Benchmarking experiments for the proton backscattering on ^{23}Na , ^{31}P and $^{\text{nat}}\text{S}$ up to 3.5 MeV

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Abstract

The benchmarking procedure in IBA (Ion Beam Analysis) regards the validation of microscopic charged-particle differential cross-section data via the acquisition of EBS (Elastic Backscattering Spectrometry) spectra from uniform thick target of known composition followed by their detailed simulation. In the present work such benchmarking measurements have been performed for the elastic scattering of protons on ^{23}Na , ^{31}P and $^{\text{nat}}\text{S}$ in the energy range of 1-3.5 MeV in steps of 250 keV at three backward angles, at 120.6° , 148.8° and 173.5° in an attempt to validate the existing evaluated cross-section datasets from SigmaCalc and to facilitate their extension at higher energies. The EBS spectra acquired were compared with simulated ones using the DataFurnace code, along with an *a posteriori* treatment of the surface roughness. All the experimental parameters were thoroughly investigated and the results obtained and the discrepancies found are discussed and analyzed.

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1. Introduction

Ion Beam Analysis (IBA) critically depends on the accuracy of the available differential cross sections for the reactions involved. Unfortunately, the existing experimentally determined differential cross-section datasets [1] are in many cases quite scarce and discrepant, thus limiting the applicability of the IBA techniques. The evaluated cross-section data, available through the online calculator SigmaCalc [2], are the most reliable data to be used in analytical studies, since they involve a critical assessment of the available experimental data followed by a proper tuning of the corresponding nuclear model parameters [3]. However, most of the evaluated datasets are still not adequately validated. A carefully designed benchmarking experimental procedure (i.e. the validation of microscopic differential cross-section data via the acquisition of thick target spectra followed by their simulation) is thus mandatory. Benchmarking can also provide feedback for the adjustment of the parameters of the nuclear model used in the evaluation process, and can help in assigning realistic uncertainties to the cross sections. Moreover in the absence of evaluated cross sections, it can indicate recommended experimental datasets. In the past benchmarking results have usually been reported, related to the validity of specific experimentally determined differential cross-section datasets [e.g. 4-6]. However, such measurements have never been performed in a systematic and consistent way, taking into account all the fine steps and details of the benchmarking process.

The present work coherently benchmarks important cross sections relevant to IBA. It also aims at facilitating the extension of the corresponding evaluations to higher energies. The whole experimental procedure for benchmarking the proton elastic scattering on ^{23}Na , ^{31}P and $^{\text{nat}}\text{S}$ involves: (i) the acquisition of EBS thick target spectra from uniform thick targets of known composition with good

counting statistics, (ii) the accurate calibration of the accelerator over a broad energy range, (iii) the minimization of target-related effects such as channeling, and (iv) the *a posteriori* treatment of surface roughness through a detailed mathematical model [7]. The results obtained and the discrepancies found are discussed.

2. Experimental setup

The measurements were performed using the 2 MV Tandatron Accelerator of the Ion Beam Centre of the University of Surrey. Spectra of elastically backscattered protons from ^{23}Na , ^{31}P and $^{\text{nat}}\text{S}$ using uniform thick compound targets were measured and simulated in the energy range 1-3.5 MeV, in steps of 250 keV, at 120.6° , 148.8° and 173.5° with an uncertainty of 0.1° . The scattering angles were measured directly using a beam-line laser and a six-axis goniometer [8]. The goniometer also allowed for the correct positioning of the targets along the z-axis according to their thickness (1-5 mm).

The detection system consisted of three Silicon Surface Barrier (SSB) detectors (thickness of $100\mu\text{m}$), placed at the corresponding angles, along with the standard electronics for spectroscopy. The thick target spectra from the three detectors were simultaneously recorded at each energy point. The detectors were set at a distance of ~ 14 , 12 and 19 cm from the target, with orthogonal slits having a width of about 2 , 2 and 5 mm in front of them, in order to reduce the effective angular uncertainty to $\sim 1^\circ$, 1.4° and 2.3° respectively. The proton beam spot was focused to ~ 1 mm in diameter, while the beam current was kept lower than 20 nA during all measurements, in order to minimize the pileup effects.

