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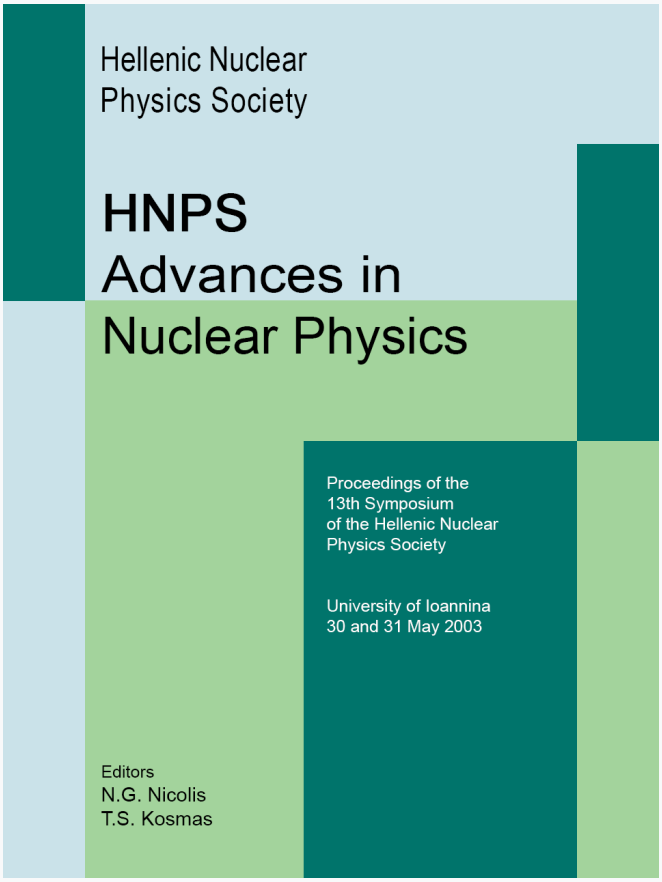

Beryllium determination in non-optimal matrices using the ^{9}\textbf{Be(d,n$\gamma)$}^{10}\textbf{B}

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Beryllium determination in non-optimal matrices using the ^{9}**Be(d,n**γ**)**^{10}**B**

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Abstract

An alternative method for the detection of beryllium in light element matrices is proposed, implementing the use of a deuteron beam at energies from 1 to 2.1 MeV and the ^{9}Be(d,nγ)^{10}B reaction. A HP GE detector of 20% relative efficiency was used to detect the 718 keV gamma ray of ^{10}B. The minimum detection limits obtained for beryllium, are compared to those taken with other NRA techniques (PIGE, heavy-ion and charged-particle spectroscopy) in complex matrices containing high concentrations of light elements. The absolute γ-ray yield of the reaction is also compared to absolute γ-ray yields from literature.

1 Introduction

Beryllium and its chemical compounds are toxic, even in small quantities [1],[2]. Toxification by beryllium is connected to the development of rhinal and dermatological diseases. It is furthermore suspected to be responsible for human carcinogenesis in many cases. The goal of the present work was to determine the concentration of beryllium in soil, asphalt and concrete samples taken from various areas of the former airport of Athens that is now closed. Different types of samples were taken from various areas of the airport. All the samples contained large amounts of light elements.

The detection of beryllium is a very challenging task of ion beam analytical techniques. Several pioneer works for the detection of beryllium, using PIXE and PIGE, have been presented in the past [3],[4]. The alpha-induced Gamma-Ray Emission Spectroscopy (AIGE) [5],[6] and heavy ion gamma ray spectroscopy [7], have also been used with successful results.

The sensitivity of these methods is limited due to the low reaction yield and/or the high background. In addition, in most of them gamma rays with energies higher than 3 MeV have to be detected, thus requiring a detector of relatively high efficiency. AIGE seems to be the most promising technique for, but a high intensity alpha-beam has to be used. Furthermore, the use of alpha particles as well as heavy ions limits the detection ability to the surface of the sample due to the low penetration depth of the beam.

