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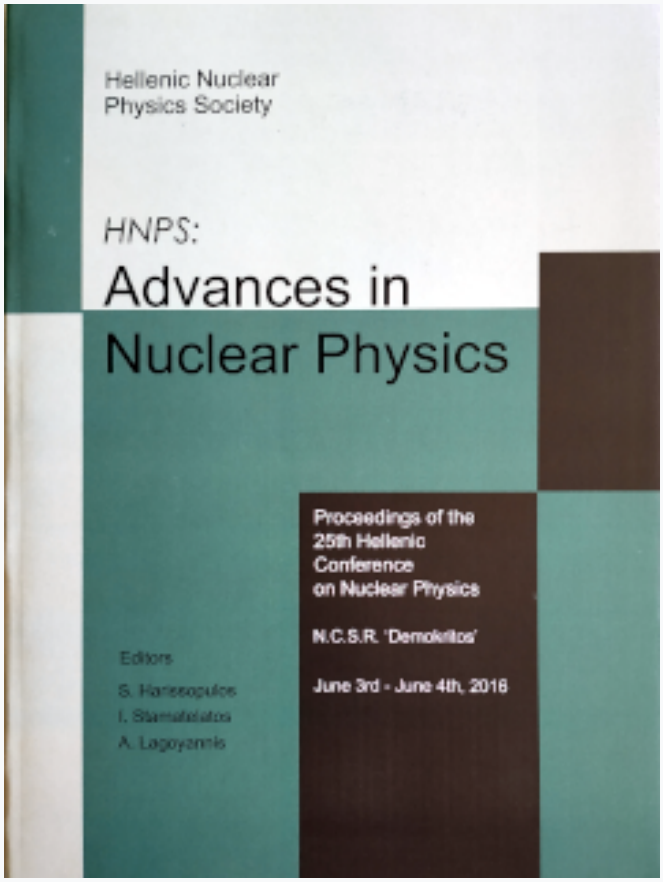

A benchmarking procedure for PIGE related differential cross-sections

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A benchmarking procedure for PIGE related differential cross-sections

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Abstract The application of standard - less PIGE requires the a priori knowledge of the differential cross section of the reaction used for the quantification of each detected light element. Towards this end, a lot of datasets have been published the last few years from several laboratories around the world. The discrepancies found can be resolved by applying a rigorous benchmarking procedure through the measurement of thick target yields. Such a procedure is proposed in the present paper and is applied in the case of the $^{19}F(p,p'\gamma)^{19}F$ reaction.

Keywords Benchmarking, PIGE, ^{19}F, ^{23}Na

INTRODUCTION

Proton Induced Gamma ray Emission (PIGE) is a well-known Ion Beam Analysis (IBA) technique used for identification and quantification of mainly low Z (Z<13) elements. The quantification of the detected elements is based, since its first applications back to the 1960's, on the comparison of the accumulated gamma ray yield between the under study sample and a standard one of known concentration. As the production of the gamma-ray yield depends on the stopping power of the studied matrix, the successful application of the method requires the comparison to be performed against a standard of similar stoichiometry, at least for the major elements of the sample. The expansion of Ion Beam Analysis in a variety of interdisciplinary fields would therefore require a large number of standards to be owned by an analytical laboratory.

A standard-less approach would, on the other hand, require the precise knowledge of fundamental quantities entering the calculation of the yield such as the stopping power and the cross section of the reaction used for the detection of the element. While the stopping power of protons in various matrices can be reliably calculated within a 5% uncertainty using suitable codes like SRIM [1], there is an evident lack of reliable cross sections in literature. Until recently, only the publication of C. Boni et al.[2] aimed at measuring differential cross sections suitable for PIGE analysis. A few years ago, IAEA has launched a Cooperative Research Program [3] aiming at curing the lack of reliable differential cross section data in the literature. This initiative resulted in a number of publications [4–11] dealing with the measurement of differential cross sections of gamma ray producing reactions. While in some

cases, like the one of 23 Na(p,p'γ)23 Na, the measurements from different laboratories coincide, there are some cases where the different experimental setups, as well as, the different normalization procedures involved in the determination of the differential cross sections, led to discrepancies in the data which, in some cases, can reach up to a factor of 5. These large discrepancies could be resolved by applying a benchmarking procedure. Such a rigorous procedure is proposed in the present work.

