Investigation of the $^{19}\text{F}(p, \alpha, e^{-}e^{+})^{16}\text{O}$ reaction for use in ion beam analysis


Department of Nuclear Physics, Lund Institute of Technology, Box 118, SE-221 00 Lund, Sweden

Received 31 October 1996; revised form received 30 January 1997

Abstract

The $^{19}\text{F}(p, \alpha, e^{-}e^{+})^{16}\text{O}$ reaction has been investigated with the aim to employ it for ion beam analysis. An experimental set-up was designed, in which the $\alpha$ particle was detected in coincidence with either the emerging electron or the positron. The reaction was studied in the energy interval between 2.00 to 2.55 MeV. The minimum detection limit for thin samples was found to be 30 ng/cm$^2$ for an integrated beam charge of 10 $\mu$C. An analysis of odontological material was performed with the technique. The possibility to only utilise the electron-positron pair for the analysis of fluorine samples is proposed and discussed.

PACS: 2590; 2930; 2970; 2990
Keywords: Fluorine analysis; Hydrogen analysis; NRA; $^{19}\text{F}(p, \alpha, e^{-}e^{+})^{16}\text{O}$; PIXE; Nuclear microprobe

1. Introduction

The analysis of fluorine has been extensively studied by many authors [1-6]. Especially the depth profiling properties have attracted great interest. Both the $^{19}\text{F}(p, \alpha, \gamma)^{16}\text{O}$ reaction and the $^{19}\text{F}(p, \alpha, \alpha')^{16}\text{O}$ reaction (where $\alpha'$ denotes the various excited states) have been widely used for this purpose. However, the first excited state in $^{16}\text{O}$ (at 6.05 MeV) has not gained large attention for utilisation in ion beam analysis. This is a $0^+ \rightarrow 0^+$ transition, and thus a gamma ray de-excitation is rigorously forbidden. The de-excitation takes place mainly through internal pair production, by which an electron and a positron are created. These share the remaining 5.03 MeV (6.05-1.022 MeV) [7,8]. In this paper, an investigation of this reaction is undertaken, both with the aim of employing it as a technique for depth profiling and for trace element analysis.

There are applications for fluorine analysis in various areas. Both bulk analysis and micro-analysis have been performed, the latter by depth profiling as well as by nuclear microprobes (NMP) [9]. In odontology, problems related to the fluorine distribution in teeth have been investigated [10,11]. In medicine, some drugs contain F and would be interesting to analyse [12]. In geology, various issues involve the analysis of F [13,14]. Dating of archaeological artefacts has been done by a technique based on the depth analysis of fluorine [15,16]. Fluorine analysis has also been performed in material science, for example Si [17], F implantation of diamond [18] and metallurgy [19].

2. The experiments

The experiments were carried out at the Lund Nuclear Microprobe, which is a single-ended NEC 3 UH accelerator. The beam line has been described in Ref. [20]. The proton beam had an energy of between 2.00 and 2.55 MeV and a current between 100 and 500 pA. Broad beam spots were used as well as focused ones. The current was limited by the count rate in the detector (see below).

The basis of the design of the experiment was to measure the $\alpha$ particle in coincidence with either the electron or the positron. The $\alpha$ particles were detected with a silicon surface barrier detector (SBD) and the electrons and positrons with a thin plastic scintillator. The experimental arrangement can be seen in Fig. 1. The $\alpha$ particles from the $^{19}\text{F}(p, \alpha, e^{-}e^{+})^{16}\text{O}$ reaction were detected by an annular silicon surface barrier detector with an active area of 450 mm$^2$ and a hole diameter of 12 mm. Its thickness of 300 $\mu$m stops all $\alpha$ particles and protons, whereas electrons and positrons lose only a slight fraction...
Fig. 1. The experimental set-up. The α particles from the $^{19}$F(p, α, e$^-$ e$^+$)O reaction were detected by the SBD, and the electrons and positrons passed through the SBD and were detected by the plastic scintillator. The SBD was shielded at the back from scattered protons from the beam by an Al tube. The light guide led the light through to a photomultiplier tube (PMT) placed on top of the chamber.

