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THE INFLUENCE OF RECOIL LOSSES IN SUBSEQUENT α DECAY ON THE DETERMINATION OF α-BRANCHING RATIOS

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ABSTRACT

The fraction of daughter nuclei that recoil out of the catcher foil after α decay of mass–separator implanted mother nuclei is determined experimentally and compared with calculations. The correction that has to be applied on the α–branching ratio, obtained by comparing the mother and daughter activity in one α detector, is at least 20%. Previously published branching ratios can be wrong due to the underestimation of the correction procedure. As an example, the $^{202}Rn \rightarrow {}^{198}Po \rightarrow {}^{194}Pb$ decay chain is studied.

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Abstract

The fraction of daughter nuclei that recoil out of the catcher foil after α decay of mass–separator implanted mother nuclei is determined experimentally and compared with calculations. The correction that has to be applied on the α–branching ratio, obtained by comparing the mother and daughter activity in one α detector, is at least 20%. Previously published branching ratios can be wrong due to the underestimation of the correction procedure. As an example, the $^{202}Rn \longrightarrow {}^{198}Po \longrightarrow {}^{194}Pb$ decay chain is studied.

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1. INTRODUCTION

Far from stability, the α decay is sometimes, due to the lack of suitable projectile–target combinations, the only available spectroscopic tool. The relative α width (δ^2[MeV]), one of the observables of the α decay, is proportional to the ratio of the α–transition probability λ to the penetration factor P [1]. The latter is the probability for an α particle to tunnel through the Coulomb and centrifugal (if angular momentum is transferred) barrier. The δ^2 value is the probability of forming an α particle inside the nucleus and contains all the information on nuclear structure. For instance the systematics of δ^2 values in the Pb region have been used to question the Z=82 shell closure for the neutron–deficient Pb nuclei [2]. The experimental uncertainty on the relative α widths is mainly due to the uncertainty on the half life and on the α–branching ratio (α_{br}); the accuracy on the α–decay energy is normally sufficient. Alpha–branching ratios are reported in literature on the basis of three different techniques:

–comparison of α yields with absolute cross–section estimates

–comparison of α and β^-, β^+/EC and/or IT decay

–comparison of the α decay of the feeding parent nucleus and the daughter nucleus.

The first method is as reliable as the absolute cross–section calculation. Far from stability, this can lead to order–of–magnitude errors. The second method has as main disadvantages the fact that one uses a two–detector set–up and that one needs a precise knowledge of the β (and/or IT) decay scheme in order to scale the intensity of the observable (γ rays, X rays, electrons and/or positrons) to the number of decays. The third method is in principle the most interesting as it requires only one detector of which the absolute efficiency ought not to be known. Neither is the knowledge of the other decay channels necessary. However, the method has to be used in connection with a mass or recoil separator. In the case of a recoil separator the relation between the mother and daughter α decay can, for short–living nuclei, be verified by the time correlation. At an on–line mass separator the mass resolving power is in most cases high enough to ensure a sufficient suppression of unwanted A–4 mass contamination (A is the mass of the mother nucleus). The α–branching ratio is then in principal the ratio of the daughter to mother α–ray intensity, corrected for the different half–lives. However, corrections need also to be made for the fact that the daughter nucleus, due to the recoil energy after α decay, is sitting at a different place than the mother

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After extraction, the ions are mass separated and implanted at the detector set–up. The described process for on–line produced activity holds also for the decay of the target atoms. The recoil energy is now coming from the α decay, this means that only α–decay daughters are obtained as ion beams: e.g. ^{227}Ac, ^{223}Ra, ^{219}Rn, ^{215}Po, ^{211}Pb and ^{207}Tl are possible beams, but ^{227}Th and ^{211}Bi are not produced directly as they are β–decay daughters. The ion–guide separation is fast: also the 1.8 ms ^{215}Po is separated efficiently ($\sim 10^3$ atoms/s).

The ^{215}Po beam is implanted into an aluminized mylar tape in front of which an α detector is placed (see figure 2). The α detector records the α lines of ^{215}Po and ^{211}Bi. The data taking is organized in a cycle (see figure 3). The time elapsed between the beginning of the "beam–on" period and the detection of the α particle is measured with a Time to Digit Converter (TDC). Figure 3 shows the TDC spectra of the α lines of ^{215}Po (a) and ^{211}Bi (b). After 4320 seconds of beam–on time, the separator beam is switched off and the detection of the ^{215}Po α line falls to zero (fig 3a). The α detector records the ^{211}Bi activity sitting on the tape and on the detector itself due to recoil–implantation. After another 1500 seconds, the tape is moved and only the ^{211}Bi activity on the detector is recorded. The time pattern of the ^{211}Bi activity is mainly determined by the ^{211}Pb half life.

