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On the Application of Elastic Scattering Analysis as a Complement to PIXE for the Determination of Light Elements in Thin Aerosol Samples

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ABSTRACT

The potential of elemental analysis of light elements by measuring elastically scattered particles as a complement to PIXE (Particle Induced X-ray Emission analysis), when using a 3 MV electrostatic tandem accelerator, is discussed. The discussion is based on protons and He ions with energies suitable for PIXE. Some experimental results of scattering yields are included.

INTRODUCTION

Since its introduction as an analytical method Particle Induced X-ray Emission (PIXE) (1) has been used in several studies of aerosols. The multielemental nature of the method and its capacity to give fast and reliable analysis of high sensitivity is ideal for determining low amounts of elements in large quantities of small aerosol samples.

The lightest elements cannot be analysed with PIXE due to the low energies of their characteristic X-rays. In a transition region ($12 \leq Z \leq 16$) the possibility of performing quantitative analysis is highly dependent on the sample homogeneity and the ability to determine the X-ray absorption in the sample and between the sample and the sensitive volume of the detector.

In contrast to X-ray fluorescence (XRF) PIXE offers the possibility of detecting the light elements by measuring signals originating from the nuclei of these elements. Particle Elastic Scattering Analysis (PESA)

and Particle Induced γ-ray Emission (PIGE) can be performed simultaneously with PIXE. In this paper the combined use of PESA and PIXE using protons and He-ions from a 3 MV electrostatic tandem accelerator is discussed.

ELEMENTAL RESOLUTION

By bombarding samples with protons and He ions elemental analysis can be performed by detecting particles elastically scattered. Proton energies below 1 MeV and He-ions below 2 MeV are often used. These energies are lower than the Coulomb barrier for most nuclei. Thus scattering yields can often be predicted in these cases by using the classical Rutherford formula for the differential cross-section of scattering from a point charge (2). The method is usually called Rutherford Back-Scattering Analysis (RBS) and is well established (3). By measuring the energies of the scattered particles the masses of the target nuclei can be determined using the following formula (4)

$$E_M = E_0 \left\{ \frac{m\cos\theta + \sqrt{M^2 - m^2\sin^2\theta}}{m + M} \right\}^2$$

θ = scattering angle in the laboratory system
m = mass of the projectile particle
M = mass of the target nucleus
E_0 = energy of the incident particle
E_M = energy of particle scattered by a nucleus with mass M

The elemental resolution is highest for light elements. The simultaneous detection of scattered particles with a surface barrier detector and X-rays with a Si(Li) detector is an attractive combination. However, for thin PIXE-samples (e.g. aerosol samples) the projectile energies used for traditional RBS-analyses are not sufficiently high for elemental resolution of the light elements which can not be analysed with PIXE. Cahill et al. (5) have detected forward scattered He ions with an incident energy of 30 MeV for the determination of elements from hydrogen to fluorine in aerosol samples. Nelson and Courtney (6) have analysed light elements in

aerosol samples by detecting protons, with an incident energy of 16 MeV, scattered 135°. For small accelerators the maximum energy available limits the thickness of the samples which can be analysed. Determinations of surface concentrations can, however, be performed without completely separated signals from the elements. Kemp (7) has used 3 MeV protons for the analysis of C, N and O in aerosol samples by comparing the concentrations of the elements in the samples with the concentrations in the aerosol filter.

To facilitate the discussion of the potential of elastic scattering analysis two scattering geometries are given in fig 1. In the arrangement shown in fig 1 a particles scattered 155° are detected with a circular surface barrier detector. This arrangement is used in the present PIXE facility in Lund (8). Figure 1 b shows an arrangement with an annular detector with a 174° scattering angle which is intended to be used in a future construction.