EXPERIMENTAL SETUP

The proposed benchmarking procedure was applied in the case of the $^{19}F(p,p'\gamma)^{19}F$ reaction where the measured differential cross sections [2, 6, 13–17] exhibit discrepancies especially for the $^{19}F(p,p'\gamma_{2-0})^{19}F$ reaction. Two multielemental standards were used containing Fluorine and Sodium: a NIST standard (Phosphate Rock 120b) and a home-made one containing 5.3% Fluorine and 5.3% Sodium in a carbon matrix. The existence of Sodium in both standards enabled the cross checking of the method and the elimination of possible systematic uncertainties as there is a remarkable agreement in the available data [13–14, 18–23] for the $^{23}Na(p,p\gamma_{1-0})^{23}Na$ reaction, at least for the energy range between 2000 and 4000 keV. The targets were placed in the center of a cylindrical chamber which was electrically isolated in order to act as a Faraday cup. The proton beam, delivered by the 5.5 MV Tandem Accelerator of the Institute of Nuclear and Particle Physics (I.N.P.P.), National Centre of Scientific Research (N.C.S.R.) "Demokritos", was led in the chamber through a system of tantalum collimators placed ~1 m before the target. A voltage of -300 V was applied to the collimators in order to suppress the emission of secondary electrons. The beam energy ranged from 2000 to 4000 keV with a variable step, while the current was kept below 20 nA throughout the experiment to avoid pile-up effects and high ADC dead time. The detection of the emitted γ-rays from ^{23}Na and ^{19}F was achieved with the use of two HPGe detectors of 18% and 100% relative efficiency placed at 55^o and 90^o with respect to the beam axis. Their efficiency were calculated using a ^{152}Eu and a ^{226}Ra calibrated point sources, mounted at the position of the target. The detector signals where processed and recorded using standard NIM modules.

ANALYSIS AND RESULTS

Two integrating codes, SPECTRW [24] and TV [25] were used for the analysis of the 110, 197 and 440 keV photopeaks produced by the $^{19}F(p,p'\gamma_{1-0})^{19}F$, $^{19}F(p,p'\gamma_{2-0})^{19}F$ and $^{23}Na(p,p'\gamma_{1-0})^{23}Na$ reactions, respectively. The results from both codes agreed within 1% eliminating thus possible systematic errors coming from the background subtraction or the integrating procedure. The yields were calculated using

$$Y = \frac{N}{4\pi Q \varepsilon_{abs}} \quad (1)$$

where N is the integrated area corrected for the dead time, Q is the accumulated charge and ε_{abs} is the γ - ray detectors efficiency. It has to be noted that the yields calculated with this procedure correspond to multielemental targets.

In order to facilitate the thick target yields calculation, a c++ code was developed that takes as input the differential cross sections of the reaction under examination and the composition of the multielemental target and outputs the yield produced by this target. As a first step, the code divides the target into a large number of sublayers. As the beam enters the first layer, the yield is calculated by multiplying the corresponding cross section with the amount of the investigated element. After passing the first layer the energy loss of the beam is calculated using either the Ziegler, Biersak and Littmark [30] or the SRIM 2013 [1] compilation for the stopping power. The difference found using both approaches is only 2.6% at high energies. In the case of composite targets Bragg's rule [27] was applied. The same procedure is followed for each layer of the target until the energy of the beam drops to zero or there are no cross section data available, whichever comes first. The number of layers is user-defined, enabling thus the correct yield calculation in the regions where the cross section is changing rapidly i.e. in the vicinity of resonances. The case of ^{23}Na was chosen as a test case for the code as there is a remarkable agreement between most of the published differential cross section data. The elemental composition of the two targets along with the existing data from literature were fed to the program and the expected yield was calculated. It has to be noted that the calculated yields were normalized by adding an initial yield but only for presentation reasons. The comparison is performed between the shapes or slopes of the thick target yields and not of the absolute values. The same procedure was followed for the ^{19}F(p,p'γ_{1-0})^{19}F and ^{19}F(p,p'γ_{2-0})^{19}F reactions using the available datasets and the results are presented in Figs 1 and 2, respectively.