Particle spectroscopy by means of the ^{9}Be(d,a)^{7}Li reaction is proposed for routine measurements [9],[10]. In the present work a complementary method for beryllium detection is presented. It is based on the analysis of the 718 keV gamma-ray deexiting the first excited state of ^{10}B.The latter nucleus is produced via the ^{9}Be(d,nγ)^{10}B reaction. The results obtained with this reaction in this work are compared to those taken using two standard nuclear reactions ^{9}Be(p,γ) and ^{9}Be(d,α) as well as to results already existing in the literature.

2 Experimental Procedure

The experiments were performed at N.C.S.R "Demokritos", Athens, Greece, using the 5.5 MV TN11 TANDEM Accelerator. The deuteron beam energy varied between 1Mev and 2.1MeV. The energy of the proton beam used was 1.2 MeV. For the ^{9}Be(d,a) reaction the detection system consisted of one Si surface barrier detector, of 300μm thickness, at an angle of 150^{o} with respect to the beam axis. For gamma spectroscopy measurements, a HPGe detector, of 20% relative efficiency, was used.

Several reference targets were irradiated: a) one disk of pure metallic Be, b) one cylindrical shaped dental alloy sample containing 2-3% Be c) Three disk shaped reference targets ($Na_2B_4O_7.5H_2O$, NH_4Cl and CaF_2), d) Three of the samples taken from the old airport of Athens including soil, asphalt from the runway and concrete from hangars, were also examined. All targets were irradiated with a deuteron beam at the energy of 1.5 MeV and with a proton beam at the energy of 1.2 MeV in order to compare the results from the three reactions, ^{9}Be(d,a), ^{9}Be(p,γ) and ^{9}Be(d,nγ). At the energy of 1.5 MeV cross section data for most relevant reactions are available. The selection of the energy of 1.2 MeV for the proton beam was made after considering the equivalent deuteron penetration depth in the samples. At the same time, this proton energy covers the region of the broad resonance of the ^{9}Be(p,γ) reaction cross section at 989 keV and the narrow resonance at 1083 keV. The accumulated charge in the case of the (d,a) reaction was 4μCb for all targets and for the (p,γ) and the (d,nγ) reactions it was 20μCb. Additionally the pure beryllium target was irradiated in the energy interval of 1-2.1 MeV to obtain the thick target yield of the ^{9}Be(d,nγ) reaction and thus determine the optimum deuteron beam energy for Be detection. The dead time for the (d,a) reaction measurements did not exceed 2% while for the (p,γ) reaction measurements it was negligible (less than 0.03%). In the case of the (d,nγ) reaction the dead time for the measurements was less than 8%.

At the optimum beam energy additional measurements were carried out with the use of a more optimized setup, which allowed the increase of the beam current without increasing the dead time. This was achieved using a small shaping time, of 1μs and via the selection of an energy window around the energy of 718 keV, using a discriminator. The use of this more optimized setup allowed the accumulation of 150 μCb in a reasonable and applicable time due to the higher beam current.

3 Results-Discussion

The spectra taken with the ^{9}Be(d,a) reaction at the deuteron energy of 1.5 MeV for the pure Be target, the dental alloy and of the typical soil sample are shown in figure 1. In the pure Be spectrum, the counts present in the area between channels 70-250, come from the ^{9}Be(d,a_0), ^{9}Be(d,a_1), ^{9}Be(d,p_0) and ^{9}Be(d,p_1) reactions. In the case of the dental alloy sample, no other light elements are present, apart from a peak coming from the ^{12}C(d,p) reaction. However in the case of the soil sample, due to the existence of other light elements, in order to identify the observed peaks and counts, reference targets had to be irradiated using the same setup.

In figure 2, the spectra of the reference targets, $Na_2B_4O_7.5H_2O$, NH_4Cl and CaF_2, together with the pure beryllium spectrum are presented. The peaks present in the spectra come from the (d,a) and (d,p) reactions on B, O, N, F and Be correspondingly. In the region where the beryllium counts appear, there are also counts coming from all other reactions, concerning elements whose expected concentration levels are several orders of magnitude higher than those of beryllium. There is a small region in the spectrum that can be considered as a low background region and thus can be used for the deduction of the minimum detection limits (MDLs) of Be in the sample.

For gamma spectroscopy, the study of the ^{9}Be(p,γ) and the ^{9}Be(d,nγ) reactions, was carried out via the detection of gamma rays from the de-excitation of ^{10}B which was the produced nucleus in both reactions. The gamma ray of interest was the 718 keV transition from the first excited state to the ground state, due to its high intensity.

