Thickness and uniformity analysis of thin and heat-resistant targets

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Abstract. The thickness and uniformity characterisation of the first prototypes of thin tellurium and germanium targets evaporated on Highly Oriented Pyrolytic Graphite for NUMEN project is reported. The contribution of such targets to the energy resolution on reaction ejectiles is evaluated.

1 Introduction

The NUMEN (NUclear Matrix Elements for Neutrinoless double β decay) project proposes an innovative technique to access experimental information on Nuclear Matrix Elements (NMEs) involved in neutrinoless double β decay (0νββ), by measuring the differential cross section of heavy-ion induced Double Charge Exchange (DCE) reactions. The idea to use DCE reactions to obtain information on NME of 0νββ is based on the fact that the two processes are characterized by important similarities, as for example the same initial and final nuclear states, and a similar mathematical structure of the transition operators [1,2].

So far, the NMEs have been calculated using different theoretical models, but the discrepancies between the values from the various calculations are large. In addition, experimental constraints are loose. The main goal of NUMEN is to establish an alternative experimental path for the determination of the 0νββ NMEs. An experimental campaign has already started by using targets made with 0νββ candidate isotopes [1]. The experiments are performed at INFN - Laboratori Nazionali del Sud (LNS, Catania, Italy) mainly with ^{18}O and ^{20}Ne beams produced by the LNS cyclotron at 15-60 AMeV and with the MAGNEX spectrometer to detect the reaction products. Since the DCE reactions are characterised by very small cross-sections, an improvement of the LNS cyclotron is underway to enhance the intensity of the beams up to 10^{13} pps. Consequently, an upgrade of the whole detection apparatus is also ongoing [3-5] to withstand these intense beams. In particular, a new target system, capable of dissipating the power released by these beams is under study [4].

In the following, the requirements for NUMEN targets are discussed. Then, the characterisation of tellurium and germanium target prototypes is described in detail. On the basis of thickness and non-uniformity measurements, the impact of the targets on the energy resolution of the ejectile is evaluated.

2 New target system for NUMEN

The power released by the intense beam in the NUMEN targets is of the order of 1 W, two orders of magnitude higher than the typical power released in a target for nuclear experiments. Analytical and numerical thermal calculations have been performed with the MATLAB framework, which allows to conclude that a stand-alone target is not able to dissipate this amount of power [6]. The heat dissipation can be enhanced by depositing the target material on a highly thermally conductive substrate, i.e. High Oriented Pyrolytic Graphite (HOPG), characterised by a thermal conductivity which could range between 1700 and 1950 W m^{-1} K^{-1} [7-8]. It has been estimated that a 450 μg/cm^2-thick HOPG is able to dissipate up to 10 W [9]. Moreover, a dedicated cooling system has also been designed to help the heat dissipation. The target will be encased in a copper holder, mounted on top of a cryocooler and kept at 40 K [3]. Thanks to the HOPG and associated cooling system, the target reaches, in a few hundred microseconds, a

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maximum temperature lower than the melting point [9,10]. Thus, the NUMEN targets will be characterised by the presence of a substrate thicker than a usual target backing. Thick HOPG substrates could produce significant straggling and cause a degradation of the energy spread of the products. To distinguish the ground state from the ground state of the DCE transition, the energy resolution of the reaction products should be lower than the energy of the first excited state of the residual nuclei, typically ≈ 500 keV. In order to obtain this resolution, and because of the thick HOPG backing, the target material thickness (i.e. the tellurium and germanium layer) should range between 250 and 500 μg/cm^2. The presence of non-uniformities in the HOPG and/or in the target can also contribute to worsening the energy resolution. For this reason, as will be explained in the following, a detailed analysis of the non-uniformity has to be performed. The maximum acceptable non-uniformity has to be estimated for each reaction considered.