DISCUSSION

The present pure elemental thick target yields for the ^{23}Na(p,p'γ_{1-0})^{23}Na are in very good agreement with the ones reported by M. Chiari et al. [12], Anttila et al. [30] and M. J. Kenny et al. [27]. There is also a fair agreement with the data of F. Bodart et al. [31] up to 2200 keV and A. Savidou et al. [32] up to 3200 keV. The only dataset having a large deviation from all the others is the one measured by Kiss et al. [33] where, judging by the last point at 4200 keV which is lower than that at 3800 keV, there is evidently an error. The agreement is a first indication that the present measurement has no significant systematic errors. The same comparison was also performed for the measured thick target yields of the ^{19}F(p,p'γ_{2-0})^{19}F reaction with the available data. While the present data agree with the ones by Kenny et al. the data of Savidou et al. are lower by a factor of ~1.8 which, in their turn, agree with those

of A. Anttila et al. and A. Z. Kiss et al. The latter two datasets are not indepedent, as Kiss et al. have normalized their data with those of A. Anttila et al. at 2400 keV.

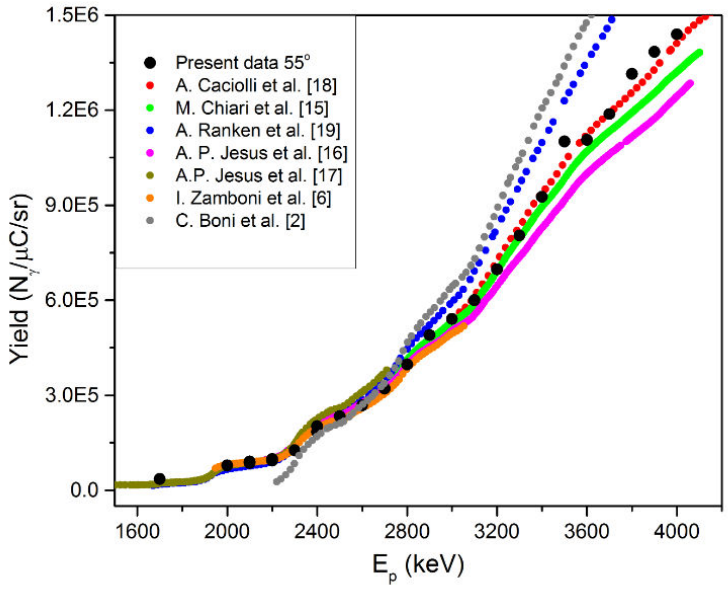

Fig 1: Comparison of the integrated differential cross sections of the 110 keV de-excitation of ^{19}F with the present data for the phosphate rock standard