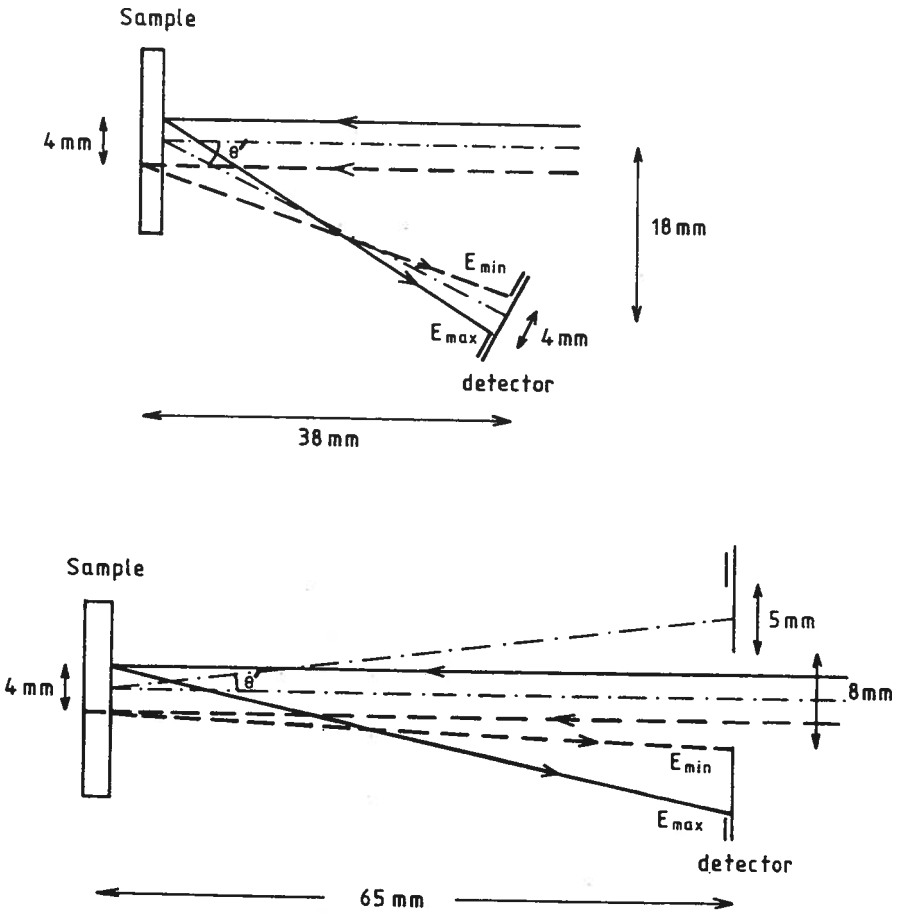


Figure 1.

Two geometrical arrangements, a) with a circular detector and b) with an annular detector, suitable for elastic scattering analysis. The solid angles are 7.2 and 48 msr in a) and b) respectively. Scattering angle = $= 180° - \theta$.

The annular detector covers a much larger solid angle at equal differences between maximum and minimum scattering angle ($\Delta\theta$) than the circular detector. The annular detector also detects particles in a more backward direction, making the path in the sample of the scattered particle shorter, which reduces the energy loss. With increasing scattering angle $dk/d\theta$ ($k = E_M/E_0$) decreases which gives better mass resolution for a certain range of possible scattering angles of the particles detected. The increase in dk/dM with scattering angle also contributes to a better mass resolution for more backward directions.

Figures 2a-d illustrate the resolving power for light elements at the given scattering geometries and different particle energies. In the calculations stopping power data from Andersen and Ziegler (9) were used and $\Delta\theta$ is given as the difference beteween the maximum and minimum scattering angle according to figure 1. For 2.55 MeV protons as projectiles (figure 2a) it is obvious that all light nuclides cannot be separated. However, these projectiles may be used for special applications when samples contain a few light elements are analysed, or for the determination of beryllium, which is a toxic metal of great hygienic interest and abundant in some working environments. The sensitivity for beryllium for 2.55 MeV protons is comparatively high (table 1). For 5 MeV protons (figure 2b) the mass resolution is improved and all elements with masses below 20 u are separated for a matrix with particle stopping corresponding to that in 200 $\mu g/cm^2$ carbon.