If we denote the recoil fraction by R, the detection efficiency of an α signal by Ω, the solid angle of the detector divided by 4π, one has a total efficiency for the detection of the parent α decay :

$$\epsilon_P = \Omega \qquad (1)$$

and in first order for the daughter decay :

$$\epsilon_D = (1 - R).\Omega + R.\frac{\Omega}{R}.\epsilon_I \qquad (2)$$

with ϵ_I, the efficiency for detecting the α radiation from nuclei implanted in the detector. In the first and second part of the cycle, we can use these expressions and fit the mother–daughter–grand daughter relation in order to get the correction term $(1–R+\epsilon_I)$. In the third part of the cycle the daughter activity, recoil–implanted in the tape, is moved away from the detector by the tape transport and the efficiency for detecting the daughter activity is reduced to $(\Omega.\epsilon_I)$. As a fit parameter, we will

extract here the term ϵ_I. The results from the fits are :

$$\epsilon_I = 0.50\ (1)$$
$$1 - R + \epsilon_I = 1.19\ (4)$$

and as deduced value for the recoil fraction one obtains :

$$R = 0.31\ (4).$$

The production rate of ^{211}Bi was obtained from the ^{215}Po data (fig. 3a). The error on 1–R+ϵ_I is quite large due to the difference in peak shape between the mother and the daughter α line. An example of this difference in shape will be given in section 4. The value for ϵ_I is in agreement with what is expected for nuclei implanted in the detector: half of 4π or 0.5. We must remark here that these values are in first order independent of the absolute efficiency of the α detector (Ω).

3. Calculation of the recoil fraction

The experimental situation as described above can be simulated using a computer code that calculates the passage of radiation through matter. We have used the TRIM85 code [4]. In order to simulate our situation, the program, originally written to calculate implantation profiles, was modified as follows : first, 50 keV ^{215}Po atoms impinge on aluminum, at an angle of 30° with the normal on the surface. When a ^{215}Po atom comes to rest, it's identity is changed : it becomes a ^{211}Pb atom with an energy of 140 keV (the recoil energy from the ^{215}Po α decay). Note that the ^{211}Bi will be at the same place as the ^{211}Pb since it is a β decay daughter. The direction in which the ^{211}Pb atom starts moving through the aluminum is chosen randomly, corresponding to an isotropic α emission. The ^{215}Po implantation profile and the ^{211}Pb recoil distribution, as well as the energy and angle of the ^{211}Pb atoms recoiling out of the aluminum, are stored. The calculated fraction of daughter atoms that recoil out of the aluminized mylar tape is 0.34, in agreement with the experimental value: 0.31 (4).

4. The influence of the recoil fraction on the determination of α–branching ratios; an example : ^{202}Rn to ^{198}Po decay

The percentage of daughter nuclei that recoil out of the medium in which the mother activity has been implanted by a 50 kV mass separator is minimum 31% for a thick catcher (like an aluminized mylar tape) but can be substantially higher for thin catcher foils (carbon foils of 20 $\mu g/cm^2$ are often used). In the literature, this effect is described as recoil loss. But when the mother decay is in front of the detector, there is also a substantial enhancement in detecting the daughter activity. The net result of these two effects is, on its turn, strongly dependent on how the data acquisition is organized. For instance, if the data is taken at the decay station of a moving tape system, corrections have to be made for recoil losses at the implantation station, for decay losses during transport, for recoil losses at the decay station and for recoil implantation in the detector at the decay station. All these corrections are very sensitive to the decay characteristics of the mother and the daughter nucleus.

The α decay of neutron–deficient Rn isotopes has been studied by P. Hornshoj et al. [3]. The α–branching ratios of the Polonium daughters with mass ranging from 202 down to 197 have been deduced by comparing the α decay of the Rn mother to the Po daughter. The detection was at the decay station of a moving tape collector. The authors indicate that the correction for the loss of daughter activity was small.

