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Alpha spectroscopy for thoron progeny implantation in different materials

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Abstract

A study involving thoron daughters implanted at the surface of glasses or other materials can be useful in the simulation of radon progeny implantation. This simulation is possible due to the equilibrium of implantation obtained in a short time (about 50 h) compared to about 100 years in the case of ^{222}Rn. The purpose of this work is to obtain some implantation characteristics using alpha spectra for thoron daughters implanted into the surface of different materials and to show the possibility of this method to simulate the radon progeny implantation. Measurements were made using a Canberra PIPS detector (900 mm^2) with 25 keV energy resolution. The samples were obtained by exposure of different materials in a small (0.3 l) thoron vessel containing 200 g of $Th(NO_3)_2 \cdot 5H_2O$ as a source of thoron. The first results showed that alpha spectra of implanted thoron progeny can be obtained and that the width of the alpha peaks depends on the exposed materials. The constant of implantation estimated for all used samples varies between 8 and 52 mBq/m^2:Bq/m^3 for the case of equilibrium.

Keywords: Thoron progeny; Deposition; Alpha spectra; Implantation factor; Energy peak

1. Introduction

In order to estimate the risk of indoor radon in producing lung cancer, a lot of epidemiological studies were developed, using underground miners data. Extrapolating these results for the general public, based on the linear no-threshold relationship between radon exposure and lung cancer risk, is now a problem widely discussed by the scientific world [1,2]. In the US, for example, the BEIR IV committee estimated that about 15,400 or 21,800 lung cancer deaths per year can be attributed to radon among eversmokers and neversmokers. Considering the results of the latest epidemiological studies, the BEIR VI committee, based on the uncertainties of these analyses, extended this interval to 3000–32,000 deaths per year. However, the committee admits that it could not exclude the possibility of a threshold relationship between exposure and lung cancer risk at very low levels of radon exposure.

To improve the results of the latest epidemiological case–control studies, and therefore to diminish the influence of the multiple parameters

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involved in radon relative risk estimation, an alternative would be a correct assessment of the past radon exposure of each subject in these case–control studies. This past exposure parameter is very important and the reduction of its uncertainties can be done using dose reconstruction of past radon exposure [3,4]. Many retrospective studies regarding past radon exposure are based on the accumulation of ^{210}Pb on the surface of glass objects or other materials, and measuring the specific alpha-activity of ^{210}Po ($T_{1/2} = 138$ d, $E_{\alpha} = 5.3$ MeV) resulting from radon daughters implanting due to their alpha recoil. Because ^{210}Pb, grandmother of ^{210}Po, has a lifetime of 22.3 yr, the glass acts as a memory for the airborne radon-activity over several decades. Another way [5] to measure the specific surface-activity of implanted radon daughters is beta-activity counting of ^{210}Bi (parent of ^{210}Po). The measurement of implanted ^{210}Po or ^{210}Bi activity can be correlated to the average past exposure using the Jacobi room model [6–9]. In the case of thoron exposure (^{220}Rn) this method is not applicable because there is not a long lifetime daughter to allow a significant accumulation over many years of any descendant. The longest lifetime of thoron progeny is that of ^{212}Pb ($T_{1/2} = 10.6$ h). But a study involving thoron daughters implanted on surface of glasses or other materials (plexiglass, paper, wood, copper, aluminum or other metals) can be useful for the simulation of radon progeny implantation and for the study of aerosol influence on this phenomenon. By following the intensity modification of implanted activity of alpha peaks over time in different conditions it could be possible to assess the desorption constant of the implanted atoms. This simulation is possible due to the equilibrium of implantation, which is obtained in a short time (about 50 h) compared with 100 yr in the case of radon daughters. The alpha spectroscopy method used for the plate-out of radon and thoron progeny was that of Bigu [10]. Some studies characterizing the adsorption–desorption of thoron daughters on aerosols have also been made lately [11–13].

The purpose of this work is to obtain alpha spectra for thoron daughters implanted on the surfaces of different materials and to find some characteristics of the implantation process.