When using He-ions the increased value of dk/dM gives a substantially increased energy difference between the elements. However this advantage is limited to very thin samples. Due to the large particle stopping the mass resolution rapidly decreases when sample thickness increases. Figure 2 d illustrates the mass resolution for He ions of 9 MeV, which is the highest energy attainable with a 3 MeV tandem accelerator. For an energy loss corresponding to 200 $\mu g/cm^2$ carbon, ^{11}B and ^{12}C cannot be completely separated.

In common aerosol applications (e.g. working environments and atmospheric aerosols) the boron concentration is far lower than the carbon concentration, therefore the practical target thickness limit

is often determined by the separation between ^{14}N and ^{16}O for nuclides with mass numbers below 20. The first nuclide of interest above 20 u is ^{23}Na. To separate the signal from ^{23}Na from that of ^{24}Mg, extremely thin samples are required.

For projectiles obtainable from a 3 MV tandem accelerator the highest particle energies can give sufficient elemental resolution for masses below 20 u, at least for samples of fine particles (diameter below 2 μm).

ANALYTICAL SENSITIVITY

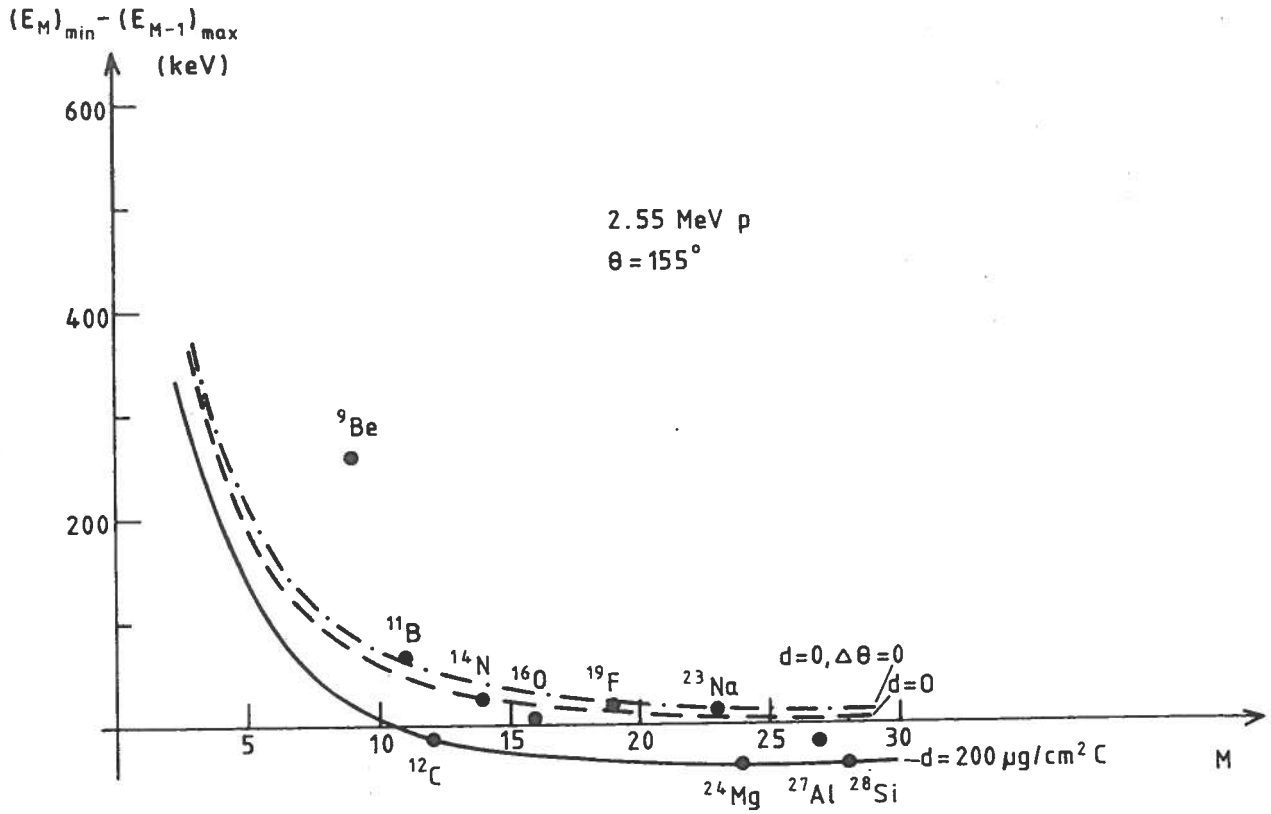
The analytical sensitivity cannot generally be predicted from existing theoretical models. However, sensitivities may sometimes be estimated from published cross section data. The differential elastic scattering cross sections of light nuclei are typically in the 1-200 mb/sr range for 2-6 MeV protons and 3-9 MeV He ions, giving analytical sensitivities between 1 and 100 pulses/(($\mu g/cm^2$)·msr·μC). The sensitivities obtained from published data cannot normally be used for determining amounts in elastic scattering analyses. The scattering parameters used in these measurements seldom coincide with the optimal ones for analytical purposes. A useful compilation of experimental cross section data for elastic scattering and nuclear rections has been performed by Jarjis (10).