of their energy, which could be discriminated by setting a threshold in the discriminator. The detector was placed in the backward direction at a distance of 9 mm from the target and covered an angular interval between 125° and 145° relative to the beam direction. The proton beam went through an aluminium tube when passing the scintillator and the SBD so that no particles could enter the SBD from behind. The electrons and positrons were detected with a 2 mm-thick plastic scintillator (NE-110). The scintillator was situated behind the SBD at a distance of 25 mm from the target. It was glued onto the Plexiglas light guide with optical glue. The size of the scintillator was 50 X 50 mm$^2$.

It was covered with aluminium foil in order to enhance the light collection. A thin layer of carbon tape was attached outside the aluminium to stop scattered protons from reaching the scintillator. The light guide led the light through the chamber wall to a photo multiplier tube (PMT) placed on top of the chamber. The PMT was a Hamamatsu R1166 which has a circular cathode with a diameter of 19 mm.

The lateral homogeneity and efficiency of the light collection of the scintillator was verified with collimated irradiation by sources of $^{207}$Bi (an internal electron conversion source) and $^{143}$Pm (a β-source).

In Fig. 2 the energy spectra of the plastic scintillator are shown for the case of a thin CaF$_2$ sample and of a thick Teflon target. The signal from the PMT was directly connected to a charge-integrating ADC. The energy interval in which the minimum ionizing electrons lose their energy when passing through the scintillator is marked with a bar. The enhanced ratio of electrons and positrons for a thick sample can be observed. A discriminator threshold is imposed at the indicated level in the coincidence set-up described below.

The signal from the scintillator was used only as a trigger. Due to the low count rate in the scintillator no fast coincidence unit was necessary to reduce the data flux. The signal from the SBD was processed by standard electronics. The energy signal from the SBD was collected when an event above the threshold occurred in the scintillator. Further, the time between an event in the SBD and an event in the scintillator was measured by a time-to-pulse height converter (TPHC). The time and the energy signals were acquired by peak-sensitive ADCs event by event. These ADCs were part of a Camac data acquisition and beam control system [21]. The integrated charge was also recorded.

The background in the scintillator is small and originates mainly from Compton-scattered γ-rays. The background from pair production and Compton scattering taking place in the SBD due to the high energy γ-photon, is estimated to be less than 1% of the coincidence signal from the reaction. Some background arises also from
processes in surrounding material, such as the chamber wall. The total background from the $\alpha_2$ transition, which de-excites through $\gamma$-decay and is separated only by 0.08 MeV from the $\alpha_1$ transition, can be estimated to be about 4%. Very few other reactions giving rise to electrons exist for proton energies lower than 3 MeV. The amount of internal conversion electrons is determined by the internal conversion factor, which for light elements is normally less than $10^{-4}$. For thick samples the enhanced electron flux, due to back-scattering of the electrons and positrons in the sample, must be considered. This effect depends strongly on the Z of the material and the energy of the electrons. Thus calibration must be performed with a standard target with a similar matrix when analysis of thick samples is done.

Three types of samples were irradiated. Nuclepore covered with 50 $\mu$g/cm$^2$ (+5%) CaF$_2$ was used as a thin standard. Teflon, a polymer with the chemical formula (CF$_2$)$_n$, was employed as a thick standard. As examples of possible applications for fluorine analysis, two different odontological materials, utilised as mending material, were investigated. One was a glass ionomere material (containing fluorine), and the other a fluorine composite material. They were covered with a carbon foil, 40 $\mu$g/cm$^2$ thick, to prevent charge build-up in the sample. The issue of interest for this kind of analysis is to obtain information on the concentration and the distribution of fluorine in the development process of new mending materials in odontology.

