PIXE Detection Limits for some Aerosol Collection Substrates by Excitation with protons and 4He2+ Ions from a 3 MV Tandem Accelerator

Swietlicki, Erik; Bohgard, Mats

Published in:
Nuclear Instruments & Methods in Physics Research. Section B: Beam Interactions with Materials and Atoms

DOI:
10.1016/0168-583X(84)90414-2

1984

Nuclear Instruments and Methods in Physics Research B (1984) 441–445
North-Holland, Amsterdam

PIXE DETECTION LIMITS FOR SOME AEROSOL COLLECTION SUBSTRATES BY EXCITATION WITH PROTONS AND $^4He^{2+}$ IONS FROM A 3 MV TANDEM ACCELERATOR

Erik SWIETLICKI and Mats BOHGARD

Department of Nuclear Physics, Lund Institute of Technology, Sölvegatan 14, S-223 62 Lund, Sweden

Comparisons of PIXE detection limits for K_α X rays using 2–5 MeV protons and 7–8 MeV $^4He^{2+}$ ions as projectiles have been performed. The comparisons have been made for common aerosol backings. According to simple theoretical considerations regarding X-ray production cross sections and the production of background radiation, detection limits for $^4He^{2+}$ should be two to four times lower than for protons for equal velocity projectiles of equal numbers. However, the background in X-ray spectra arising from γ quanta being Compton scattered in the Si(Li) detector can strongly affect the detection limits. The detection limits using protons and $^4He^{2+}$ ions from a 3 MV electrostatic tandem accelerator are determined and discussed.

1. Introduction

Through the years PIXE has developed many different approaches to the problem of obtaining low detection limits for a specific sample of interest. This paper deals with the suitability of using $^4He^{2+}$ as a projectile instead of the more commonly used proton for production of K_α radiation. It is not possible to analyze the lightest elements with PIXE, a drawback that can be overcome in various ways. In PIGE the induced γ rays may be detected simultaneously with the X rays, and particle elastic scattering analysis (PESA) uses the elastically scattered projectiles to determine the amounts of the low − Z elements. As the mass resolution of PESA for very thin samples increases with the mass of the projectile, a combined measurement of both X rays and elastically scattered He^{2+} projectiles could be a better alternative than using protons as projectiles for simultaneous analysis of most elements. The γ production yield is usually higher when $^4He^{2+}$ is used as the projectile thus making $^4He^{2+}$ ions more advantageous than protons for PIGE [1–3].

The minimum detection limits are in this paper defined as $3(N_b)^{1/2}/S$, where N_b is the number of pulses within an interval of two full widths at half maximum around the K_α peak. If $N_b < 11$, then $3(N_b)^{1/2}$ is set to 10. S is the sensitivity, or the K_α yield, measured in: number of detected K_α counts/mass per sample area. The sensitivity S decreases rapidly with increasing Z due to the fact that the production cross section for K_α radiation σ_p is a steadily declining function of Z in the relevant Z interval. According to BEA calculation [4], the production cross section using $^4He^{2+}$ as the projectile is four times higher than for protons, for equal velocity projectiles. Thus, S becomes four times larger, hinting of a possibility of improved detection limits. Another approach is to use a higher projectile energy, as σ_p increases with increasing energy of the projectile for the energies in question. Doing this one must keep in mind the effect of these parameters on the counterbalancing factor N_b.

The background radiation comes mainly from secondary electron bremsstrahlung, projectile bremsstrahlung and Compton-scattered γ quanta. The secondary electrons give the main contribution to the background up to the energy $T_m = 4m_eE/(M_{proj})$, where m_e and M_{proj} refer to the electron and projectile mass, respectively, and E is the projectile energy. This usually corresponds to energies of characteristic K_α radiation for elements in the region $Z = 30$. The γ quanta that are Compton scattered in the Si(Li) detector give rise to a very slowly decreasing background level above T_m, and determine the background level at these energies. The projectile bremsstrahlung is in a first approximation [5] proportional to $(Z_t/A_t\text{-}Z_{proj}/A_{proj})^2$, suffixes t and proj referring to the target and the projectile, respectively. As most elements have a Z/A ratio of around $\frac{1}{2}$, this term should be considerably lower when using $^4He^{2+}$ as projectiles instead of protons. The design of the experimental chamber is also of importance when trying to reduce the background radiation.