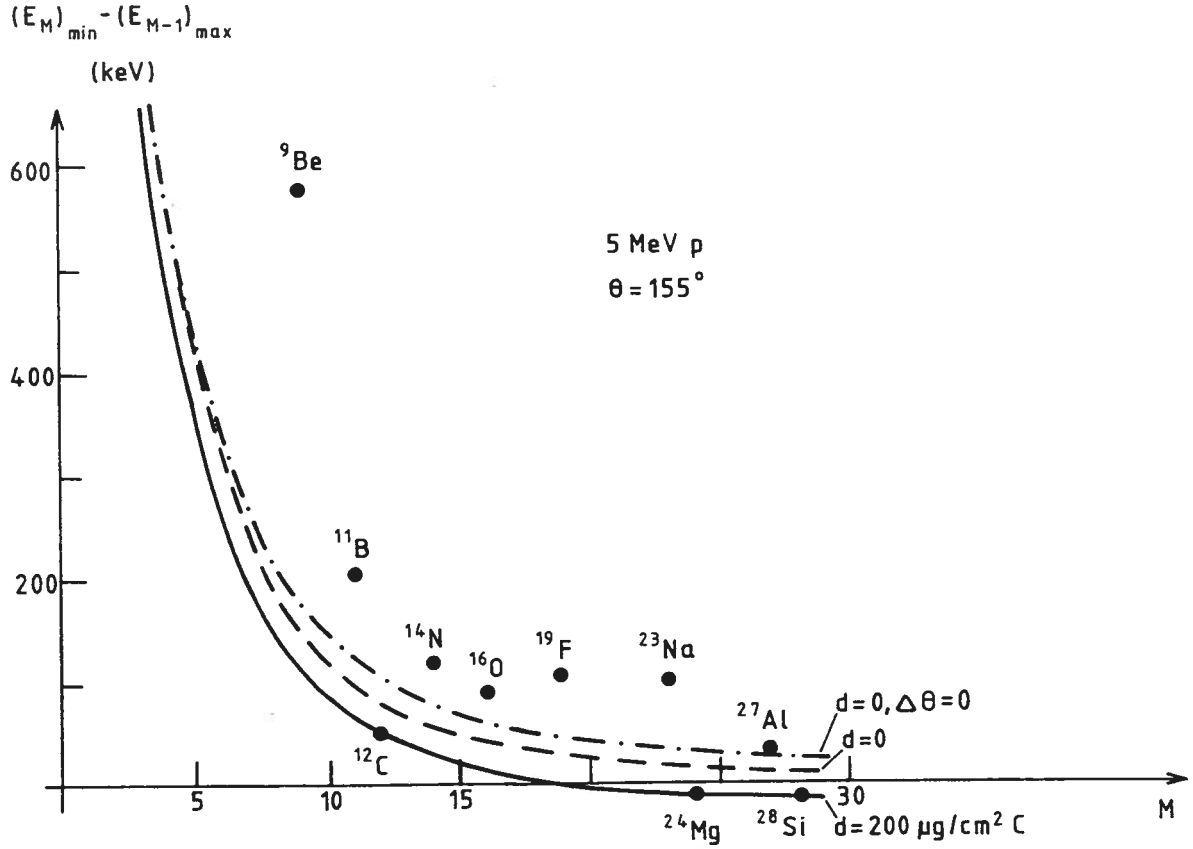
EXPERIMENTS AND RESULTS

Estimation of sensitivities

For the estimation of analytical sensitivities thick targets can be bombarded with particles of various energies. For calibration purposes thin foils of various thicknesses have to be used. Analytical sensitivities have been estimated for two proton energies and the scattering geometry shown in fig 2a.

Protons were accelerated with an electrostatic tandem accelerator (NEC 3 UDH) to 2.55 and 5.00 MeV respectively and the scattered particles


$(E_M)_{min} - (E_{M-1})_{max}$
(keV)
600
400
200
2.55 MeV p
θ = 155°
^{9}Be
^{11}B
^{14}N
^{16}O
^{19}F
^{23}Na
d=0, Δθ=0
d=0
5
10
15
20
25
30
^{12}C
^{24}Mg
^{27}Al
^{28}Si
d = 200 μg/cm² C
M


$(E_M)_{min} - (E_{M-1})_{max}$
(keV)
600
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5 MeV p
θ = 155°
^{9}Be
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^{16}O
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^{12}C
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^{28}Si
d = 200 μg/cm² C
M