The targets used were high-purity ($>99.99\%$), highly pressurized tablets of NaBr and MoS_2 in the case of ^{23}Na and $^{\text{nat}}\text{S}$, and a polished crystalline GaP wafer in the case of ^{31}P . A thin layer of gold (of $\sim 50 \times 10^{15}$ at/cm²) was evaporated on top of all targets in order to protect them from corrosion and for normalization purposes. Before the measurements, they were kept in nitrogen-filled containers with silica gel. The targets were mounted all together on the six-axis goniometer allowing us to avoid

channeling effects by tilting the sample and automatically switch from one target to another for each energy step.

3. Benchmarking steps

Benchmarking is an experimental procedure that needs to be very carefully designed. In order to simulate the thick target spectra acquired, one must investigate and accurately determine all the parameters involved. A thick target spectrum is essentially a convolution of the stopping power and the straggling function in the material, the resolution of the detector and the corresponding scattering cross-section dataset. Several other parameters are also important for accurate simulations, including the beam energy, the ADC calibration and the target roughness, if present. In addition to the above, the code used for the simulation should be capable of taking into account any possible fine structure of the corresponding differential cross section (sharp narrow resonances).

In the present study, the DataFurnace code [9], capable of taking into account the cross-section fine structure and self-consistently fitting multiple spectra, which is of great value when working with multiple detectors, was used for the simulations. It should be noted here that up to now, all popular analytical codes, including DataFurnace, do not take the uncertainties in the experimental differential cross-section datasets into account, while the assessment of the uncertainties in the evaluated datasets has been the subject of recent studies [10, 11]. All the other parameters for the present benchmarking measurements were treated as follows:

3.1 Energy calibration of the GVM accelerator

The final energy of the protons was determined by calibrating the accelerator (GVM) with the use of three narrow resonances, namely the (991.9 ± 0.1) keV one of the $^{27}\text{Al}(p,\gamma)^{28}\text{Si}$ reaction ($\Gamma=110$ eV), the (1747.6 ± 0.9) keV of the $^{13}\text{C}(p,\gamma)^{14}\text{N}$ reaction ($\Gamma=122$ eV) [12] and the (3379 ± 1) keV of the $^{32}\text{S}(p,\gamma)^{32}\text{S}$ reaction ($\Gamma=700$ eV) [13] using a 10% HPGe detector. The linearity of the energy with

respect to accelerating voltage, as shown in Figure 1, was found to be excellent over the whole energy range studied. The uncertainty of the proton energy was calculated to be less than 0.1%.

3.2 Detector resolution and ADC calibration

The simulation of the backscattered protons from the polished GaP target at all three angles studied, compared to the corresponding experimental spectra acquired, enabled simultaneously the calibration of the ADC (Au peak) and the determination of the resolution of all the detectors.

3.3 Stopping power, straggling and plural scattering

The models, which are considered to be the most accurate ones, were used in the DataFurnace code for the simulations. These are the model of Ziegler–Biersack–Littmark [14] for the proton stopping power and the one of Chu & Yang [15] for the straggling function. The effect though, of these two parameters, as well as the effect of plural scattering, especially when a heavy element is present in the target, were investigated in the present study by comparing the spectra obtained at close energies (i.e. at resonances and then, in small energy steps above the resonances) and using different stopping power compilations. It was found that the effect of those three parameters could not be simulated in a satisfactory way at lower energies (deeper in the targets), as it can be observed in Figure 2, for the plural scattering effect. Although each of these parameters has a different dependence on depth, all of them are more pronounced, with decreasing energy of the incoming particles. In order to diminish such problems in the simulations, the analysis was limited relatively close to the surface leaving a narrow window of opportunity for the validation of the cross-section data. The energy step for all the benchmarking runs was thus small, not larger than 250 keV, over the whole energy range studied.