A complete theoretical evaluation of the detection limits using different projectiles at different energies on many different backings would indeed be a formidable task. It is necessary to perform experiments on which to base further discussion on the subject.

2. Experiment and results

All experiments were made at the 3 MV tandem accelerator laboratory (NEC 3 UDH) with adjoining

0168-583X/84/$03.00 © Elsevier Science Publishers B.V.
(North-Holland Physics Publishing Division)

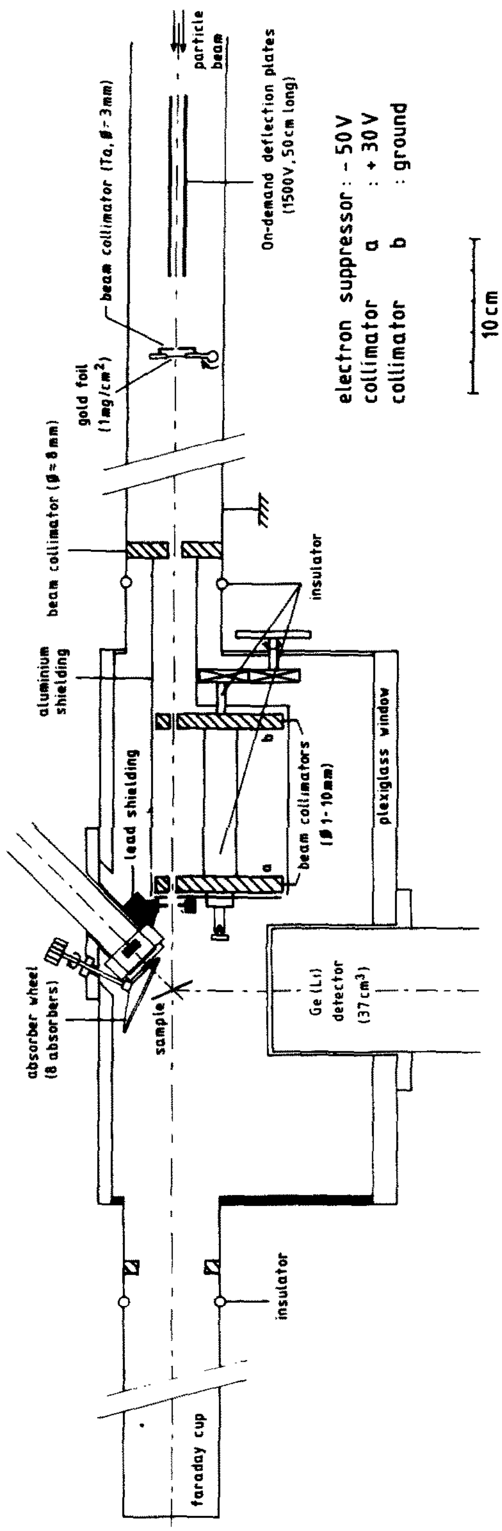

Fig. 1. The experimental chamber. The 6.5 mm Ni collimator was covering the 8 mm carbon collimator at a distance of 29 cm before the target.

PIXE set-up at the Lund Institute of Technology, Sweden. The set-up is described in detail by Malmqvist et al. [6]. The X rays produced are detected in an 80 mm^2 (collimated to an effective area of 28 mm^2) Si(Li) Kevex detector with an energy resolution of 158 eV at 5.9 keV. The detection was made at an angle of 135° to the projectile beam, through a 109 μm Be chamber window, a 4 mm air gap and a 25 μm Be detector window, at a total distance of 4 cm from the sample. Simultaneously, the γ radiation was detected at a 90° angle using a Ge(Li) detector. This was done to make it easier to evaluate the effect of the γ radiation on the X-ray spectra. The charge was measured with an Ortec Model 439 Current Digitizer.