2. Experimental methods

Alpha spectrometry was carried out using a Canberra PIPS alpha detector (900 mm^2) with 25 keV energy resolution operated under vacuum conditions and connected to a pulse height analyzer of the type ICA-70-Hungary. Alpha energy calibration was determined using the internal standard pulses generated by the alpha spectrometer. The detection efficiency was calculated taking into consideration the solid angle under which the detecting surface (900 mm^2) is seen from the sample area. The samples consisting of squares of 1 cm side are placed on the detector axis, at 5 mm

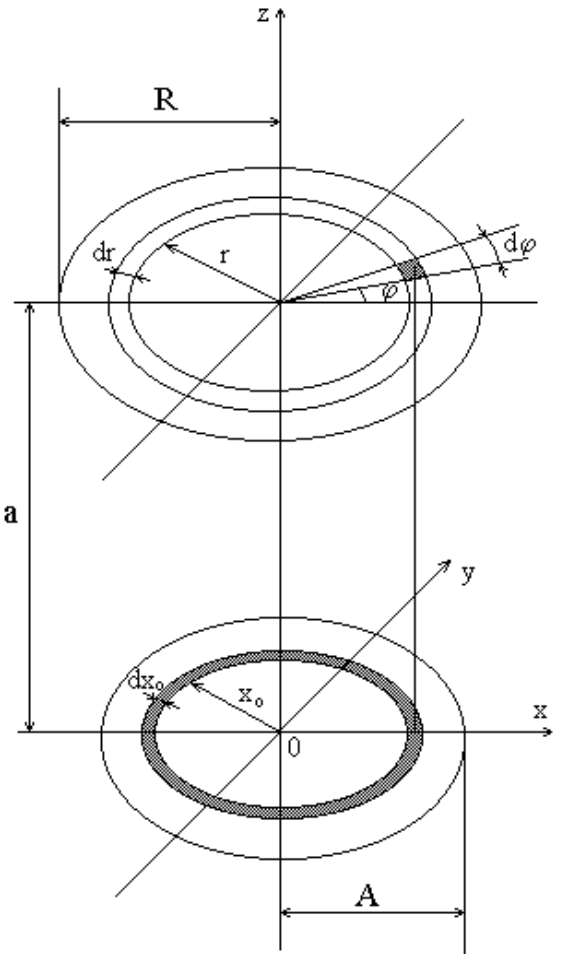

Fig. 1. Geometric arrangement for solid angle calculation (detection efficiency).

distance. The solid angle for this arrangement (Fig. 1) was computed by solving the following triple integral [14]:

$$\Omega = (a/\pi A^2)\int_0^A 2\pi x_0\,\mathrm{d}x_0\int_0^R r\,\mathrm{d}r \times \int_0^{2\pi}\mathrm{d}\varphi/(r^2+x_0^2+a^2-2rx_0\cos\varphi)^{3/2}, \quad (1)$$

where $a = 5$ mm is the distance between the sample and the entrance of detector, R is the detector radius and $A = 5.64$ mm is the equivalent radius of a circle having the same area as the samples (100 mm^2). The efficiency calculated in this way was 35.5%.

3. Samples exposure

Two hundred grams of $Th(NO_3)_2 \cdot 5H_2O$ powder were put into a metallic box of 8.5 cm diameter and 5.0 cm height, Fig. 2. This substance was purchased over 25 years ago, therefore the equilibrium between ^{232}Th and ^{234}Ra is almost reached (95%). A thin plastic sieve was placed on the thorium nitrate. The thickness of the $Th(NO_3)_2 \cdot 5H_2O$ layer was 2.9 cm and that of the layer of air above was 2.1 cm. Taking into account the very short live time of thoron (1 min), the equilibrium concentration is quickly reached in V_2 (165 cm^3) after closing the box. We considered that a time period of about 30 min allows the diffusion of the thoron generated from $V_2 = 165$ cm^3 into $V_1 = 120$ cm^3. The amount of thoron in volume V_1 was determined in a separate experiment by isolating the thorium nitrate in a 0.5 l plastic bottle hermetically closed, the thickness of thorium nitrate being the same as in the box from Fig. 2.

