Angular variation of the energy straggling in alpha particles emitting sources*

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The present paper presents firstly a review of the fundamental results and conclusions obtained through alpha spectrometry, and the influence of energy straggling, energy loss and self-absorption, on the determination of the energy of alpha samples. Moreover, measurements made by us, with a silicon surface barrier detector, show that the peak-asymmetry and peak-shift of an alpha spectrum increases with the angle of emission, and that the magnitude of this effect depends on the thickness and homogeneity of the sample, as well as on the geometry of the measuring system. Data analysis let us to conclude that, in general, there is a larger precision in measurements made under very small solid angles around the normal to the sample, and with the necessary cases required on the production of the source and on the set up of the measuring system.

O trabalho apresenta inicialmente uma revisão dos resultados experimentais e conclusões fundamentais que demonstram a influência do *straggling* em energia, poder de freamento e auto-absorção, na espectrometria de amostras alfa-emissoras. Medidas feitas por nós, com um detetor de barreira de superfície de silício, mostram que a assimetria e o deslocamento dos picos num espectro-alfa aumentam com o ângulo de emissão da partícula. A magnitude de tal efeito depende da espessura e da homogeneidade da amostra, bem como da geometria do sistema de medição. Fica evidente que existe maior precisão em medidas feitas em pequenos ângulos sólidos em torno da normal à superfície da fonte, em que se tomaram os necessários cuidados com a preparação da amostra e com o arranjo do equipamento de medidas. Por outro lado, a técnica pode ser aplicada na medida do *stopping power* e do *straggling* em energia, como função da espessura atravessada pela partícula no material da fonte.

I. Introduction

The energy measurement of alpha particles from radioactive sources, made with conventional detectors, presents several difficulties, specially when one wishes very accurate results. The main problems arise basically from two facts:

- The average range of alpha particles in the source material is about 10 μm. Thus, even making use of extremely thin samples, produced through techniques such as vacuum evaporation, electrodeposition, etc., there is always some energy degradation of the emitted particles when they emerge from the source Moreover, owing to the statistical factors involved in its interaction with matter, the energy straggling may increase depending on the distance passed through the sample.

- On the other hand, Rutherford scattering, both in the source and in its holding, can change the angular distribution of the emission of particles.

A number of research workers have carried out researches which contribute to elucidate the matter[1−26]. Bohr[1,2] shows that when charged heavy ions pass through a certain absorbing material, they lose their energy through several independent electrostatic interactions either with electrons bound to environmental atoms or with environmental atomic nuclei.

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Due to the statistical nature of the energy loss process, the initially monoenergetic energy distribution of an incident beam of particles doesn't remain defined after the projectiles have passed through a given thickness of the absorber.

The resulting variations in the energy loss are known generally as "energy straggling". Lewis[3] has shown that this process is closely connected to the "range straggling". According to Bohr[2], heavy ions crossing thick layers of a certain material, would produce a gaussian energy distribution with an additional low energy "tail", owing to a few heavy collisions with nuclei. With the increase of the energy loss, the distribution approaches more and more a gaussian curve. With regard to such theory[1], the peak full width at hall maximum η (FWHM) of the gaussian portion of the respective energy spectrum can be determined by:

$$\eta^2 = (8.\ln 2) \cdot 4\pi \cdot z^2 \cdot e^4 \cdot N.Z.(\Delta X) \ , \qquad (1)$$

where $z.e$ is the incident ion charge, Z is the atomic number and ΔX the thickness of the crossed absorber, which contains N atoms/cm^3.

Changes to the Bohr theory, due to Bethe-Livingston[4], Titeica[5], Landau[6], Tschalär[7], Payne[8] and others, were applied to the energy loss of alpla particles with good agreement with the experiments. To Lewis, in the beggining of the path of a monoenergetic beam, when the energy loss is still small and particles energy is almost unchanged, the shape of the energy distribution is asymmetric, with a "tail" due to single collisions with significant energy losses. Landau predicted this asymmetry to fast heavy particles crossing thin absorber layers, and his theory had shown very good agreement with experimental results.

We shall restrict our discussion to the experimental results of alpha-spectrometry developed with silicon surface barrier detectors (SSB), associated to conventional electronics.