3.4 Roughness of the targets

The roughness of the NaBr and MoS₂ pellets used in the present study was treated *a posteriori* using an algorithm based on the mathematical model developed by Molodtsov et al. [7]. To take into account the possible secondary crossings of protons in the asperities of the target surface, according to the model, the uncorrected simulated spectrum $Y_0(E)$ is convoluted with a parameterization function $f(x)$, which depends on two free parameters, namely the sharpness p of the asperities and σ , a random height, chosen from a Gaussian distribution of variance σ^2 . These parameters were determined using the MINUIT code for χ^2 minimization [16]. As shown in Figure 3, the surface roughness can significantly affect the shape of the spectra and thus, it is very important to treat it carefully when present.

4. Assessment of the uncertainty factors

The assessment of the uncertainties in the benchmarking procedure is very important and must be the result of the detailed study of all the parameters involved in the simulating and validating steps.

The obtained simulated yield is directly related to the stopping power systematics. The effect though of different stopping power compilations (e.g. ZBL and Andersen-Ziegler [17]) in the integrated yield of ~40-50 channels (corresponding to 250 keV from the surface) which were used in the validation procedure was always less than 1%. The pulse height defect, related to the energy loss in the dead layer of the detector, has also a negligible effect on the analysis for the proton energy range studied and the same applies to the ADC width.

The important uncertainty factors in the present work are thus related to the counting statistics and the accurate determination of the $Q\Omega$ factor. The effect of these parameters is strongly target dependent. Whenever the target consists of a compound with a high-Z element, on which the elastic cross section does not deviate from the Rutherford formula, the uncertainty in the determination of the $Q\Omega$ factor is minimized, while the corresponding uncertainty in the statistics is maximized, because one has to subtract the large Rutherford signal of the high-Z element from the total experimental one, in order to validate the cross section for the light element. In the present work, in order to minimize the

effect of the $Q\Omega$ factor uncertainty at high proton energies, where possible deviations from the Rutherford formula could in principle exist for Ga and Br, all data were normalized relative to the Rutherford backscattering on Au. The thickness of the deposited thin Au layers was calculated for each target, by fitting the simulated spectra at several low beam energies (namely at 1531, 1782 and 2033 keV) and detector angles, where proton elastic backscattering on Br, Mo and Ga follows the Rutherford formula and by taking the average value. This procedure yielded an estimated uncertainty of ~3% in all cases, and this was in fact the dominant uncertainty in the validation procedure, except for the case of the NaBr target, which is described in the following section. On the other hand, at lower proton beam energies the uncertainty in the determination of the $Q\Omega$ factor was minimized, since it was obtained directly from the Br, Mo and Ga signal, following the roughness correction and the dominant uncertainty was thus the statistical error in the experimental yield. In all cases, however, with the exception of NaBr, the total combined uncertainty in the present work, including all statistical errors, did not exceed 4%.

5. Results and Discussion

For the $^{nat}\text{S}(p,p)$ backscattering evaluated cross-section data exist up to 3500 keV [2]. The most representative benchmarking results of the present work concerning these data are shown in Figures 4a-b, where one can see that the simulated spectra, using the evaluated cross sections reproduce the experimental ones in an excellent way (within 5%). This is the case for all the backward angles studied, up to 3287 keV (which was the last benchmark point, where the simulation and the experiment perfectly agree), thus also validating cross sections for all intermediate backward detection angles, typically used for EBS measurements. In Figure 4c, it is seen that using the evaluated results from SigmaCalc 1.6 for the $^{nat}\text{S}(p,p)$ backscattering, there are discrepancies between the experiment and the simulation around 3.5 MeV. Following the benchmarking results of the present work, the evaluation was revised, leading to a very good reproduction of the experimental spectra, as shown in Figure 4c. A comparison between

the cross sections obtained using the previous and the current SigmaCalc version (1.6 and 2.0 respectively) is presented in Figure 4d.