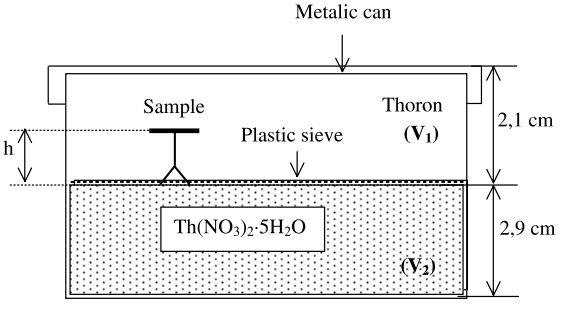

Fig. 2. The container for the irradiation of samples with thoron.

The equilibrium concentration of thoron above the thorium nitrate in the bottle was measured with the LUK 3A Plch device using the RnTh+ mode. Using this mode, the device can measure both the radon and thoron concentrations when their values are higher than 1 kBq/m^3 [15]. In this particular case, the Janet syringe (part of the LUK 3A Plch device) was used to extract a precise quantity of air out of the plastic bottle. The extraction was done using this syringe (150 cm^3) fitted with a very thin medical needle and pricking the cork and lateral sides of the bottle. The average of five extractions leads to a value of 1210 ± 85 Bq. The same equilibrium quantity is found in the canister in Fig. 2, where the thoron concentration in volume V_1 is $A_0 = 6.0$ MBq/m^3. This value will be used to determine the implantation factor under equilibrium conditions. Considering the lifetime of the descendant with the longest life ^{212}Pb ($T_{1/2} = 10.65$ h), the implantation equilibrium could be reached in two days (48 h). Calculations show that after this time interval 95.6% of the equilibrium is reached. The samples exposed to implantation (glass, paper, polyethylene, aluminum, etc.) are squares of 1–2 mm thickness and 1 cm^2 area, placed in the thoron exposure vessel as shown in Fig. 2, above the plastic sieve. In each case, the implantation was measured on the upper side face. Before measuring the implanted activities, the samples were cleaned by wiping with a soft material and ethanol.

4. Results and discussions

Fig. 3 shows the alpha spectrum obtained for the glass sample. The spectrum peaks are only those at the energy of 6.05 and 8.78 MeV. The first peak is obtained from alpha disintegration of ^{212}Bi (36%) and the second from ^{212}Po disintegration generated by the beta disintegration of ^{212}Bi (64%). The 6.5, 7.5 and 8.5 MeV peaks from this spectrum are peaks identified to be at these energies by the standard pulse generator of the spectrometer, in order to be used at energy calibration. If in integral

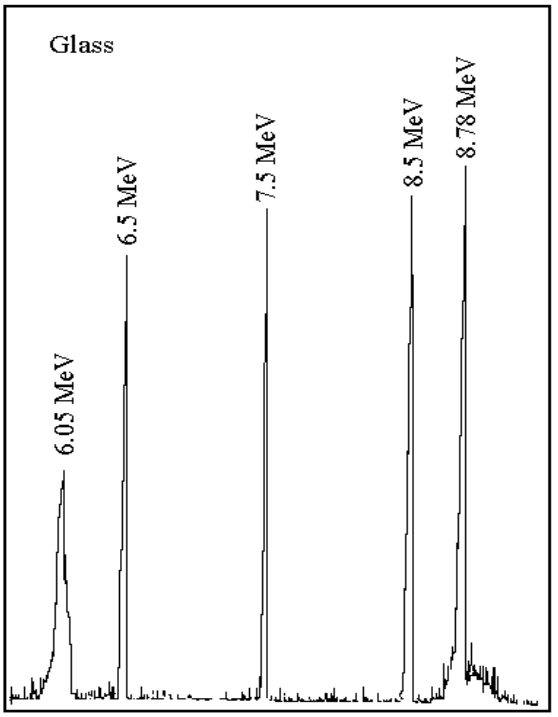

Fig. 3. The alpha spectrum obtained for the glass sample.

mode the energy threshold is used for the energies $E_1 \geqslant 5$ MeV (N_1) and $E_2 \geqslant 7$ MeV (N_2), the areas of these two peaks can be obtained.

$$A_1 = N_1 - N_2, \quad A_2 = N_2. \tag{2}$$

One can also calculate the alpha weighting factors at the ^{212}Bi disintegration:

$$g(6.05\ \text{MeV}) = A_1/(A_1 + A_2) = (N_1 - N_2)/N_1,$$
$$g(8.78\ \text{MeV}) = A_2/(A_1 + A_2) = N_2/N_1. \tag{3}$$

As this spectrum shows, the peaks are relatively narrow, especially the 8.78 MeV peak. The width of the peaks depends much on the degree of the surface roughness as is presented in Fig. 4, where the alpha spectrum for a Cu sample is shown. This sample was previously chemically cleaned using a HNO_3 solution.