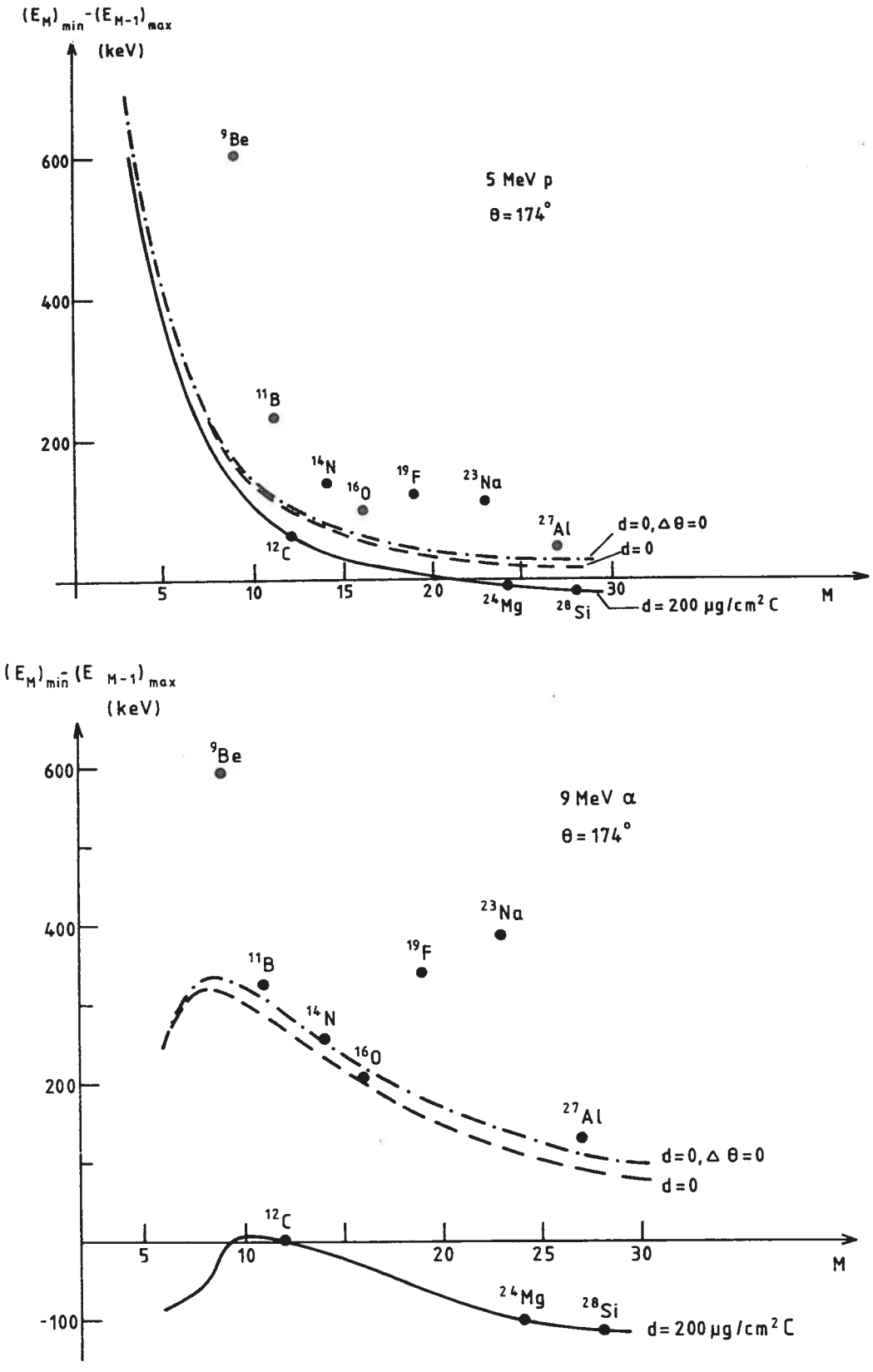

Figure 2

Illustrations of elemental resolving powers for scattering geometries shown in figur 1. and a sample thickness corresponding to 200 μg/cm² carbon. The solid line shows the difference between minimum and maximum energies for consecutive mass numbers. For comparison the resolution is also illustrated by curves for d (thickness) = 0 and Δθ (maximum difference in scattering angle) = 0. The dots show the differences between minimum and maximum energies for the major isotopes of consecutive elements (excluding neon) calculated for a sample thickness of 200 μg/cm². To obtain completely separated peaks $(E_M)_{min} - (E_{M-1})_{max}$ should be greater than 20-40 keV due to detector resolution and energy straggling.

were detected (scattering angle = $155^{\circ} \pm 2^{\circ}$, solid angle = 1.8 ±0.1 msr) with a partially depleted surface barrier detector (ORTEC TA-014-25-300). The experimental set-up for PIXE analysis in Lund described by Malmqvist et al. (8) was used. Pellets (made of LiCl, Li_2CO_3, H_3BO_4, NH_4Cl, $MgSO_4$ and $Na_3C_6H_5O_7$), metallic foils (Be and Al) and SiO_2 were used as targets.