3. Results

The thin CaF$_2$ sample was used as a standard target. In Fig. 3 the two-dimensional spectrum of the energy of the SBD versus the coincidence time gated with the condition that there be one event in the SBD and one above the threshold in the scintillator is shown. The marked region shows the fluorine events from the reaction under investigation. The region at lower energy is the result of the $^{19}$F(p, p$\gamma$)$^{19}$F reaction, where the $\gamma$-photon of 110 keV has been Compton-scattered in the scintillator and detected in coincidence with the proton. The proton events appear at slightly longer time because of non-linear effects in the leading-edge discriminator. The energy projection of the spectrum in Fig. 3 is shown in Fig. 4.

The analogue of Fig. 3 of an analysis made on the thick Teflon sample is shown in Fig. 5. The events for the reaction are demarcated down to the maximum probing depth, which is in the order of 10 $\mu$m. The maximum probing depth is determined by the depth from which the $\alpha$ particle can escape from the sample, which for a proton
Energy of about 2.5 MeV is approximately 1/4 of the penetration depth of the proton. The projected energy spectrum is disturbed by a background from the $^{19}$F($p$, $p,\gamma$)$^{20}$F reaction, which has to be corrected for. The coincidence time increases with decreasing energy of the $\alpha$ particle, as is to be expected.

3.1. Energy scan

An energy scan for the reaction was made from 2.00 to 2.55 MeV in steps of 50 keV using the CaF$_2$ target. The beam head was about 10 $\mu$m in size and was scanned over the sample over an area $0.5 \times 0.5$ mm$^2$, divided into $64 \times 64$ steps of 8 $\mu$m. The result of the scan is shown in Fig. 6. The yield values are calculated from two-dimensional histograms of the type shown in Fig. 3. The highest value measured is 35 counts/µC/µg/cm$^2$.

3.2. Minimum detection limit

The minimum detection limit (MDL) is often defined as $\text{MDL} = 3\sqrt{N_B}$, where $N_B$ is the number of pulses in the background of the relevant signal. In the case of non-existent background, a significant signal must contain at least 10 counts.

The maximum yield of the set-up is 35 counts/µC/µg/cm$^2$ with a small background. Thus the MDL is 290 ng/cm$^2$ for an integrated charge of 1 µC, a reasonable charge for an NMP. For 10 µC, which is a feasible charge, an MDL of 29 ng/cm$^2$ is achieved. The MDL varies inversely proportionally to the integrated charge.

3.3. Analysis of odontological samples

The odontological materials were scanned over an area of $8 \times 512$ $\mu$m$^2$ in $64 \times 64$ steps of 1/8 and 8 $\mu$m with a
beam spot size of about 3 \mu m. The glass jonomere material showed no structure in the F distribution in the scanned area. In the fluoride composite, some structure, interesting to further study, was found. There was no attempt made to deconvolute the data in order to obtain depth profiles. To calculate the fluorine concentration the Teflon sample was used as a standard. The backscattering effect mentioned in Section 2 was estimated to be negligible, as both Teflon and the odontological samples consist mainly of light elements. For the glass jonomere material, the total F concentration was determined to be about 8\%, and for the fluoride composite material about 4\%.

4. Discussion

The most common method of ion beam analysis to measure fluorine is PIGE (Particle-Induced Gamma-ray Emission). Either the \gamma-energies of 110 and 197 keV from the $^{19}\text{F}(p, \gamma)^{16}\text{O}$ reaction for proton energies between 2.00 and 2.55 MeV. The steps between the points are 50 keV. It shows large variations depending on the beam energy, but the top value is large, 35 counts/\mu C/cm$^2$. The dashes between the points are to guide the eye.

Fig. 6. The excitation curve for the $^{19}\text{F}(p, \alpha\gamma e^- e^+)^{16}\text{O}$ reaction for proton energies between 2.00 and 2.55 MeV. The steps between the points are 50 keV. It shows large variations depending on the beam energy, but the top value is large, 35 counts/\mu C/cm$^2$. The dashes between the points are to guide the eye.

beam spot size of about 3 \mu m. The glass jonomere material showed no structure in the F distribution in the scanned area. In the fluoride composite, some structure, interesting to further study, was found. There was no attempt made to deconvolute the data in order to obtain depth profiles. To calculate the fluorine concentration the Teflon sample was used as a standard. The backscattering effect mentioned in Section 2 was estimated to be negligible, as both Teflon and the odontological samples consist mainly of light elements. For the glass jonomere material, the total F concentration was determined to be about 8\%, and for the fluoride composite material about 4\%.