The width of the lines is in this case four times larger than that in the case of the glass sample. For a mechanically polished Cu sample the lines are thinner. A similar spectrum to that of the glass

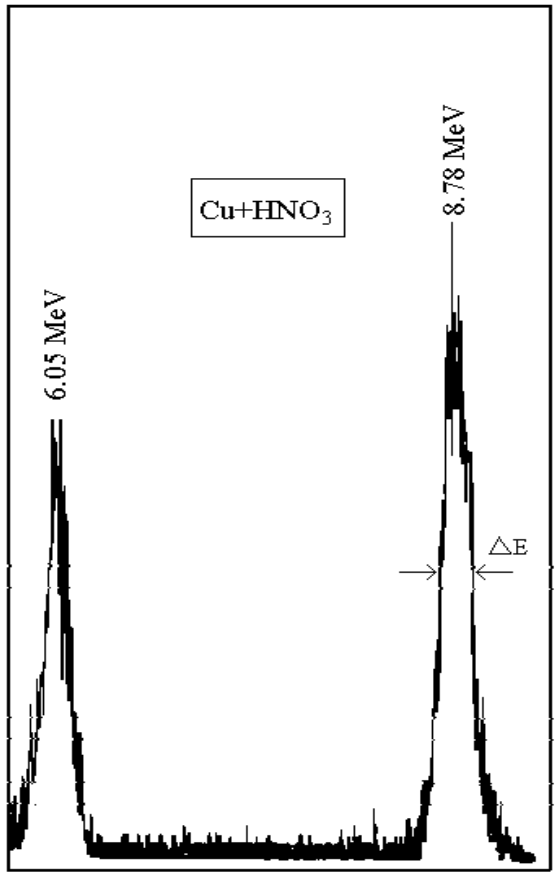

Fig. 4. Alpha spectrum of a copper sample cleaned with HNO_3.

sample was obtained in the case of polyethylene, the widths of the lines being very alike, Fig. 5 and Table 1.

Table 1 presents the implantation data obtained from eight types of samples. As one can see from this table, the weighting factor (64%) for beta decay of ^{212}Bi was found. The average value for these results is $63.25 \pm 0.95\%$ obtained from the third column of this table. The width ΔE of the line depends on the nature and degree of the material processing. The narrowest lines were recorded when working with glass and plexiglass, these materials having a very glossy surface. Taking for glass and plexiglass samples an average width of 50 keV for the 8.78 MeV peak (Table 1) and considering that the alpha detector protective cover introduces a supplementary width of the

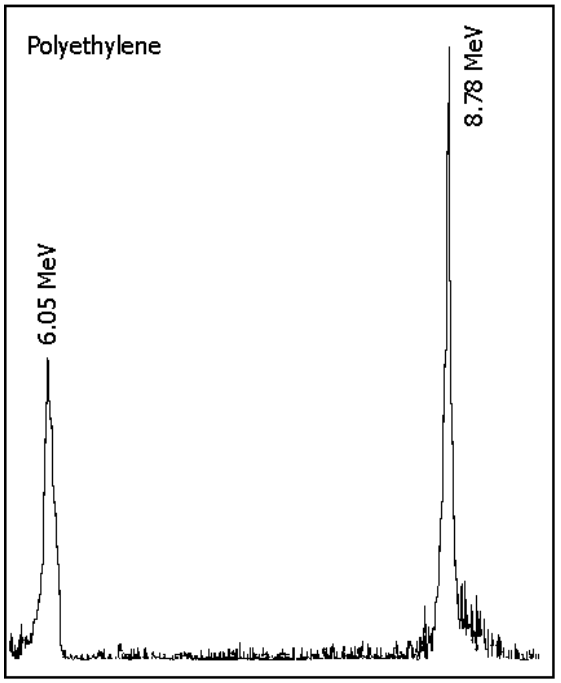

Fig. 5. Alpha spectrum for a polyethylene sample.

line, from 25 to 35 keV, we can calculate that the excess width of 15 keV is the result of a depth implantation of about 100 nm for ^{212}Pb. Compared with other depth implantation data this value is almost the same [16].