We have remeasured the α–branching ratio of ^{198}Po in order to verify the validity of the corrections mentioned in reference [3]. The ^{202}Rn activity is produced at LISOL by the heavy–ion fusion–evaporation reaction of ^{20}Ne on ^{nat}Ir. The reaction products recoil inside a FEBIAD source, are ionized, mass separated and implanted into an aluminized mylar tape. More details on the LISOL separator can be found in reference [8]. The used detector set–up is very similar to the one described in figure 2. Time–sequential singles α spectra are taken at the implantation station. The cycle is divided in six periods of 100 seconds; the tape moves every 600 seconds the activity away. Figure 4 gives the sum of the α spectra taken over all periods andcycles. The α–branching ratio can be deduced out of the intensity of the ^{202}Rn (I_{Rn}) and ^{198}Po (I_{Po}) peak and using the following correction:

$$\alpha_{br} = \frac{I_{Po} \cdot C_{Rn}^{T1/2}}{I_{Rn} \cdot \{C_{Po}^{T1/2} \cdot [1-R] + C_{Rn}^{T1/2} \cdot \epsilon_I\}} \qquad (3)$$

with $C^{T1/2}$ the correction term for the 600 s implantation (0.976 (1) for Rn and 0.727 (5) for Po). By using our measured values for R and ϵ_I we obtain for the α–branching ratio of ^{198}Po a value of 0.61 (7). The ^{198}Po peak intensity in the spectrum of figure 4 is for 50% coming from activity sitting in the tape ($C_{Po}^{T1/2}.[1-R]$) and for 50% coming from activity recoil–implanted in the detector ($C_{Rn}^{T1/2}.\epsilon_I$).
Ignoring these corrections would have led to an alpha branching ratio bigger than 1.
The 50–50 fraction of α decays from the tape or from the detector explains the time behavior of the ^{198}Po peak in the 6 times 100s implantation cycle. The rather high uncertainty of the α–branching ratio, mainly due to the intensity determination of the ^{198}Po peak, makes it impossible to evaluate the correction procedure of reference [3]. The values agree within the error limits (α_{br} value from [3] is 0.70 (8)).
A possibility to obtain a more accurate intensity of the ^{198}Po peak is to reduce the isobaric contamination of ^{202}At. The ISOLDE isotope separator has a target–ion–source system with a cooled transfer line between the target and the ion source: only the Rn noble gas passes through the transfer line [9]. The Rn nuclei, with masses ranging from 199 to 228, are produced in the spallation of a thorium carbide target by 600 MeV protons. In order to minimize the half–life correction factors, the activity is implanted for several daughter half–lives into the tape; the α detector is mounted at the implantation site. As the implantation characteristics are slightly different compared to the LISOL conditions (at ISOLDE : 60 kV acceleration and an implantation angle of 45^0), the 1–R+ϵ_I value is remeasured using the ^{218,219,220}Rn decay chains. For the three decay chains a value of 1.19 (4) is obtained, identical to the value measured at LISOL. This means that the effect of the differences in implantation energy, –angle and α–decay energy (6.288 MeV for ^{220}Rn and 7.133 MeV for ^{218}Rn) on the 1–R+ϵ_I value is not visible within the error bars. Figure 5 gives the α spectrum obtained after 3670 seconds of implantation. Only one peak from ^{202}At is present in the spectrum. This is the α decay of the low–spin ground state of ^{202}At fed in the β^+/EC decay of ^{202}Rn [10]. The right shoulder of the ^{198}Po peak is due to the activity implanted in the detector; when moving the tape, only this part remains. The precision on the intensity of the ^{198}Po peak is much better in this case and we obtain a more precise value for the α–branching ratio of ^{198}Po: 0.57 (2).

The value obtained from the 6x100s implantation cycle at LISOL [0.61 (7)] is in agreement but the published value [0.70 (8)] from Hornshoj et al. [3] is too high. We have remeasured all the branching ratios from ^{204}Po down to ^{197}Po [10] and our values are systematically 20% lower than the values from reference [3]. This deviation is probably caused by the used correction procedure for recoil loss out of the tape and recoil implantation in the detector.