The sensitivities were calculated from the height of the edges in the particle spectra originating from scattering in a surface layer with thickness, δx, corresponding to a particle energy stopping, δE, of one channel width in the spectrum. The following expressions were used (3),

$$A = Q \sigma \Omega N_A t$$

$$H = Q \sigma \Omega N_H \delta x$$

$$\delta x = \frac{\delta E}{(E_M/E_0)(-dE/dx)_{E_0} + (-dE/dx)_{E_M} |1/\cos\theta|}$$

Ω = solid angle (sr)
A = number of pulses in a peak from the element in a "thin target spectrum"
Q = number of projectiles
σ = average differential cross section, (cross section for scattering (cm^2/sr))
N_A = number of atoms per cm^3 in the thin sample
t = sample thickness (cm)
$-dE/dx$ = stopping power (keV/cm)
N_H = number of atoms/cm^3 in the thick sample
H = height of the edge in spectrum from thick samples (pulses)

and the number of pulses for a given amount of an element is calculated from:

$$A/(N_A t) = H/(N_H \delta x)$$

Stopping power data from Andersen and Ziegler (9) were used for

ELEMENT	SENSITIVITY (pulses/((μg/cm^2)·msr·μC))	
	2.55 MeV protons	5 MeV protons
Li	22	31
Be	76	11
B	42	20
C	36	42
N	23	19
O	18	25
F	3.0	3.6
Al	6.9	3.3
Si	6.5	7.6
P	7.4	20

Table 1.