The large width (221 keV) for the Cu sample treated with HNO_3 suggests that the implantation of thoron and radon progeny could be used in the study of surface roughness. When the sample was mechanically polished, it gives approximately only half of the width of the chemically treated sample. A similar value can be also found for the Al sample. Another remark refers to glossy paper, with intermediary values of the line width. The last but one column in Table 1 represents the implantation factor at equilibrium (f_i), obtained in these experiments. This factor is defined as the implanted activity on the material surface (Bq/m^2) divided by the thoron concentration in the chamber (Bq/m^3). Higher values of the implantation factor are obtained in the case of glass and paper these being over six times higher as in the case of Al sample. The increasing of the surface roughness leads to increase of the implantation factor as shown by the comparison of rows 5 and 6 in Table 1, respectively, for Cu samples mechanically and chemically cleaned. Comparing the value found for glass in the case of equilibrium for thoron (^{212}Pb), 52 (Bq/m^2)/(kBq/m^3) those found experimentally for radon (^{210}Pb), about 25 (Bq/m^2)/(kBq/m^3) [17], a significant difference is observed. This may be explained by important losses due to glass corrosion and diffusion of ^{210}Pb from the glass surface as remarked in the works [9,18]. For the glass sample, the exposure was repeated placing the sample at four different heights: 2, 6, 10 and 16 mm above the thorium level in the box. The implantation result is observed in Fig. 6. The quantitative values of implanted ^{212}Pb decrease. This is similar to the gradient of thoron and its progeny one may observe as one moves away

Table 1
Some characteristics of thoron progeny implanted in different materials

Nr	Sample material	g (8.78 MeV) (%)	ΔE (keV)	N_1 (c/100 s)	f_i (Bq/m^2:Bq/m^3)	Obs.
1	Glass	63.7	55	1080	0.052	Normal
2	Polyethylene	61.8	64	672	0.032	
3	Paper	62.6	89	1010	0.048	Glossy
4	Plexiglas	65.1	46	355	0.017	
5	Cu	63.3	137	620	0.030	Mechanically polished
6	Cu + HNO_3	63.2	221	862	0.040	Chemically cleaned
7	Al	63.5	146	175	0.008	Metallic foil
8	PdAg	62.8	74	340	0.015	Alloy

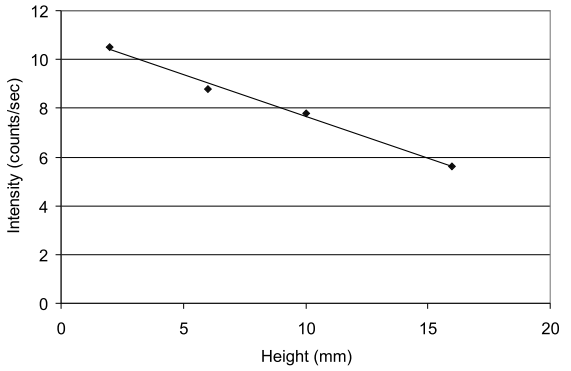

Fig. 6. Influence of the sample position in the thoron chamber on implanted ^{212}Pb.

from the walls of a room which is producing thoron.

In addition, another experiment was made for the glass sample, consisting of five consecutive measurements as follows. The first measurement was made after the sample was removed from the container, without touching the implanted surface (a). After this, the sample was carefully cleaned with a soft material and re-measured (b). The sample was again removed from the spectrometer chamber and cleaned with ethanol solution 95%, then re-measured (c). The operation (c) was repeated (d). Finally the sample was measured again the next day, after 19 h (e). The results are shown in Table 2. In each case, three measurements of 100 s were made over the whole spectrum. The first four operations (a)–(d) took 0.5 h total. In the first case (a) there is a great contribution of the aerosols gathered on a surface, which can be easily removed (b). A cleaning operation with ethanol (c) removes a part of the ^{212}Pb and ^{212}Bi atoms implanted even on the surface, but by repeating this operation the quantity implanted is very little influenced (d). Calculating the lifetime of the implanted progeny, from the values (d) and (e) a value of $T_{1/2} = 10.55$ h is obtained, very close to the value of the lifetime constant of ^{212}Pb.