Four different non-exposed backings were bombarded: polystyrene, Kimfol, Nuclepore and Teflon. The polystyrene film was made in our laboratory and has an estimated thickness of 0.03–0.04 mg/cm^2. This very thin and delicate foil is well suited for different chemical analyses demanding high purity backings. Kimberley-Clark's Kimfol backing is about 0.2 mg/cm^2 thick and is a polycarbonate. Nuclepore (also a polycarbonate) is approximately 1 mg/cm^2 thick and often used as an aerosol collection substrate. The last backing, Millipore's commonly used Teflon filter Fluoropore FHUP is about 2 mg/cm^2 thick. The difficulty with Teflon is that it is polytetrafluoroethylene, containing fluorine as a constituent of the foil. This gives rise to many γ transitions from fluorine, causing a distinct increase of the background level in the X-ray spectra.

As the Lund PIXE group regularly uses 2.55 MeV protons for their analyses, the design of the experimental chamber has been adapted to this energy with regard to background yield. When running the proton experiments no changes were made in the chamber, but when $^4He^{2+}$ was used as projectile a Ni collimator (6.5 mm diameter) was placed 29 cm before the target, covering the 8 mm diameter carbon collimator and defining the beam (fig. 1). The choice of Ni as collimator material was based on the work by Giles and Peisach [3] indicating the absence of (α, $\alpha'\gamma$) reactions when using 5 MeV He^{2+} as projectiles. The revolving set of carbon collimators situated between the target and the Ni collimator was then turned to its maximum diameter collimator (13 mm diameter), thus preventing the beam from hitting any carbon directly. By doing this the otherwise very intense 4.4 MeV γ rays from the $^{12}C(\alpha, \alpha'\gamma)^{12}C$ reaction completely disappeared in the γ spectra.

The ionization cross sections used in the calculations of the detection limits were those derived from Akselsson/Johansson's fifth-degree polynomial [7]. According to BEA theory the cross sections scale as the square of the ratio between the charge of the different projectiles, for equal velocity projectiles. It means that, comparing protons with $^4He^{2+}$, the ionization cross sections are four times higher using $^4He^{2+}$ with four times the

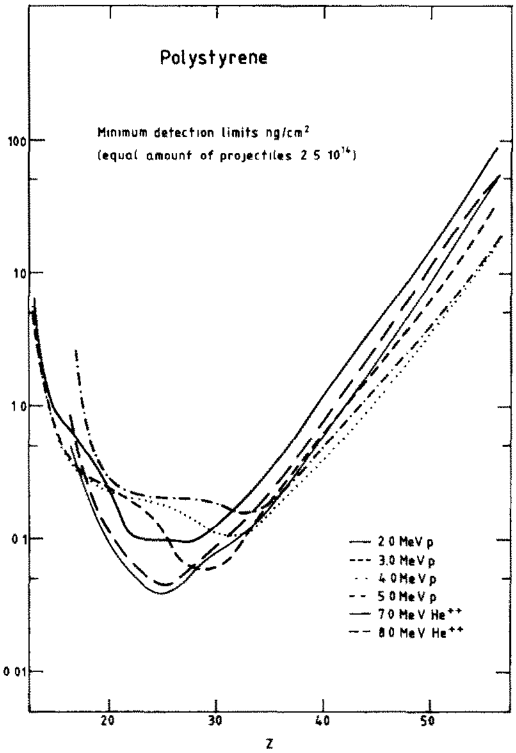

Fig. 2. Minimum detection limits in: ng per cm^2 sample area/2.5×10^{14} projectiles, for 2, 3, 4 and 5 MeV protons and 7 and 8 MeV $^4He^{2+}$ on polystyrene.

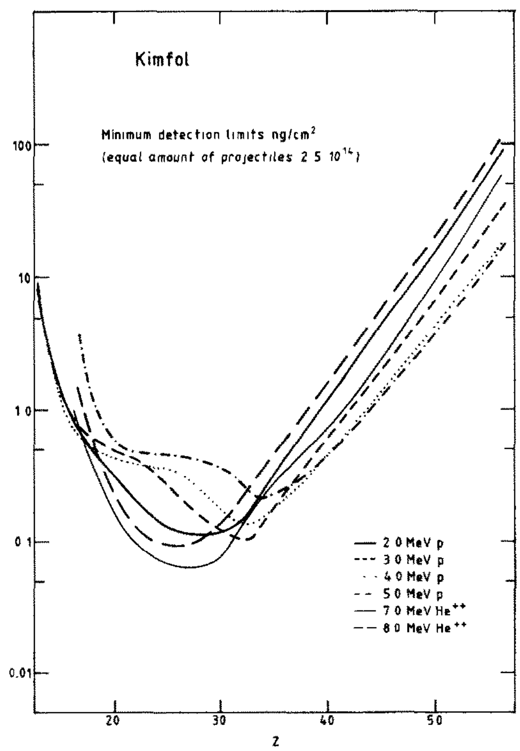

Fig. 3. Minimum detection limits in: ng per cm^2 sample area/2.5×10^{14} projectiles, for 2, 3, 4 and 5 MeV protons and 7 and 8 MeV $^4He^{2+}$ on Kimfol.