The sensitivities obtained using 2.55 and 5.0 MeV protons (scattering angle = 155°) for light elements The uncertainties are below 15%. The sensitivities for Na and Mg were below 5 pulses/((μg/cm^2)·msr·μC).

calculations of δx. The estimations of the sensitivities are given in Table 1. For carbon, sensitivities were obtained from irradiation of thin polystyrene foils.

Evalutation of spectra from thin samples

The peaks in an elastic scattering spectrum are not gaussian. The continuum on the low-energy side of a peak has only recently been explained. With the present knowledge it is not possible,

in general, to give an analytical expression for the shape of non-Rutherford elastic scattering spectra. This makes it difficult to perform theoretical fits to experimental data points in the spectra.

To perform an evaluation of some simple methods for peak integration thin polystyrene foils spiked with cobalt were prepared (11). The foils were irradiated by 2.55 MeV protons. X-rays and scattered particles were detected simultaneously. The foils were spiked with 2060 ± 70 ppm cobalt and the thicknesses of the foils were calculated from the cobalt determination. Figure 3 shows a schematic one-peak spectrum. The magnitude of the carbon signal was determined in three different ways; a) the counts in the peak (P+B) and the low-energy continuum (T) were summed, b) the total number of counts in the peak (P+B) was found by integration and c) the number of counts in the peak corrected for a linear background was calculated (P).

Figure 4 shows the number of counts as a function of the foil thickness for the three measures of estimating the intensity of the carbon signal. In all three methods the intensity of the carbon peak increased linearly with foil thickness upp to a thickness of 200 $\mu g/cm^2$.

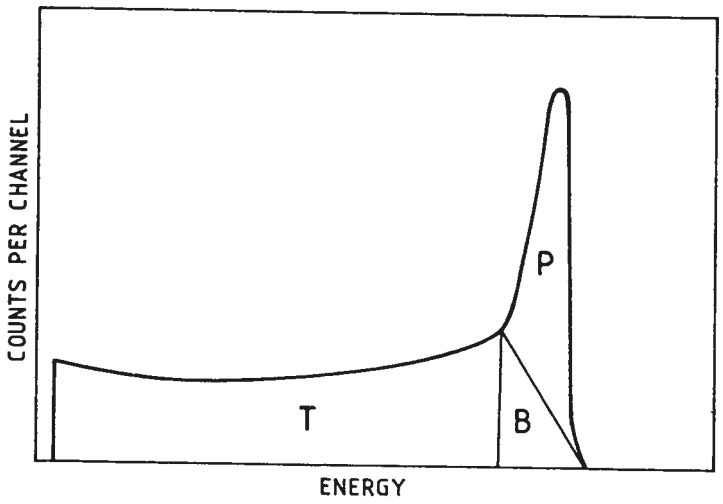

Figure 3.

A schematic one-peak elastic scattering spectrum.