3 Tellurium and germanium target production and analysis

First depositions of tellurium and germanium have been produced with an electron-beam evaporation source at the TRUSTECH Company laboratories (Turin, Italy), specialized in micro and nanotechnology. Natural tellurium and germanium were used to save costly isotopes. To define the best parameters for the evaporation (see [10,11] for details), several attempts have been made by varying the evaporation rate, by changing the HOPG substrate thickness and temperature, and using a deposition buffer for helping the diffusion of the evaporated material onto the backing surface. For the materials considered in the present work, the best results were obtained by performing the evaporation process at standard conditions, i.e. without heating of the backing, without a deposition buffer and by applying a low evaporation rate (0.2-0.5 Å/s). For the mentioned depositions, the HOPG substrate (nominal thickness: 450 μg/cm^2) was obtained from the OPTIGRAPH Company (Berlin, Germany). It has to be underlined that the HOPG substrate is always analysed before the evaporation using the alpha-particle transmission technique, to determine its actual average thickness and uniformity. This procedure showed that the HOPG foils used for these first studies have a significant non-uniformity, both globally and locally (i.e. inside the alpha spot). The average thickness could differ from the nominal value by up to 50%, with a local non-uniformity variation from 5% to 25%. Because of these characteristics, these kinds of HOPG foils cannot be used as backing for the final NUMEN targets. However, they have been useful for the definition of the evaporation procedure. A detailed characterisation of the HOPG foils will be reported elsewhere. The here presented target prototypes have been characterised using Field Emission Scanning Electron Microscope (FESEM), Rutherford Backscattering Spectrometry (RBS) and Alpha Particle Transmission (APT). FESEM provides information on the morphology of the sample surface. The analyses have been performed with a Zeiss Merlin instrument , hosted in the laboratories at the Department of Applied Science and Technology of the Polytechnic of Turin, with a magnification of up to one million and a resolution of 0.8 nm.

The RBS technique gives information on the thickness, and on the elemental composition of the sample. One of the advantages of the RBS technique is that the study of an evaporated film is not influenced by the type of sample substrate used. RBS measurements have been performed at the AN2000 accelerator facility of the INFN – Laboratori Nazionali di Legnaro (Padova, Italy). This facility provides proton and alpha beams with energies up to 2 MeV with a circular spot diameter of around 1 mm (Full Width at Half Maximum). The proton beam has been exploited to study the HOPG backings, whilst for studying the target depositions, the 2 MeV alpha beam was used. The scattered ions were detected with a silicon detector at an angle of 160° with respect to the beam. The uncertainty of these measurements has been estimated around 10%.

The APT method was used to characterise the sample in terms of thickness and uniformity, with the set-up hosted at the Department of Applied Science and Technology of the Polytechnic of Turin [11]. The average thickness value was deduced from measuring the energy lost by an alpha particle, emitted by an ^{241}Am source, in traversing the target. In this procedure, the main source of uncertainty is given by the stopping power values obtained with the SRIM code [12]. According to the comparisons of SRIM calculations with measured stopping powers [12], the uncertainty of the calculations, for the materials studied in the present work, is in the order of 5%. The target non-uniformity can be deduced, as described in [13], by comparing the Full Width at Half Maximum (FWHM) of the experimental spectrum with the FWHM of a simulated spectrum, obtained by assuming a uniform target with the measured average thickness and convoluting it using the detector energy resolution as input. In the present analysis, the simulations were performed with the SRIM [12] program.

3.1 Tellurium target

Figures 1-2 show the results of the FESEM, RBS and APT analyses of a tellurium target evaporated at the standard conditions previously mentioned. In the FESEM image (Figure 1a), the presence of tellurium grains, with dimensions in the order of tens of nanometers can be observed. The RBS analysis (Figure 1b) allows to determine the target thickness (322 ± 32 μg/cm^2) and shows that no contaminants are present in the target. Figure 2 shows the residual energy spectra for the alpha-particles passing through the HOPG backing alone (top) and the HOPG backing + tellurium (bottom). The black line is the experimental spectrum and the red and green lines are the simulated spectra used to determine the non-uniformity inside the alpha-particles spot (also referred to as "local non-uniformity"), as described in [13].

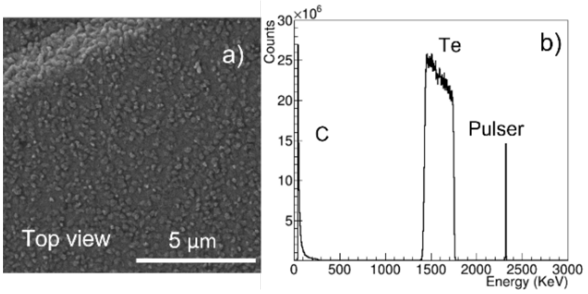

Fig. 1. (a) FESEM view of Te sample, deposited on a HOPG substrate at room temperature without any buffer (b) RBS spectrum of the Te target, measured with a 2.2 MeV alpha beam, detected at an angle of 160° with respect to the incident beam (see text for details).

According to this analysis, the HOPG substrate resulted in a thicker than the nominal value by about 20% (533 ± 27 μg/cm^2) and presented a large local non-uniformity of 24%. Regarding the thickness of the deposited tellurium, the value deduced by APT (323 ± 16 μg/cm^2) is in excellent agreement with the one obtained with RBS. The simulated spectrum (green curve) already includes the HOPG backing non-uniformity, thus the difference in the FWHM of the simulated and the experimental spectra is attributed to the tellurium non-uniformity, which is estimated to be about 10%.