The ^{9}Be(p,γ)^{10}B reaction spectra, at the energy of 1.2 MeV, are presented in figure 3. The 718 keV peak is clearly observed in the spectra of the pure Be target. In the case of the 2% concentration in beryllium of the dental alloy, the 718 keV peak just emerges from the background, while in the case of the soil sample spectrum there are no counts in the corresponding area that could safely be distinguished from the background.

Due to the low absolute gamma ray yield and the high background coming from the interaction of the light element matrix with the proton beam, this reaction is less preferable than the ^{9}Be(d,a) one (higher MDLs).

The spectra taken at the deuteron energy of 1.5 MeV for the ^{9}Be(d,nγ) reaction are presented in Figure 4. The 718 keV gamma ray is clearly observed in the spectra of the pure Be target and the dental alloy having a much higher absolute peak-to-valley ratio than the corresponding (p,γ) spectra. The detection limits deduced with this reaction are considerably lower. In the pure beryllium spectrum and in the dental alloy spectrum, the 1022 keV γ-ray coming from the transition of the second excited state to the first excited state of ^{10}B (1740.15 keV $\rightarrow$ 718.3 keV) is also observed. Additionally in all spectra, peaks coming from reactions of the produced neutrons with the surrounding materials (holder, scattering chamber, etc.) or the detector itself, are also present.

In order to obtain the optimum deuteron beam energy, the thick target yield of the reaction, using the pure beryllium foil, was measured, in the range of 1 MeV to 2.1 MeV The ratio of yield/$\sqrt{BGD}$ was then used as the "figure of merit" of the statistical variations, proposed by Currie [11]. The optimum deuteron energy was found to be 1.85 MeV.

Table 1 presents the extracted absolute gamma ray yields for the (p,γ) and

(d,nγ) reactions studied in this work, together with absolute gamma ray yields for reactions studied in the literature. The absolute yield of the (d,nγ) reaction is among the highest γ-ray yields of the reactions present in this table.

The MDLs for all reactions studied in the present work, for the typical soil sample, are presented in table 2. The MDLs extracted with the ^{9}Be(d,nγ) reaction for all samples, together with the dental alloy, are presented in table 3. Due to the alloy composition of high-Z elements, these MDLs are much lower.

As far as the soil sample is in concern , further reduction of MDLs could be achieved via the use of smaller pulse shaping time ,by reducing even more the defined energy window, or by acquiring more charge on target. However, there are some limiting factors to the use of this reaction which concern the contributions of the deuteron break-up that become important for energies above 2 MeV and since this is a neutron production reaction, the use of higher beam currents increases the background coming from neutrons. Despite these limitations, the absolute gamma ray yield is comparable or even higher than other reactions studied in the literature, which have impressive results. Some of these reactions obtained MDLs for beryllium lower than 1 ppm. This is strong evidence that the detection limits obtained with this reaction can be further reduced with the use of a more optimized setup.

4 Conclusions

The ^{9}Be(d,nγ) reaction can be used as an alternative reaction for the detection of beryllium. The study of this reaction takes place via the detection of the 718 keV gamma ray of ^{10}B with the use of a 20% relative efficiency Ge detector. Apart from the fact that this reaction offers a high absolute gamma ray yield, it also has the advantage of the use of a low efficiency detector due to the detection of a low energy gamma ray. Although for the specific case the MDLs deduced at the present work were satisfactory, with the use of a more optimized setup, the minimum detection limits are expected to be further reduced.

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Table 1: Absolute gamma ray yields for reactions studied in the present work, together with results from the literature.

Reaction	**E_γ (keV)**	**E_{Beam} (MeV)**	**Absolute γ-ray Yield/μCb.Sr**
^{9}Be(d,nγ)^{10}B	718	1.85	8.5×10^6
^{9}Be(p,γ)^{10}B	718	1.2	4×10^3
^{9}Be(p,γ)^{10}B [4]]	3562	2.4	2.5×10^4
^{9}Be(^{12}C,a)^{17}O [7]	3842	28	6.1×10^6
^{9}Be(^{12}C,2a)^{13}C [7]	3853	28	6.1×10^6
^{9}Be(^{12}C,p3n)^{17}F [7]	3857	28	6.1×10^6
^{9}Be(a,γ)^{12}C [6]	4439	2.4	1.6×10^6

Table 2: The ^{9}Be MDLs for the three reactions studied in the present work, for the typical soil sample.