6. Conclusion

At on–line mass separators, the recoil energy of the daughter nuclei after α decay is higher than the implantation energy of the mother nuclei, part of it recoils out of the tape. This means that the α radiation from the daughter decay comes from a different location than the α radiation from the mother decay. This can lead to systematic errors in the measurement of α–branching ratios. We have studied this effect by using α–decay chains with precisely known branching ratios. Two corrections are necessary: one for the recoil loss of daughter activity out of the catcher material and one for the enhanced detection efficiency for the α radiation of daughter activity recoil–implanted in the detector. Furthermore, if the catcher is moved periodically, the correction for the time behavior of the daughter nuclei has to be treated separately for the activity in the catcher and in the detector.

The experimental value for the recoil fraction can be reproduced by the TRIM85 code [4]. Although the experimental conditions at the LISOL and ISOLDE separator are slightly different, the same corrections can be used. The uncertainty on the recoil fraction is too high to be sensible to the different conditions. Also the difference in α–decay energy in e.g. the decay of ^{218}Rn (7.133 MeV) and ^{220}Rn (6.288 MeV) is not visible in the recoil fraction. The main uncertainty on the recoil fraction comes from the difference in peak shape of the mother and daughter α line. The tailing to both the low–energy and high–energy side of the daughter line due to the change in site after the recoil implantation makes a proper integration difficult.

The determination of the α–branching ratio of ^{198}Po has been used as an example. The measured value of 0.57 (2) is lower than the previously published value [3], this effect is present in all our new results [10] and indicates that previously published branching ratios can be wrong due to the underestimation of the recoil correction procedure.

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Figure captions

1. The natural radioactive family of ^{235}U. α decay is denoted by ← and β decay by ↘.
2. The experimental set–up : the α detector (PIPS 150 mm^2) faces the tape at the implantation position (distance = 30 mm).
3. The TDC spectrum of the α lines of ^{215}Po (a) and ^{211}Bi (b), where the latter includes the least square fit (see text).
4. The α spectrum of mass 202, taken in a 6x100 s. cycle at LISOL. The α energies are given for ^{198}Po and ^{202}Rn in keV.
5. The α spectrum of mass 202, taken at ISOLDE for 3670 s at the implantation site . The broadening of the ^{198}Po peak is due to the broad distribution of the ^{198}Po atoms after the recoil from the α decay of ^{202}Rn. The α energies are given for ^{198}Po and ^{202}Rn in keV.

U 235
7E8 y
Th 231
1.063d
Pa 231
32800y
Ac 227
21.77y
1.38 %
Th 227
18.72d
Ra 223
11.43d
Rn 219
3.96s
Po 215
1.78ms
2.3E-4%
Pb 211
36.1m
Bi 211
2.14m
0.273 %
Tl 207
4.77m
Pb 207
stable

Fig. 1

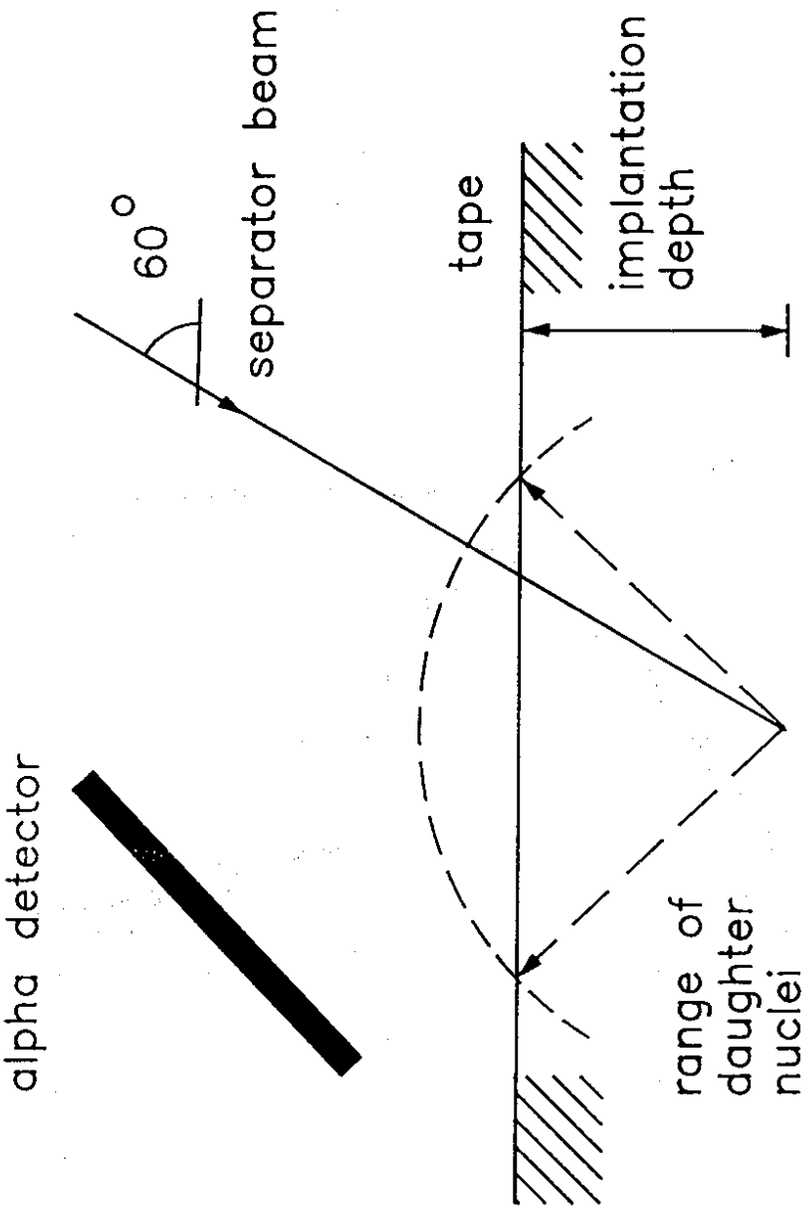
alpha detector
60°
separator beam
tape
implantation
depth
range of
daughter
nuclei