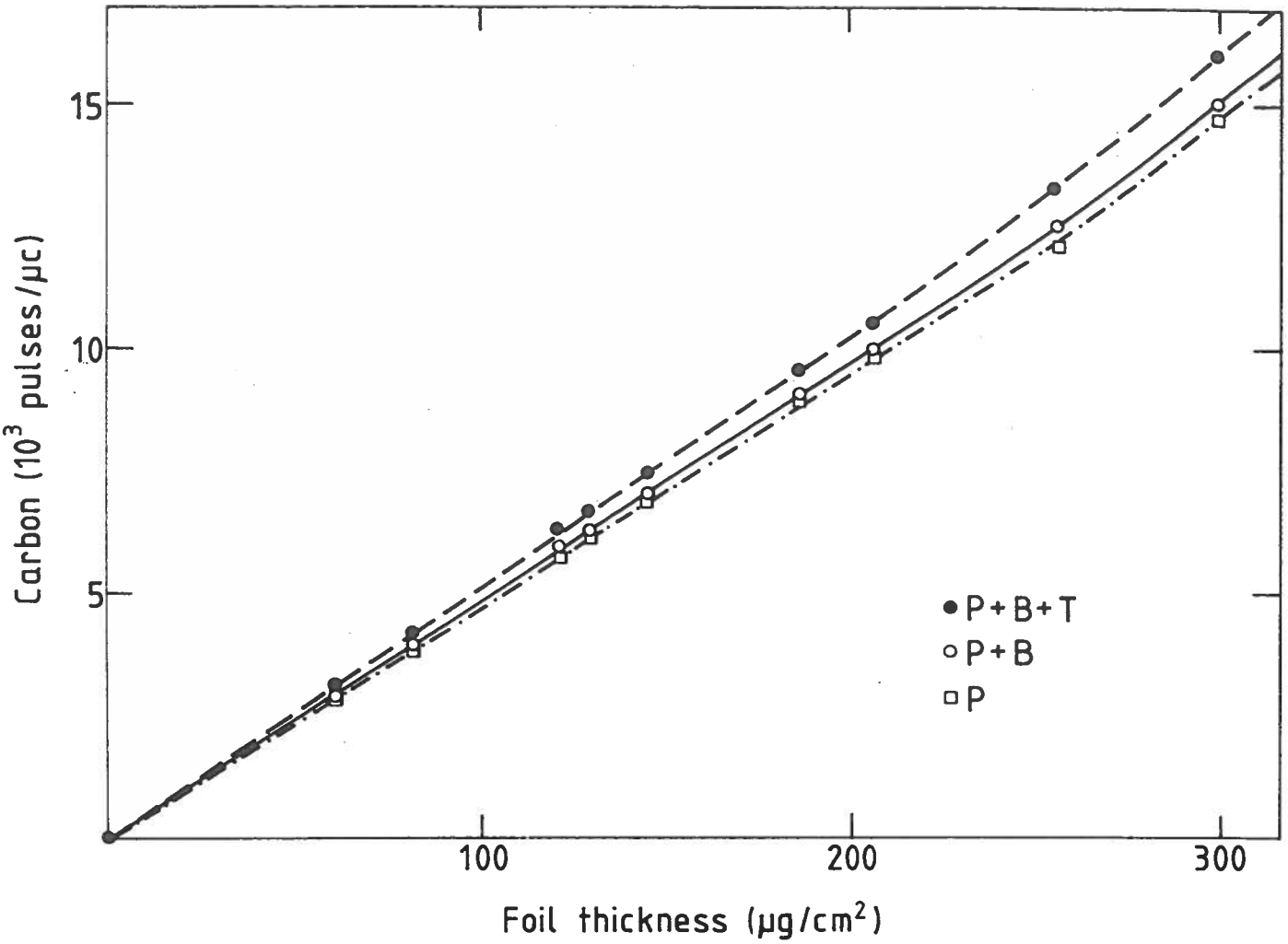

Figure 4.

Number of counts in the carbon peakas a function of sample thickness for polystyrene foils. Three different methods of estimating the number of counts in the carbon peak have been used with notations as given in figure 3.

The deviation from linearity for thicker samples is discussed in a previous paper (11). Any of the three measures can be used for a one-peak spectrum. Summation of the peak, background and tail (P+B+T) is however not applicable for spectra consisting of a number of peaks. The total counts in the peak (P+B) may be the most appropriate when the number of counts in the low energy tail is low. This can give large uncertainties if B is subtracted. For spectra with low-energy tails of a considerable size, the peaks corrected for a linear background can be a better measure because this method is less sensitive to how the width of the peak is defined.

CONCLUSIONS

Protons and He ions from a 3 MV electrostatic accelerator may, to a limited extent, be used for the analysis of the light elements, simultaneously with he PIXE analysis, in aerosol samples. To obtain high elemental resolution there are restrictions on the sample thickness and particle size which can be analysed. Calibration of carbon in a polystyrene matrix shows a linear yield below 200 µg/cm2 for 2.55 MeV protons scattered 155 degrees.

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