Reaction	**Detection Limits (ppm)**
^{9}Be(d,a)	1740±40
^{9}Be(p,γ)	17300±800
^{9}Be(d,nγ)	72±5

Table 3: MDLs for all samples using the ^{9}Be(d,nγ) reaction at the optimum energy of 1.85 MeV.

Sample	**Detection Limit (ppm)**
Dental Alloy	20±1
B5 (Soil)	72±5
G2 (Asphalt)	73±5
Υp2 (Concrete)	84±5

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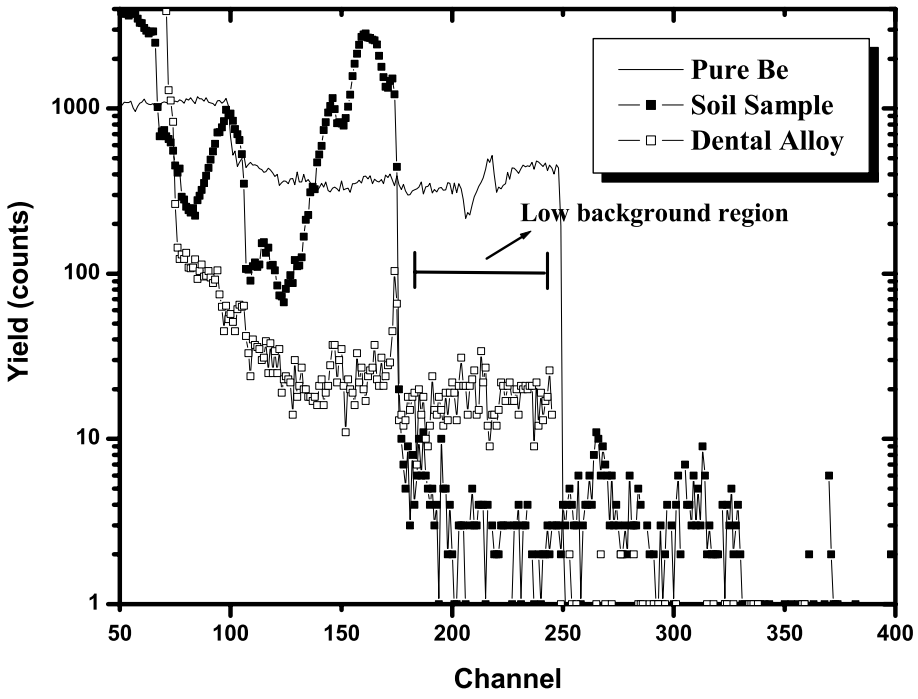

Figure 1: Spectra from the (d,a) and (d,p) reactions at 1.5 MeV deuteron beam for 4μCb accumulated charge. a) Metallic Be target, b) dental alloy c) Typical soil sample.