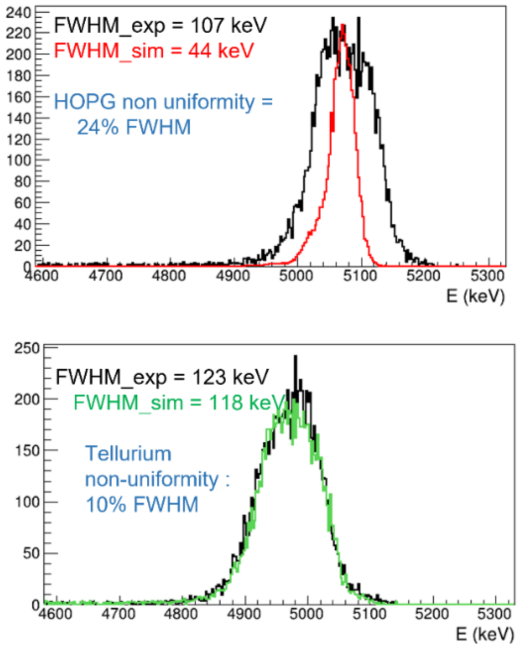

Fig. 2. Experimental (black histogram) and simulated (red and green histograms) energy spectra of α particles passing through a HOPG foil (top) and through a HOPG + Te target (bottom).

3.2 Germanium target

In the present section, similar analyses as discussed for the tellurium target are reported for a germanium target prototype produced at standard conditions. The FESEM image (Figure 3a) shows a very flat and uniform germanium deposition, with the presence of some particles with a diameter of about 0.5 micrometers.

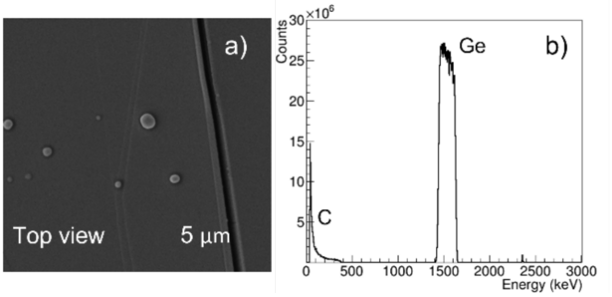

Fig. 3. (a) FESEM view of Ge sample, deposited on a HOPG substrate at room temperature without any buffer (b) RBS spectrum of the Ge target, measured with a 2.2 MeV alpha beam, detected at an angle of 160° with respect to the incident beam (see text for details).

Also in this case, the RBS analysis (Figure 3b) confirms the high purity of the deposited material, and the germanium thickness deduced from this analysis (142 ± 14 μg/cm^2) is in good agreement with the one obtained from APT measurements (152 ± 8 μg/cm^2). According to the alpha spectra analysis (Figure 4), the HOPG thickness is 496 ± 25 μg/cm^2 (i.e., 10% thicker than the nominal value) and the local non-uniformity was estimated to be about 10%. Instead, the germanium non-uniformity resulted less than 10%, in agreement with the fact that the surface seems flatter than the tellurium one.

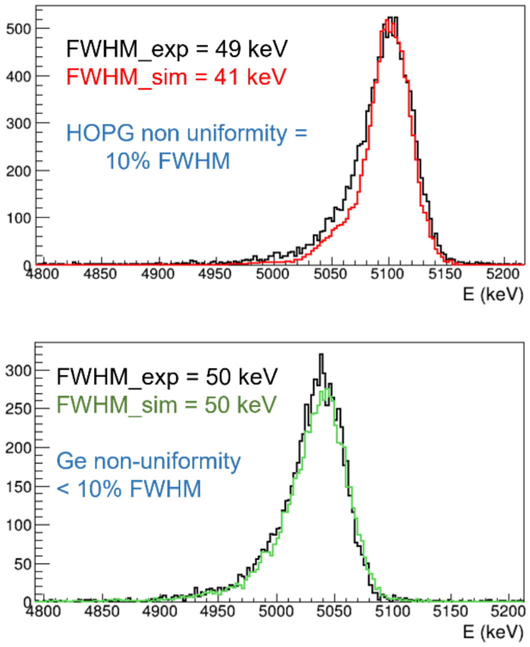

Fig. 4. Experimental (black line) and simulated (red and green lines) energy spectra of α particles passing through a HOPG foil (top) and through a HOPG + Ge foil (bottom).