Fig. 2

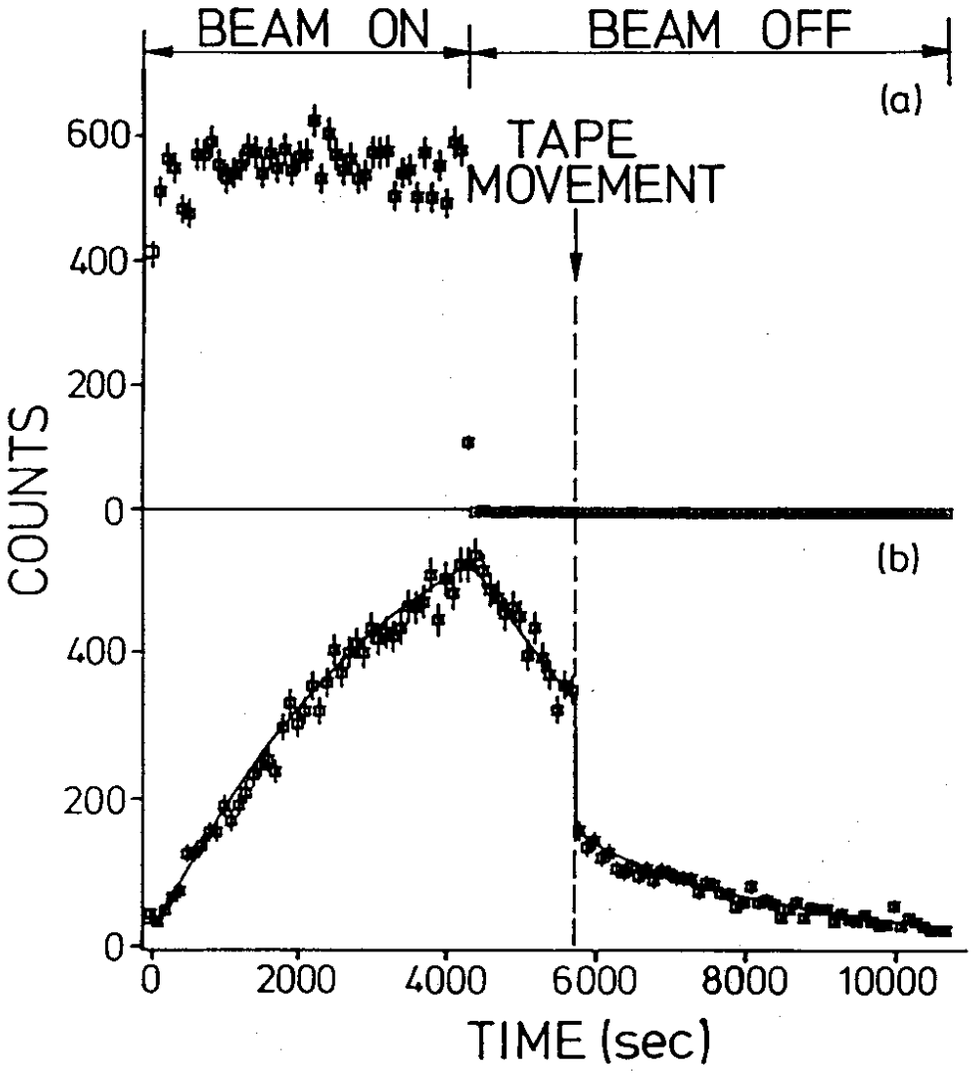
BEAM ON
BEAM OFF
(a)
TAPE
MOVEMENT
600
400
200
0
COUNTS
(b)
400
200
0
0
2000
4000
6000
8000
10000
TIME (sec)