The equipment employed is inexpensive compared to a Ge detector, although the practical utilisation may be slightly more complicated. The coincidence technique is much less matrix dependent than PIGE. On the other hand, for proton energies of about 2.5 MeV and thick samples, the range of the \alpha particle limits the MDL because the resulting \alpha particle is able to escape from the sample only for approximately the first fourth of the range of the impinging proton. In addition, the coincidence condition and lower cross section naturally decreases the MDL. The MDL for this arrangement, 30 ng/cm$^2$ for an integrated charge of 10 \mu C, can be compared to PIGE data. A MDL of 3 ng/cm$^2$ is reported in Ref. [22] for an integrated charge of 60 \mu C. The new general method, photon-tagged Nuclear Reaction Analysis (pNRA) [23], a method similar to the one presented here but in which the \gamma-photon is detected in coincidence with the proton or \alpha particle, has a comparable MDL to the set-up presented in this article.

From the discussion above, it is clear that applications with large beam charges i.e. macrobeam analysis benefit the most from the low background. At an integrated charge of 170 \mu C, the MDL is better for the system presented in this article than for the PIGE set-up mentioned above, and the difference then increases for larger integrated charges.

The excitation curve in Fig. 6 shows some structure, but it is difficult to evaluate the features for depth profiling from this scan. It would be interesting to perform a full scan made by an auto scan facility from, say, 0 to 3 MeV in very small steps in order to search for narrow resonances. Also, a smaller SBD should be employed to decrease the kinematic spread, which would also allow of a larger beam current, thereby maintaining the efficiency of the analysis. There is little doubt that the yield of the reaction is sufficiently good to be interesting for analytical purposes.

Another way to make use of the reaction under investigation is to use the electron and positron in a coincidence set-up. As was stated before, very few reactions result in internal pair production. Thus if the electron and positron are detected in coincidence, a low-background signal can be obtained. This can be done with two scintillation detectors similar to the one described in this paper, for instance one in the forward and one in the backward direction. By this arrangement several advantages are gained. First, the solid angle can be increased to make it much larger than for an SBD. Secondly, a scintillator set-up allows very high count rates, so that a high beam current can be used without impairing the performance of the detector. With this arrangement, the depth profiling properties are lost unless there are some sharp resonances between or outside the points of the energy scan presented above. Some initial calculations show that the minimum detection limit of this set-up would be better than all figures given for the F analysis by any other reaction. It would be chiefly the beam damage of the sample that sets the limit.

The detectors utilised for these experiments are in-
tended to be included in a more extensive set-up for the analysis of light elements. The signal from the surface barrier detector will then be used to analyse particles from other nuclear reactions. For example, the reaction $^7$Li(p, $\alpha$)$^4$He gives two $\alpha$ particles emitted almost back-to-back. The ones emitted in the backward direction can be detected by the SBD used in the experiments presented in this article, and another annular SBD detects those in the forward direction. Thus, this detector can be used to analyse several elements simultaneously by employing reactions with high cross sections and using large solid angles [24–27].

5. Conclusions

It has been shown that the $^{19}$F(p, $\alpha$,e$^-$.e$^+$)$^{16}$O reaction can be used for analytical purposes. The MDL is 30 ng/cm$^2$ at a proton energy of 2.55 MeV and an integrated charge of 10 $\mu$C on a thin target. A method to use solely the electron–positron pair for the analysis of semi-thick samples has been proposed and discussed.

Acknowledgements

This project was supported by the Swedish Natural Science Research Council (NFR) and the data acquisition system was supported by The Crafoord Foundation. The experimental equipment was made by Günter Matthys at the workshop of the Department of Nuclear Physics in Lund.

References