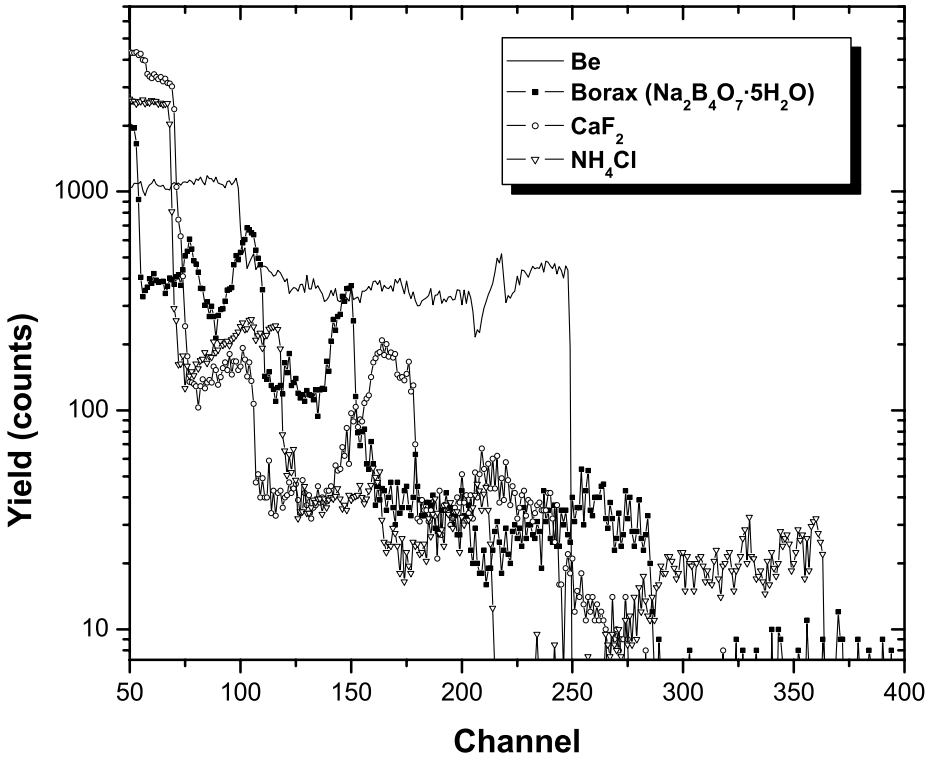

Figure 2: Spectra from d-induced reactions on the reference targets taken at 1.5 MeV deuteron beam for 4μCb accumulated charge.