Fig. 3

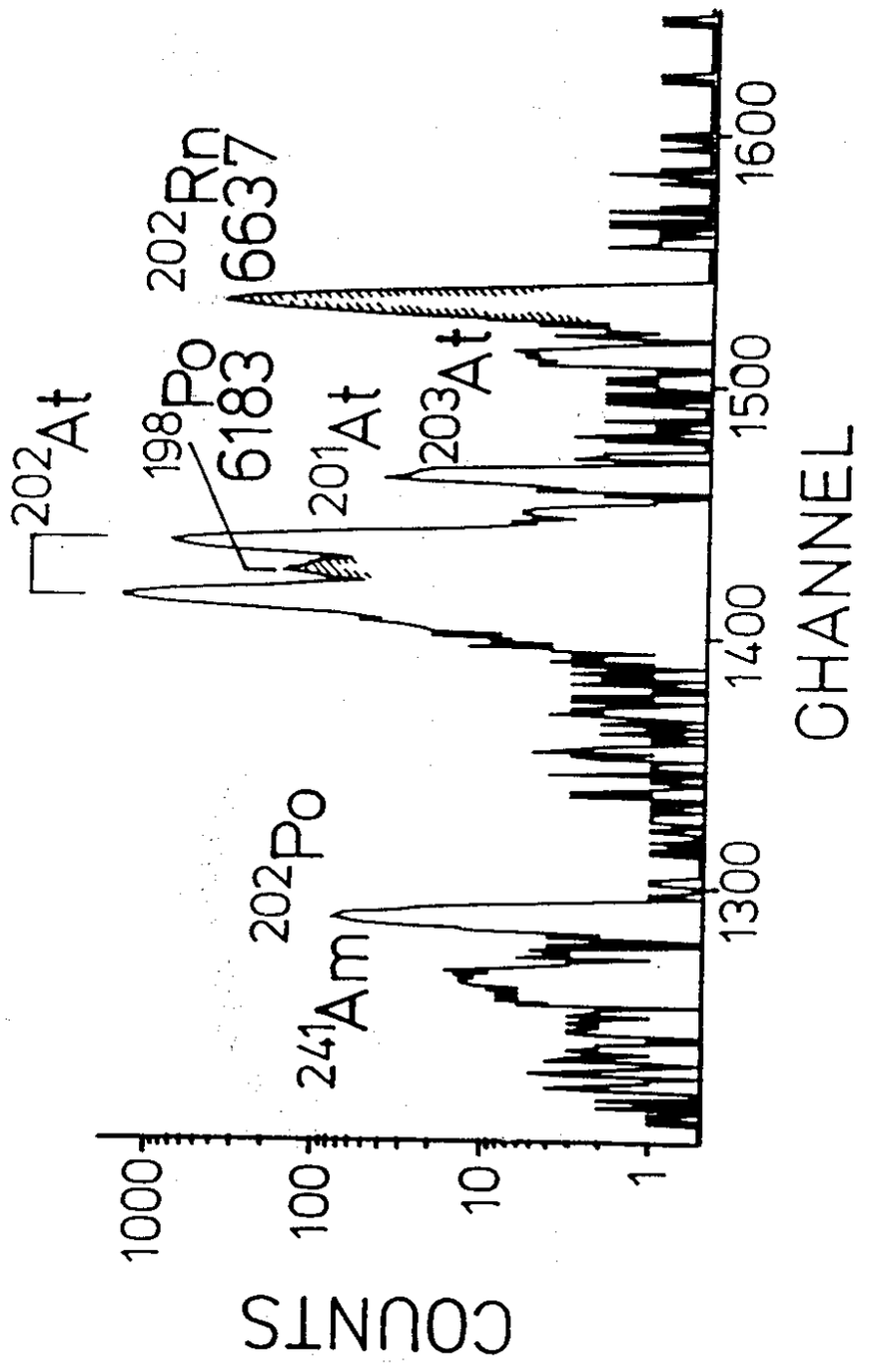
COUNTS
1000
100
10
1
CHANNEL
1300
1400
1500
1600
241Am
202Po
202At
198Po
6183
201At
203At
202Rn
6637