Concerning $^{23}\text{Na}(\text{p},\text{p})$ backscattering, one can see in Figures 5a-b some of the benchmarks of the present study, using the evaluated cross-section data for the simulations and in Figures 6a-c the ones using the data of Caciolli et al. [18] at higher energies, at 150° . The existing evaluation in the case of $^{23}\text{Na}(\text{p},\text{p})$ covers only a limited energy range, up to 1500 keV, and at such low energies, as it is shown in Figures 5a-b, the spectra are dominated by the signal of the heaviest element in the compound target, which is Br. Despite the resulting poor statistics (5% in the worst case), originating from the subtraction of the large Rutherford Br signal from the total experimental one (over the whole integrated region corresponding to 250 keV from the surface), the simulation seems to reproduce the experimental spectra quite well for all the studied angles and the evaluation is thus in principle validated. At higher energies, where the only existing dataset relevant to the detection angles studied in the present work, is the one of Caciolli et al. [18] at 150° , covering the energy range between 2210 and 5200 keV, the Rutherford cross section for $^{\text{nat}}\text{Br}(\text{p},\text{p})$ is reduced and the sodium signal is more pronounced. The simulations using these experimentally determined differential cross sections, as shown in Figures 6a-c, are in excellent agreement with the experimental spectra within the total experimental uncertainty (4%) except for the low energy case (Figure 6a) where the agreement is within 6%. This dataset is thus validated and can be recommended for EBS analytical purposes. Moreover, it can be used for the extension of the evaluation to higher energies. As far as $^{\text{nat}}\text{Br}(\text{p},\text{p})$ is concerned, the results from the present work at 148.8° up to 3.6 MeV showed no significant deviation from the Rutherford formula as reported in [19] at 150° .

The results for the $^{31}\text{P}(\text{p},\text{p})$ backscattering are presented in Figures 7a-c. In Figures 7a-b the agreement between the simulations, using the evaluated data from SigmaCalc 1.6, and the experimental spectra is good (within 7%). Above 1800 keV, following the benchmarking results of the present work, as in the case of $^{\text{nat}}\text{S}(\text{p},\text{p})$, the revised SigmaCalc 2.0 datasets led to a very good reproduction of the experimental spectra, as shown in Figure 7c. A comparison between the two versions of SigmaCalc is

again presented in Figure 7d, showing the resonant structure and the extension of the evaluation up to 2.1 MeV. At higher energies the only existing experimental dataset, related to the detection angles studied in the present work, is the one by Karadzhev et al [20] up to 3500 keV for the $^{31}\text{P}(\text{p},\text{p})$ backscattering at 150° . The simulations using these cross-section data are shown in Figures 8a-b, where it can be seen that there are serious discrepancies between the simulated and measured spectra (actually it seems that the data of Karadzhev et al. are systematically underestimated). This dataset cannot thus be recommended for analytical purposes, despite the fact that there seems to be a clear qualitative agreement. Consequently, it cannot be directly incorporated in the evaluation procedure at higher energies and therefore further experimental studies, in addition to the already existing ones (at different backward angles and/or energy range [21] and [22]), are needed in this case. It is important to note here, that in Figure 7b one can see a small peak around channel #350. This corresponds to protons scattered from ^{12}C , which is always present in the targets (carbon build up during the measurements). The same applies to oxygen and of course it concerns all targets. Therefore, these peaks should always be taken into account if needed (e.g. the $^{12}\text{C}(\text{p},\text{p})$ has already been evaluated and validated over a broad energy range), although in most cases, these small contaminants always appear at the low energy part of the spectra and do not really affect the validating results in the benchmarking procedure.