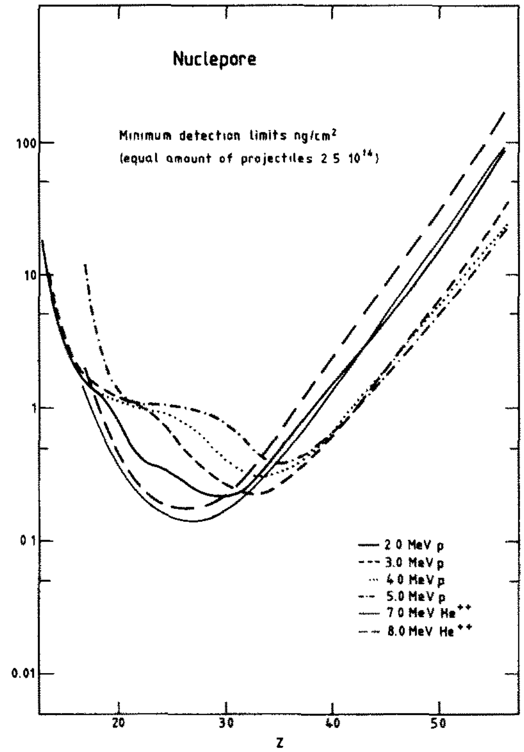

Fig. 4. Minimum detection limits in: ng per cm^2 sample area/2.5×10^{14} projectiles, for 2, 3, 4 and 5 MeV protons and 7 and 8 MeV $^4He^{2+}$ on Nuclepore.

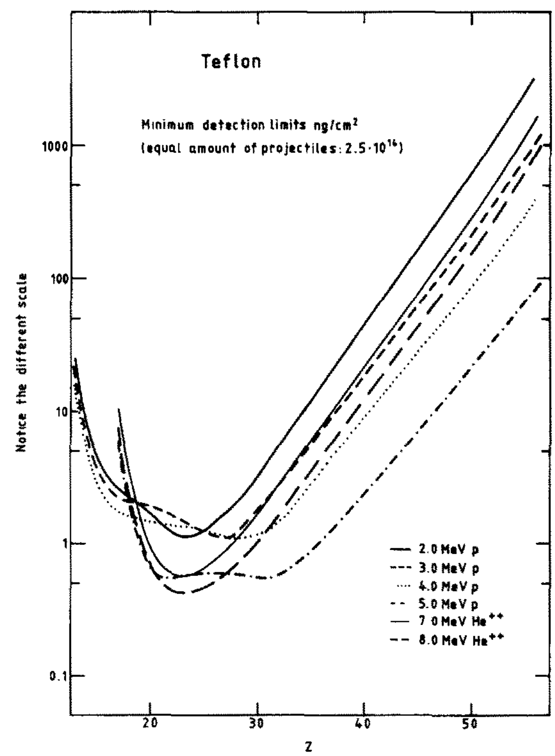

Fig. 5. Minimum detection limits in: ng per cm^2 sample area/2.5×10^{14} projectiles, for 2, 3, 4 and 5 MeV protons and 7 and 8 MeV $^4He^{2+}$ on Teflon. Note the different scale.

kinetic energy of the proton. This was used when estimating the ionization cross sections for $^4He^{2+}$, as the fifth-degree polynomial was calculated from proton data only. Deviations from the assumed scaling will cause a systematic error in the detection limits for $^4He^{2+}$ given here.

