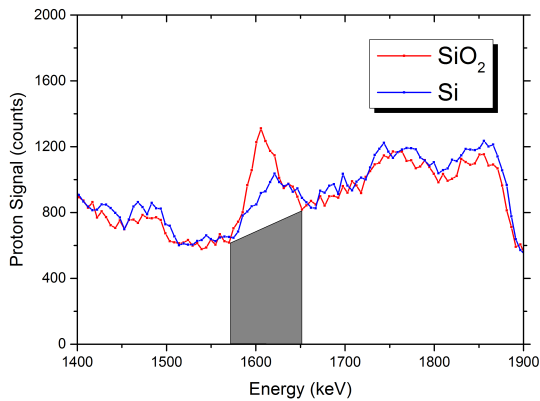


FIG. 6: Close up of the oxygen peak with a silicon peak underneath, measured at 5200 keV. The grey figure shows the subtracted background signal by the trapeze method, as done in the 680 nm sample analysis, and how wrong this assumption would have been.

plying in this case a smaller reduction: between 16 and 33 keV.

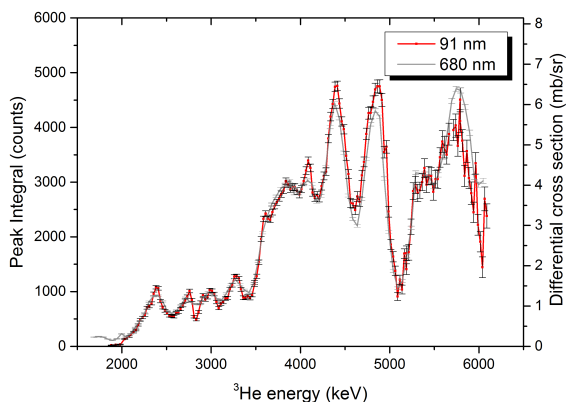


FIG. 7:  $p_0$  proton peak integral and cross section for  $^{16}\text{O}({}^3\text{He},p){}^{18}\text{F}$  reaction, for a scattering angle of  $135^\circ$  and measured with a 91-nm-thick  $\text{SiO}_2$  layer on Si 001 sample. The cross section calculated with the 680-nm-thick sample is also plotted in grey.

As one can see in this figure, the shape is practically the same for both thicknesses. No significant difference is observed in terms of the width of the high energetic resonances (4500, 5000 keV): this means that both samples can be considered thin layers and the stopping power is not influencing the peak width. However, for the low energetic resonances (2000-3500 keV), slight deviations can be detected; as both the height and the width of peaks are relatively small, we can ascribe the change of contour to the different energy steps. Regarding the source for the deviations located at high energies, it is with certainty

the subtracting background method, which is different for both experiments, and diverges in accuracy for high energies.

#### IV. VERIFICATION OF THE METHOD

As the main purpose of this thesis is to determine the quantity of oxygen in tungsten, we have performed the same experiment on some tungsten samples -three-, in order to allow a comparison between them and prove the method. Two of the samples are intentionally oxidized and the other is naturally oxidized (by being in contact with the atmosphere). The experiment was performed at the energy of the highest probability of reaction, i.e. the maximum of the cross section, so that the greatest number of protons would be detected. In this case we chose arbitrarily among the two main peaks, the energy of 4450 keV.

In Fig. 8, the backscattering spectra of all three samples is shown: the highest oxidized sample (W\_2) shows a clear step in the profile which allows a good fit and thus, a quantification of oxygen. However, in practice, this fit with one single step would be very rough and give a big range of uncertainty too big in the determination of oxygen content. Nevertheless, when examining the not so high oxidized samples, a change in the RBS profile is not even discerned. This clearly demonstrates that RBS is not suitable to detect light elements in a heavy target.

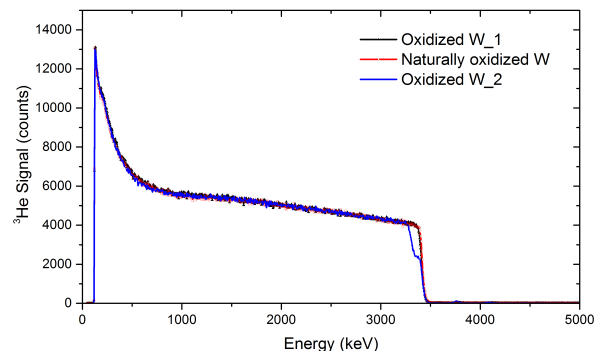


FIG. 8: RBS spectra for the three samples of tungsten measured at an energy of 4450 keV. The blue step located at about 3500 keV is the result of oxygen presence, yet too small to be accurately quantified. The other two profiles remain practically unaffected by the presence of oxygen.