6. Conclusions

In the present work, the necessary steps for the benchmarking procedure have been thoroughly described. The evaluated data, as well as few existing experimental datasets (in the absence of evaluated ones) for the elastic backscattering of protons on ^{23}Na , ^{31}P and $^{\text{nat}}\text{S}$ in the energy range of 1-3.5 MeV have been examined and validated with an accuracy of 5% in most cases. In the cases of $^{31}\text{P}(\text{p},\text{p})$ and $^{\text{nat}}\text{S}(\text{p},\text{p})$ backscattering, the results from the present work at ~ 3.5 MeV provided the necessary feedback for the successful revision of the evaluated cross-section datasets.

Following the present work, accurate experimental thick target spectra exist to facilitate the extension of the evaluations to higher energies (up to at least 3.5 MeV), thus enhancing the application of all IBA depth profiling techniques. Independent benchmarking measurements in other laboratories or using different targets (compounds) are however needed, in order to create a comprehensive library of model thick target spectra, since the benchmarking procedure is very sensitive to the accurate tuning of many experimental parameters. As it is shown in the present study, benchmarking is an integral experiment, with its own steps and uncertainties, which should be properly documented.

Benchmarking data for proton backscattering on ^{nat}Si , ^{nat}K , ^{nat}Ti , ^{nat}O , ^{nat}Fe , ^{nat}Ca and ^{nat}Cr are currently under study, using the same experimental setup and will be the subject of a future work. Moreover, the validation of practically all (p,p), (d,d) and (α,α) evaluated and experimental differential cross-section datasets for low- and medium-Z stable nuclei is mandatory in order to create a firm basis for EBS analytical work. The same procedure will also facilitate NRA studies, through the validation of the most commonly used (d,p), (d, α) and (p, α) reactions.

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Figure Captions

Figure 1. Accelerator calibration using three narrow resonances, namely the (991.9 ± 0.1) keV one of the $^{27}\text{Al}(p,\gamma)^{28}\text{Si}$ reaction ($\Gamma=110$ eV), the (1747.6 ± 0.9) keV of the $^{13}\text{C}(p,\gamma)^{14}\text{N}$ reaction ($\Gamma=122$ eV) and the (3379 ± 1) keV of the $^{32}\text{S}(p,p\gamma)^{32}\text{S}$ reaction ($\Gamma=700$ eV) using a 10% HPGe detector. The error bars are not visible due to the adopted scale.

Figure 2. Thick target spectrum (MoS_2) simulated with and without plural scattering at 1531 keV at 148.8° , along with the corresponding experimental spectrum.

Figure 3a-b. Typical experimental and simulated thick target spectra (MoS_2) at different beam energies, along with the corresponding simulation, *a posteriori* corrected for the effect of surface roughness.

Figure 4a-c: Benchmarking results for $^{\text{nat}}\text{S}(p,p)$ using the corresponding evaluated cross-section datasets for the simulations at different beam energies and detection angles. **d:** Comparison between the two versions of SigmaCalc.

Figure 5a-b. Benchmarking results for $^{23}\text{Na}(p,p)$ using the corresponding evaluated cross-section datasets for the simulations at different beam energies and detection angles .

Figure 6a-c. Benchmarking results for $^{23}\text{Na}(p,p)$ at 150° , using the experimentally determined cross-section dataset by Caciolli et al. [18] for the simulations at different beam energies.

Figure 7a-c. Benchmarking results for $^{31}\text{P}(p,p)$ using the corresponding evaluated cross-section datasets for the simulations at different beam energies and detection angles. **d:** Comparison between the two versions of SigmaCalc.

Figure 8a-b. Benchmarking results for $^{31}\text{P}(p,p)$ at 150° , using the experimentally determined cross-section dataset by Karadzhev et al. [20] for the simulations at different beam energies.

Figure

Figure 1

Figure 2

Figure 3a

Figure 3b

Figure 4a

Figure 4b

Figure 4c

Figure 4d

Figure 5a

Figure 5b

Figure 6a

Figure 6b

Figure 6c

Figure 7a

Figure 7b

Figure 7c

Figure 7d

Figure 8a

Figure 8b