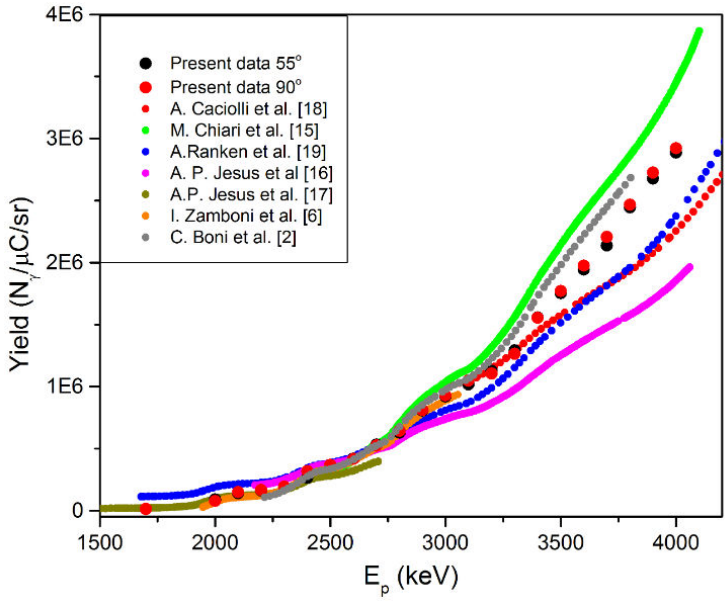

Fig 2: The integrated differential cross sections of the ^{19}F(p,p'γ_{2-0})^{19}F reaction compared with the present thick target yields for the case of the phosphate rock standard

A careful examination of the differential cross section data for this reaction at energies below 2000 keV, reveal that all the authors agree to the existence of a broad resonance at E_p = 1950 keV, even if they disagree about its actual height. A simple integration of the cross sections between 1900 and 2000 keV, even with the lowest cross section reported, resulted in a yield of ~ 3 × 10^6 which is much higher than the ones measured by Savidou and Kiss. This fact

indicates that these two datasets cannot successfully describe the emitted yield and were not taken into account in this benchmarking procedure.

The comparison of the shapes of the integrated cross section yields, as were calculated using the aforementioned c++ code, in the case of the $^{23}Na(p,p'\gamma_{1-0})^{23}Na$ reaction, reveals that the majority of the datasets under consideration can reliably reproduce the thick target yields measured with the exception of Caciolli et al. [19]. This agreement verifies not only the reliability of the conducted experiment but also that the assumptions and quantities implemented in the code are valid. The same comparison (Fig. 1) for the 110 keV de - excitation of the first excited level of ^{19}F, shows a very good agreement with the data of A. Caciolli et al. [16], I. Zamboni et al. [6], A. P. Jesus et al. [15] and M. Chiari et al. [13]. The unpublished data of Jesus et al. [14] are slightly underestimating the experimental yield while C. Boni et al. [2] and A. Ranken et al. [17] fail to reproduce the experimental thick target yields. The latter was expected as these differential cross sections deviate from the majority of the measured data. The above described situation is very different in the case of the 197 keV gamma - ray emitted from ^{19}F. In Fig. 2 only the dataset of I. Zamboni et al. reproduces successfully the obtained thick target yields. Unfortunately, this data set spans over a limited energy range from ~ 2 up to 3 MeV. The fact that most of the available cross section data for the 110 and 197 keV gamma - rays are correlated, i.e. are measured during the same experiment, indicates that the differences could be attributed to the background subtraction. This is not however the case for the present experimental procedure as the ratio peak to background is high due to the thick target used. Moreover, the agreement between the ^{23}Na thick target yields ensures that there were no systematic errors involved.

CONCLUSIONS

During the last decade a lot of differential cross section measurements related to PIGE applications have been published. However, in several cases discrepancies are observed. Aiming at resolving these discrepancies a benchmarking procedure has been proposed in the present work. The method involves the simultaneous measurement of two reactions in a pre-characterized multielemental target. The first reaction should be the one to be benchmarked while the second one corresponds to a well known reaction in order to eliminate possible systematic errors. A computer code was developed in order to facilitate the calculation of thick target yields from differential cross sections. The application of this procedure to the $^{23}Na(p,p'\gamma_{1-0})^{23}Na$ reaction revealed that there is a fairly good agreement between the majority of the published cross section data with the experimental thick target yields. The same procedure in the case of the 110 keV gamma - ray originating from the $^{19}F(p,p'\gamma_{1-0})^{19}F$ reaction showed a fair agreement with most of the available datasets. On the contrary, the existing differential cross section for the $^{19}F(p,p'\gamma_{2-0})^{19}F$ reaction fail to reproduce the shape of the experimental yields, with the exception of the data by I. Zamboni et al., and thus, they cannot be recommended for analytical purposes. The established benchmarking procedure can be applied also to several other cases where important discrepancies are observed.

ACKNOWLEDGEMENTS

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