4 Energy resolution estimations

To judge the quality of the target prototypes, analytical calculations were performed to estimate the energy resolution of an ejectile, detected at 0° with respect to the incident beam, produced in a DCE reaction in the considered target. For the tellurium case, the reaction ^{130}Te(^{20}Ne,^{20}O)^{130}Xe was considered. The first excited state of ^{130}Xe has an energy of 536 keV, and therefore, the energy resolution of the ejectile needs to be below this value. The calculations take into account the contribution of the cyclotron and the MAGNEX spectrometer, as well as the target effects (i.e. dispersion due to the random reaction point in the Te film, energy straggling in the Te film and HOPG backing, non-uniformity of the Te layer and HOPG substrate). Since calculations do not take into account other factors which contribute to energy spread (e.g. statistical counting, peak shape, presence of background, etc.), it has been estimated, thus to have a safety margin, that the energy resolution value should be at least 10% less than the energy gap. For a target with the characteristics of the Te prototype (^{130}Te film 323 μg/cm^2 thick with a non-uniformity of 10% on a HOPG backing 533 μg/cm^2 thick with a non-uniformity of 24%), the ejectile total energy resolution has been estimated to be 525 keV, which is very close to the energy of the first excited state. To investigate the role played by the backing non-uniformity, a series of calculations was performed by assuming a 323-μg/cm^2 ^{130}Te thick deposited on a 450 μg/cm^2 HOPG substrate with a non-uniformity ranging from 5% to 25%. The energy resolution values calculated as a function of the HOPG non-uniformity are given in Figure 5 (orange points). The plot also shows the lower limit of the energy resolution due to the cyclotron and the spectrometer, as well as the energy of the ^{130}Xe first excited state. A second set of calculations has also been performed, assuming a Te non-uniformity of 20% (blue squares). It is clear that the non-uniformity of the HOPG substrate contributes more to the energy resolution than the tellurium layer.

The same type of analysis was performed for the Ge target described in the previous section. In this case, the considered reaction was ^{76}Ge (^{20}Ne,^{20}O)^{76}Se. The ^{76}Se first excited state has an energy of 559 keV. For a ^{76}Ge target with the characteristics of the prototype (Ge thickness 153 μg/cm^2 with an uniformity of 10% on a 496 μg/cm^2 thick HOPG backing with a non-uniformity of 10%), the ejectile energy resolution has been estimated to be 445 keV, which is below the maximum acceptable value for the energy resolution. Given these target characteristics, it has been estimated that increasing the Ge thickness to 300 μg/cm^2 would still give an acceptable energy resolution (477 keV).

5 Conclusion and outlook

Here, the characterisation of the first tellurium and germanium target prototypes for the NUMEN project was described. Calculations to estimate the total energy resolution of DCE reaction products expected from such targets were performed. Results show that the non-uniformity of the HOPG backing plays a very important role. According to the results presented in the previous section, for Ge and Te targets, a 450-μg/cm^2 HOPG with maximum value of non-uniformity of 20% can be used. However, a HOPG backing with a non-uniformity smaller than 10% is preferred, in order to maintain a resolution close to the limit reachable with a uniform target+HOPG assembly. This would help disentangling energy peaks also in the presence of factors not taken into account in the calculations. For this reason, new HOPG substrates provided by different companies are under study. Concerning the depositions of tellurium and germanium on HOPG supports satisfactory uniformities have been obtained for these materials at standard evaporation conditions. A drawback of the evaporator used in the present study was the large amount of material required (from 10 to 20 g) to perform the evaporation, due to geometrical reasons. Given that the final NUMEN targets will be made of isotopically enriched material, it is necessary to reduce this quantity by at least an order of magnitude. For this reason, a new series of evaporations is on-going at LNS, with the aim of producing target prototypes with characteristics similar to the ones described in this work, using though amounts of material lower than 1 g.

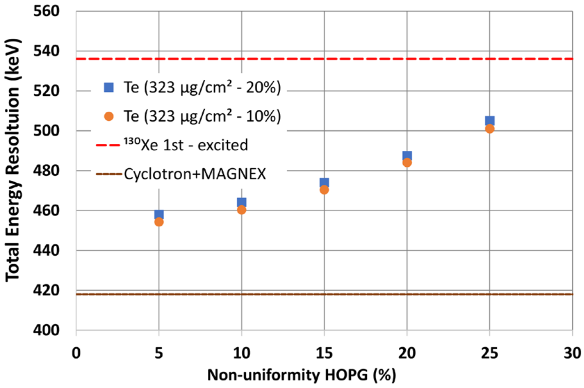

Fig. 5. Total energy resolution versus HOPG non-uniformity, for the ejectile of the reaction ^{130}Te(^{20}Ne,^{20}O)^{130}Xe, detected at 0° (see details in text). The thickness of the HOPG is 450 μg/cm^2.

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