All the detection limits are given in the units: ng per cm^2 sample area/2.5×10^{14} projectiles, using no external absorber. For the 5 MeV proton run and both $^4He^{2+}$ runs we actually used a 340 μm Mylar absorber, but the detection limits were afterwards corrected to zero absorber. The number of 2.5×10^{14} projectiles represents a collected charge of 40.0 μC for protons and 80.0 μC for $^4He^{2+}$. The detection limits are valid for low-loaded backings. The detection limits for an equal amount of charge instead of projectiles (e.g. 40.0 μC) will be obtained by multiplying the $^4He^{2+}$ values by the square root of 2, for $N_b > 11$. If the limiting factor is the beam current causing an excessive heating of the sample, a normalization to the energy deposited in the sample should be more appropriate. Bethe–Bloch's formula for the stopping power shows that the energy deposition in the backing is approximately four times higher for $^4He^{2+}$ than for protons for projectiles of the same velocity. The effect on the detection limits is a factor of 2 to the advantage of the protons.

The amount of charge being allowed to hit the target was determined by pulse statistics and time considerations. This meant a charge of 100 μC for the protons and 25–70 μC for $^4He^{2+}$, except for Teflon which gives fully adequate pulse statistics after as little as 5 μC. The maximum uncertainty in the pulse statistics is 30%. This value comes from the fact that if $N_b < 11$, the value of $3(N_b)^{1/2}$ is set to 10, which is sometimes the case for $Z > 30$. Another contribution to the statistical error arises when the number of pulses in the background below two full widths at half maximum (fwhm) is determined. This contribution to the uncertainty of the determination of the detection limits is less than 20%. We assume that the fwhm is proportional to $(\alpha + \beta E)^{1/2}$, E indicating the K_α energy. α and β are constants that are determined experimentally.

Minimum detection limits have thus been calculated for 2, 3, 4 and 5 MeV protons and 7 and 8 MeV $^4He^{2+}$ ions on four backings; polystyrene, Kimfol, Nuclepore and Teflon. This gives us 24 curves presented in four diagrams, one for each backing (figs. 2–5). Notice the different scale in the Teflon diagram. The curves stretch from Cl to Ba ($Z = 17$–56), and in the case of 2, 3 and 4 MeV p all the way from Al ($Z = 13$).

7 MeV $^4He^{2+}$-ions give the lowest detection limits in the region $Z = 18$–31 (except for Teflon) of all examined projectiles and energies. For $Z > 31$ protons give lower detection limits, with 4 and 5 MeV protons being superior for $Z > 35$. When using the high energy protons, this shows us that the higher background continuum in this region is not enough to make up for the increase in production cross sections. Below T_m this advantage is lost for 4 and 5 MeV protons, making 2 and 3 MeV protons more suitable. The minimum is

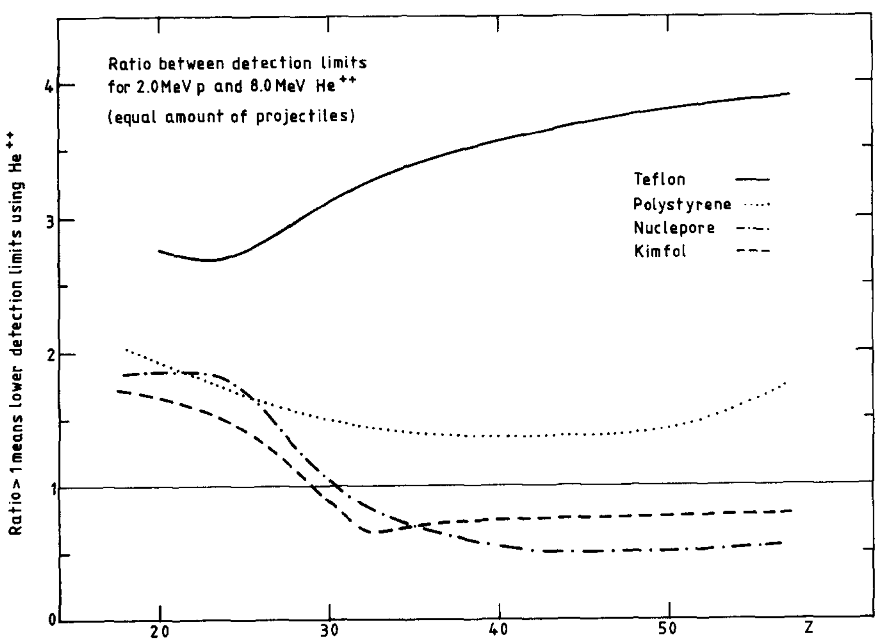

Fig. 6. Ratio between the detection limits for 2 MeV protons and 8 MeV $^4He^{2+}$ (equal velocity projectiles) for equal numbers of projectiles.

shifted towards higher Z using higher proton energies due to the direct effect of the projectile energy on T_m. For $^4He^{2+}$ at only two different energies this is not noticeable.