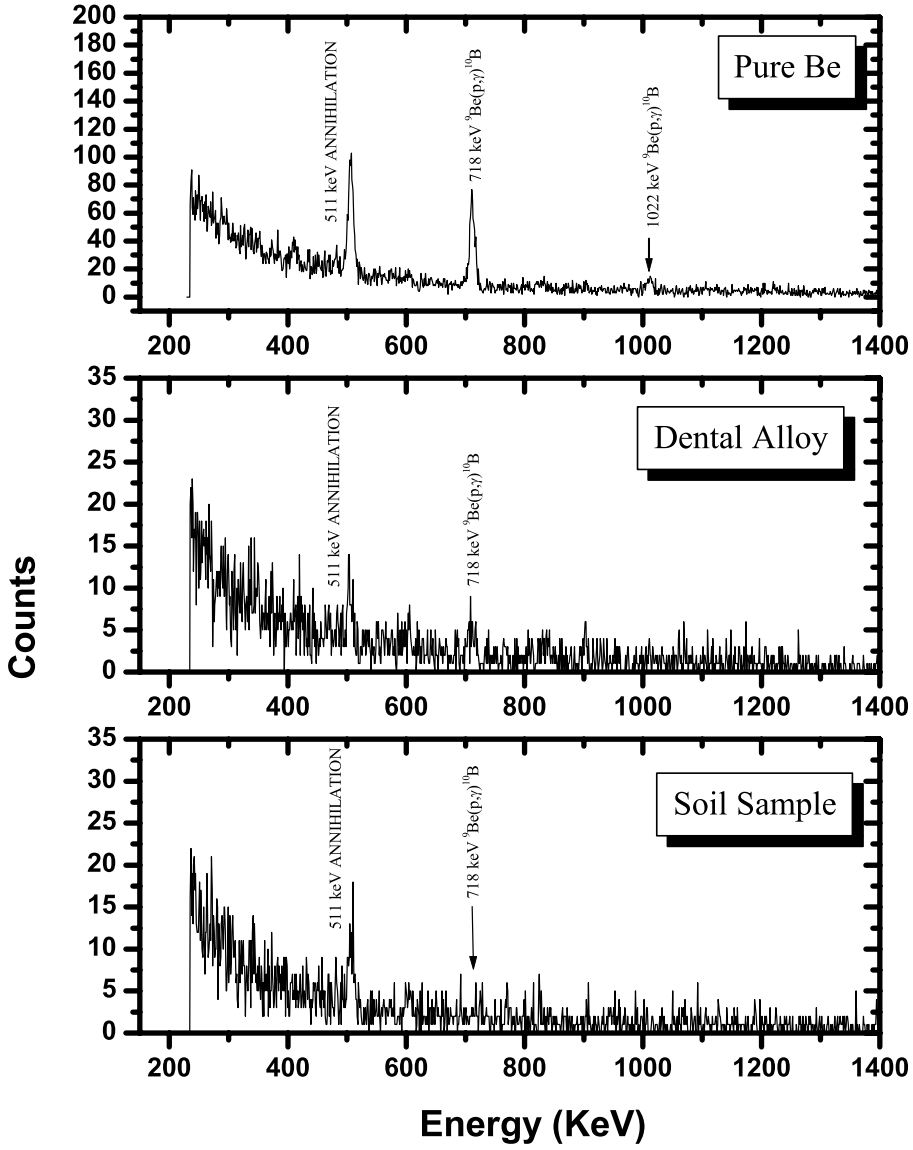

Figure 3: Spectra taken from the (p,γ) reaction taken at 1.2 MeV proton beam, for 20μCb accumulated charge.. a) Metallic Be target, b) (dental amalgam) c) soil sample.

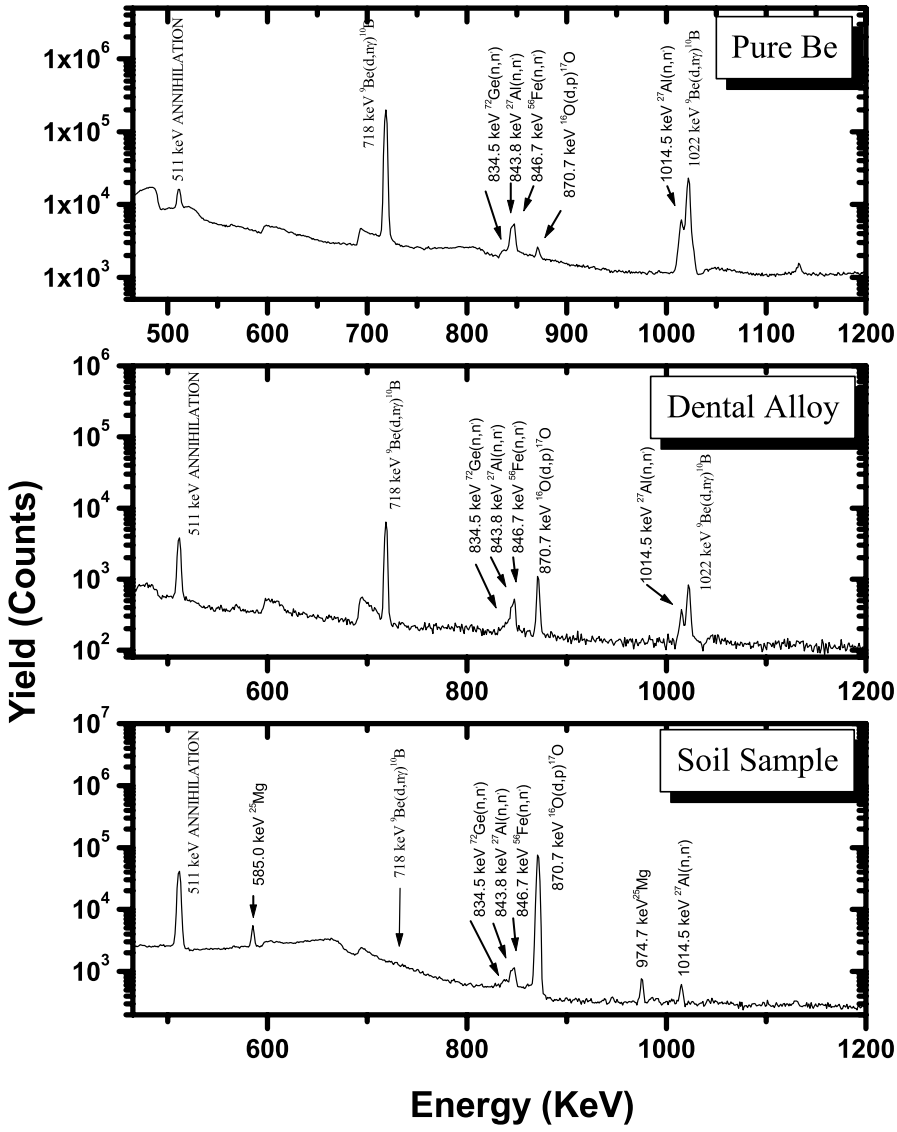

Figure 4: Spectra taken from the (d,nγ) reaction taken at 1.5 MeV deuteron beam energy, for 20μCb accumulated charge. a) Metallic Be target, b) dental alloy c) Typical soil sample