All our samples in Table 1 underwent the operations (a)–(c) before being measured. If we look at Fig. 7 and we compare the implantation schema from this figure with those of the radon progeny implantation [9] one can see that in the thoron case this mechanism is much simpler. Only the alpha decay of ^{216}Po can cause implantation in the case of thoron progeny whereas in the case of radon progeny there are two possibilities of implantation namely ^{218}Po and ^{214}Po recoil.

Also in the case of thoron progeny the attachment to aerosols may probably be neglected due to the short age of ^{216}Po (0.15 s). Porstendorfer and Reineking [11] showed that the time of attachment is much longer of about 2–100 s. The coefficients in Fig. 7 are of the same significance as in the case of radon progeny implantation [9]. The deposited part of thoron progeny is also possible to be studied because the cleaning can be done at different times after the sample is extracted from the container and after different cleaning operations.

5. Conclusion

The measured alpha spectra prove that the implantation of thoron progeny can take place in various materials. The implantation efficiency depends strongly on the material type and surface properties. Between the implantation efficiency and the roughness of surface there is a direct relation. In the case of a copper surface cleaned with HNO_3 solution before thoron exposure it was found that the peak width was doubled in size compared with the same sample mechanically polished. Therefore, the implantation ^{220}Rn prog-

Table 2
The influence of surface cleaning on the implantation parameters

Sample	(a) Untreated	(b) Soft cleaned	(c) Ethanol 1	(d) Ethanol 2	(e) Re-measured
Delay time (min/h)	0	10 min	20 min	30 min	19 h
Intensity (counts/s)	28.40	14.60	9.82	9.65	2.77

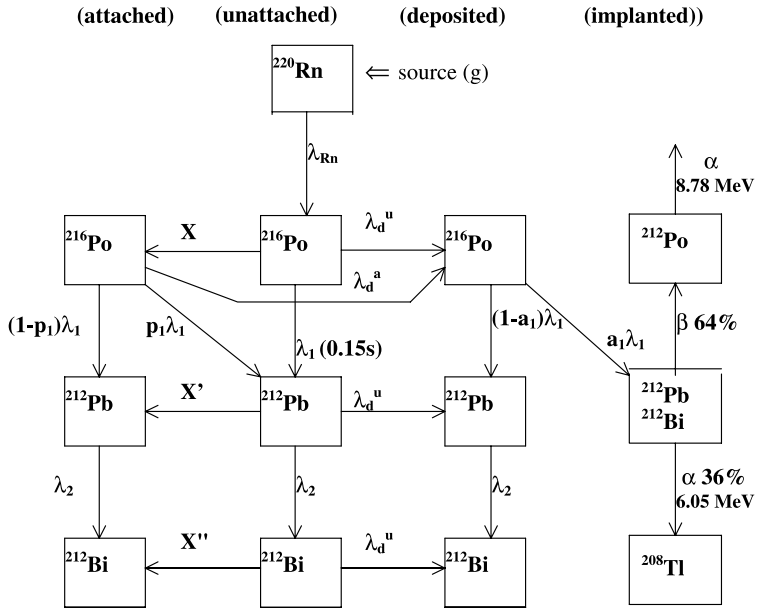

Fig. 7. The schematic decay of thoron as a daughter product from a thoron source (g). λ_{Rn}, λ_1 and λ_2 are decay constants of ^{220}Rn, ^{216}Pb and ^{212}Pb; X, X′ and X″ – attachment rates to aerosols; λ_d^u – deposition velocity of unattached ^{216}Pb; λ_d^a – deposition velocity of attached ^{216}Po; P_1 – recoil probability from aerosol for ^{212}Pb; a_1 – implantation probability of ^{212}Pb.

eny could be used in the studies of different surfaces. Because the implantation mechanism is much simpler in the case of thoron daughters these kinds of experiments are adequate for the simulation of the radon progeny implantation and therefore will assist in obtaining realistic values for the parameters used in the Jacobi model.

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