The energy straggling of alpha particles from a specific source can be experimentally determined from the measurements of η_T (FWHM, keV) of the obtained energy distribution. η_T results from the contribution of several factors which produce the peak broadening[9]: the electronic noise of the amplifier-detector system (η_E), the straggling produced by the detector window (Al or Au) and dead layer (η_D), the statistical changes in the energy loss process in the detector (η) the broadening caused by the source thickness (or by the features of the beam producing accelerator) (η_S), the eventual scattering on the measuring chamber walls and collimators (η_c) or in gases (or other absorbers) placed between the sample and the detector (η_A). The relationship among such factors and the distribution full width can be given by:

$$\eta_T^2 = \eta_D^2 + \eta_E^2 + \eta^2 + \eta_S^2 + \eta_C^2 + \eta_A^2 \qquad (2)$$

The η, η_A and η_S contribution to the total straggling was studied by different research workers. Al-Bedri, Harris and Sykes[10], among others, measured the energy-straggling of alpha-particles which passed through many *gases and gas mixtures* (η_A), using an ORTEC SSB detector, and they found reasonable agreement with the prediction of Bohr's theory to high energies and reduced thicknesses (namely, *small losses)*, although at the end of the path the straggling is smaller, because of the reduction of the beam effective charge due to the capture and loss of electrons. Comfort et al.[11] measured the energy loss and straggling of alpha particles in Al, Ni, Ag and Au, as a function of the metal thickness and of the energy, with the help of SSB detectors. The obtained spectra show a shift towards the zero energy and progressive broadening of the distribution, as a function of the energy loss increase, making the initial asymmetry predicted by Bohr[2] and Landau[6] also evident. One can verify that the data concerning the straggling η_A usually stay between the curves foreseen by Bohr and Titeica[5] but, they approach Bohr's curves in the case of small absorber thickness. Several other, researchers[9,12–14] have found similar results, both with gaseous and solid absorbers.

On the other hand, other works have studied the energy straggling produced in alpha particles beams by the crossed source material itself (η_S) and they have concluded that it is critically dependent on the chemical separation procedure, the technique of production, as well as the thickness of the source. White[15], working, with a 2n geometry gridded ionization chamber, have obtained energy spectra from U_3^{238} O_8 sources, with thicknesses of 0.02 mg/cm^2 and 0.50 mg/cm^2 and from natural uranium with thickness bigger than the alpha particles range. He could observe the growth of the peak width, especially in the low energy side, with the thickness of the sample, owing to the increase of the fraction of alpha particles which lose energy in the sample material. A similar effect was observed by Sill[16] as he studied the radiochemical separation of transuranium elements using sulfate of barium. Making use of a SSB detector and Am^{241} samples electrodeposited on polished platinum, (a) after the separation in $BaSO_4$ and (b) when homogenously distributed in $BaSO_4$ with 0.075 mg/cm^2 (c) and 0.75 mg/cm^2 thickness, Sill could get spectra with 24, 75 and 325 keV of FWHM, respectively. This study makes it clear among other things, the influence of the chemical compounds that accompany the source atoms in the spectrum resolution. Now Nitzki and Matzke[17], working with surface barrier detectors, have observed both the peak broadening and the progressive shift to lower energies of a multienergetic source of Th^{220} progeny, while successive layers of UO_2 or ThO_2 were evaporated on the sample. Regarding to the results, the energy straggling η_S varies according to the absorber thickness, in good agreement with the Bohr[1] and Bethe- Livingston[4] expressions.

Concerning the technique of production of the sources, we can mention Aggarwal's work[18], which presents a comparative study among alpha-spectrometry plutonium sources, produced by (a) direct evaporation of the Pu purified solution, (b) direct evaporation of the aqueous solution with the use of TEG (glycol tetraethilene) as spreading agent, (c) direct evaporation of an organic solution after the use of TTA (tenoil-trifluoro-cetone) for plutonium extraction and (d) electrodeposition. Alpha-spectra of such samples were obtained, using an ORTEC SSB detector, in identical conditions, and the energy resolutions (FWHM) obtained, for the Pu^{238} (5.50 MeV) and Pu^{239} + Pu^{240} (5.16 MeV) peaks, had, respectively, the following values: (a) between 80 and 90 keV; (b) between 40 and 60 keV; (c) between 30 and 60 keV; (d) between 30 and 50 keV. The sources prepared with TEG or from a solution with TTA were verified to show peak resolutions comparable to the electrodeposited sources, and those prepared by evaporation without the use of a spreading agent are, generally speaking, thicker and not uniform, as shown by their FWHM.