Regarding the NRA method, Fig. 9 manifests the clear peaks related to oxygen: as for the two oxidized samples, the peaks are enormous, and their integration would give a straightforward value of O atoms. Nevertheless, focusing on the naturally oxidized sample, a small protuberance, yet detectable, is manifested at the same position as the  $p_0$  peaks, for the other two tungsten and the  $\text{SiO}_2$  references. By repeating the measurement with

more charge collected, a clean and evident peak, centered at 1440 keV, is shown (Fig. 10), big enough to allow an integration with a reasonable statistical error, used to give an accurate value of oxygen quantity in the analyzed sample.

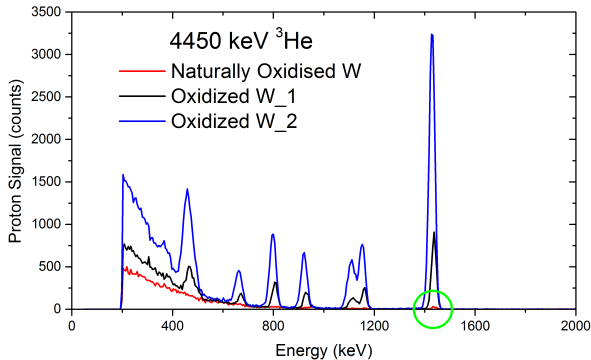


FIG. 9: Proton spectra measured at 4450 keV for the three tungsten samples. The evident peaky profiles (blue, black) belong to the purposely oxidized, whereas the green circle points out a small but detectable peak for the naturally oxidized case. A charge of 5  $\mu\text{C}$  was collected.

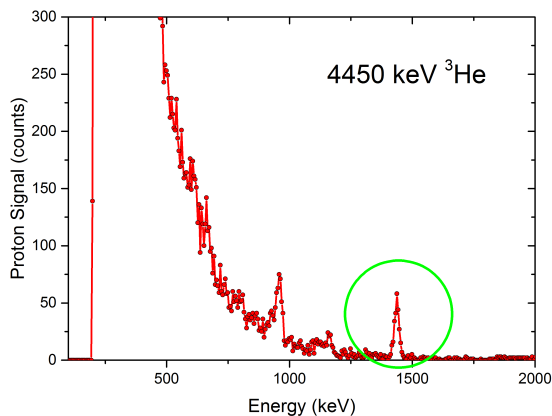


FIG. 10: Close up of the natural tungsten sample proton profile, taken at the maximum of the cross section, with a total amount of charge collected of 10  $\mu\text{C}$ .

## V. CONCLUSIONS

In this work, the differential cross section for  $^{16}\text{O}(^3\text{He},\text{p})^{18}\text{F}$  has been experimentally determined in an energy range from 1800 keV to 6100 keV for a scattering angle of  $135^\circ$ . The shape of the cross section agrees with the study done by J.C.P Heggie *et al.* measured by means of the deexciting  $\gamma$  rays [5].

For high energies, the cross section is less reliable due to large background signal, as a result of reactions of incoming  $^3\text{He}$  with the silicon present in the sample.

A method to quantify oxygen in tungsten via Nuclear Beam Analysis has been verified, very useful when it comes to small concentrations of oxygen and the Rutherford Backscattering method is not powerful enough. This thesis leaves a door open to further analysis, specially focused on the cross section, and a generalization method for any single angle.

## Acknowledgments

I would like to thank my thesis advisor, Dr. Schwarz-Selinger, for trusting me, his permanent attention and motivation for this project, even with his unbelievably packed schedule. Without him this work would have never been accomplished.

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My sincere thanks to my fellow lab-mate, V. Burwitz: his endless patience at the beginning and the interesting discussions we shared at the end, certainly contributed to make this thesis milder and more pleasant to do.

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And last but not least, my kindest words for my best friends: those who I shared with the richest adventure of my life, the career of physics.

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- [7] The reason to have gold on top of this additional sample is not related to calibration nor control of the current, but to a technical issue with the electronics of the detector: the fact that one sample would have a conductor material on top and the other would be mainly semiconductor material would give different signals, thus, the subtraction with a pure Si sample would be no longer beneficial