Fig. 4

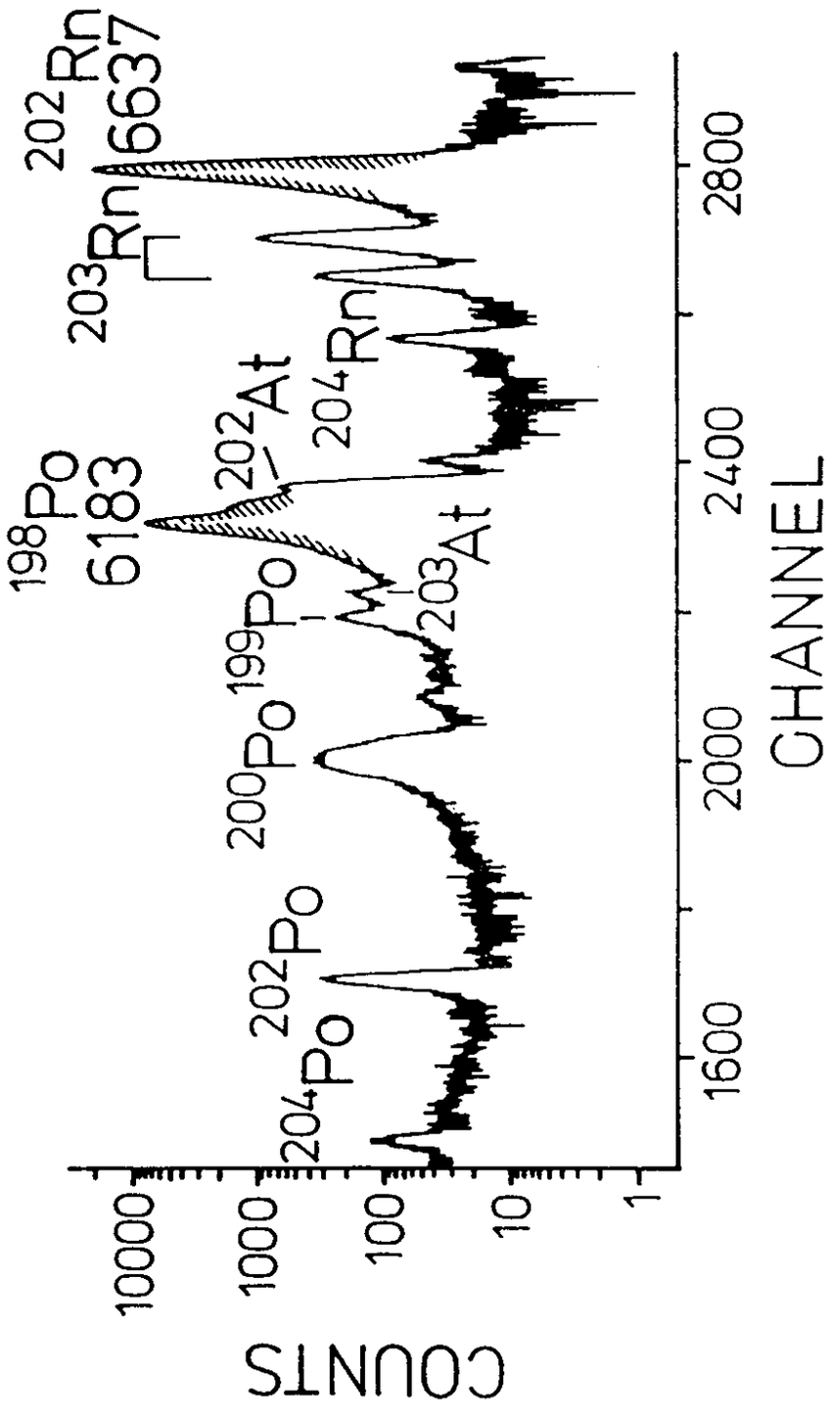

Fig. 5

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