3. Discussion and conclusions

A closer study of the curves gives an impression that the optimum proton energy should lie somewhere around 3 MeV for K_α radiation, giving us low detection limits both in the low- and high-Z region. The $^4He^{2+}$ energy of preference would be 7 MeV or lower, as the detection limits at this energy are lower than for 8 MeV $^4He^{2+}$ (except for Teflon). The choice of energy is to some extent dependent on the purpose of the analysis. Studying aerosol samples one is often interested in low-Z elements such as sulphur, motivating a somewhat lower energy. When looking at pollutants in an industrial environment high-Z elements like cadmium are often looked for, suggesting a higher optimum projectile energy. When establishing an experimental setup for extensive routine PIXE analyses a compromise must be made when choosing the energy and projectile.

Fig. 6 shows the ratio between the detection limits for 2 MeV protons and 8 MeV $^4He^{2+}$ ions for an equal number of projectiles. A value > 1 means that the detection limits are greater using protons instead of $^4He^{2+}$ ions. 2 MeV protons and 8 MeV $^4He^{2+}$ ions have the same velocity, and the production cross sections for $^4He^{2+}$ should therefore be four times larger than those of protons according to the BEA scaling rule. Knowing that the definition of the detection limits is $3(N_b)^{1/2}/S$, it can be assumed as a first rough estimate that the ratio between the detection limits of 2 MeV protons and 8 MeV $^4He^{2+}$ ions should be about 2 below T_m and 4 for high Z. As can be seen in the figure this is hardly the case. For Kimfol and Nuclepore the ratio is even below 1 above Cu ($Z = 29$). This could be explained by the increased γ radiation emanating mainly from the sample itself, which is then Compton scattered in the Si(Li) detector. Notice that the value 2 for the ratio below T_m agrees better with the data in fig. 6, probably because here the Compton background is not the determining factor.

7 MeV $^4He^{2+}$ runs with carbon collimators instead of the Ni collimator indicate that the detection limits are increased by some 10% due to the intense 4.4 MeV γ radiation from the carbon. The question arises whether the experimental chamber could be further improved. Scattering of projectiles by the target, subsequently causing the projectiles to hit the chamber walls, may give rise to an increased X-ray background that perhaps could be avoided. A study of this will be the object of further investigations.

In conclusion, the detection limits using 7 and 8 MeV $^4He^{2+}$ ions as projectiles are of the same order and in many cases ($Z = 18$–31) lower than those using 2, 3, 4 and 5 MeV protons for equal numbers of projectiles. The choice of projectile and projectile energy must be a compromise between the low- and high-Z detection limits. Complementary methods such as PIGE and PESA for the detection of low-Z elements should also be taken into consideration. The design of the experimental chamber should be made with some care when using $^4He^{2+}$ as projectile, in order to reduce the high intensity Compton scattering of γ radiation in the Si(Li) detector, causing an increased background in the X-ray spectra.

References

[1] A. Anttila, R. Hänninen and J. Räisänen, J. Radioanal. Chem. 62 (1981) 293.

[2] G. Demortier, J. Radioanal. Chem. 45 (1978) 459.

[3] I.S. Giles and M. Peisach, J. Radioanal. Chem. 50 (1979) 307.

[4] J.D. Garcia, R.J. Fortner and T.M. Kavanagh, Rev. Mod. Phys. 45 (1973) 111.

[5] F. Folkmann, C. Gaarde, T. Huus and K. Kemp, Nucl. Instr. and Meth. 116 (1974) 487.

[6] K.G. Malmqvist, G.I. Johansson and K.R. Akselsson, J. Radioanal. Chem. 74 (1982) 125.

[7] R. Akselsson and T.B. Johansson, Z. Phys. 266 (1974) 245.