The present work has as for objective, among others[19], to investigate the fundamental influence of the sample thickness on the energy of the α-particles emitted at different angles, from sources produced through direct evaporation and electrodeposition.

II. Equipment utilized for the measurements

The detection system used for tho measurements presented in this paper is an alpha-particle spectrometer which makes use of an ORTEC Si surface barrier detector, with active area of 150 mm^2 and Al window with 50.1 μ/cm^2 [19].

II.1. The measuring chamber and the vacuum system

The vaccum chamber that housed the detector (Fig. 1), made of brass and with 12.04 cm of internal diameter allowed to change and externally measure the source to detector distance (d), as well as the θ angle, between the normal to the sample plane and the detector axis. The scale of the goniometer permitted θ to be read with about 1° of precision; the reference position $\theta = 0°$ was adjusted with the help of a bubble level. The source holder consisted of a ring, with internal diameter 31.89 ± 0.04 mm and side height 3.66 ± 0.06 mm, to which the source was fixed through its support. "O-rings" were utilized to block the system to air entry.

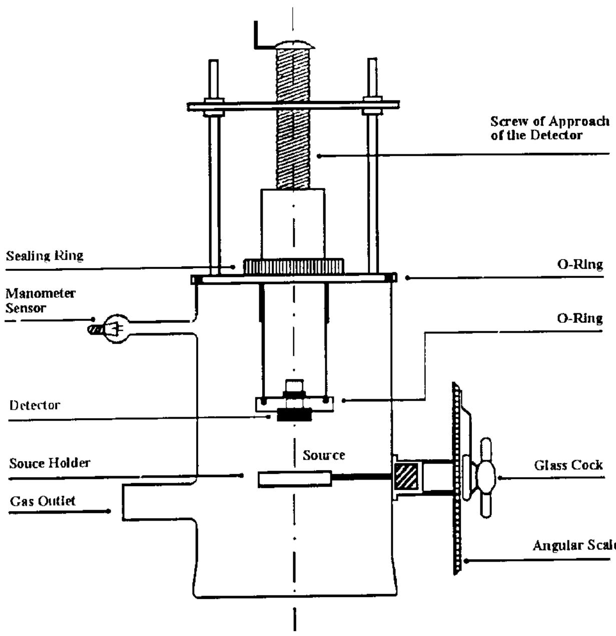

Figure 1: Vacuum chamber with detector.

Vacuum was produced by a rotative pump connected to the chamber. The chamber internal pressure was kept about 10^{-1} mmHg and a Pirani-type manometer was built and utilized for its measurement, with a circuit that consisted basically of a Wheatstone bridge made out of three equal lamps and a fine adjust-helipot. One of the lamps, used as a sensor of the manometer, was drilled and adapted to the vacuum chamber (Fig. 1). Such measurer was previously calibrated by comparison to a VARIAN NRC 836 manometer.

II.2 The amplification and signal analysis system

Three series of measurements are presented here, two of them were made using an amplification system which consists of two stages based on the CA3140 integrated circuit and of a third one with the LM301 circuit. The third series of measurements was made with another device based on the CA3140 and LM301 integrated circuits and the FET 2N3819, which presented bigger noise. In both schemes, the first stage functioned as a charge sensitive pre-amplifier. Linearity was verified in all cases and considered satisfactory[19].

Spectra were obtained with the help of an ORTEC multichannel analyzed model 6240, with 1024 channels.

III. Accomplished measurements and obtained results

We carried out measurements which consisted of:

(a) the measurement of alpha-spectra with an Am^{241} source directly evaporated on aluminized mylar, the first series series carried out with $d_1 = 27.00 \pm 0.10$ mm and the second one with $d_2 = 50.00 \pm 0.12$ mm, both for several emission angles θ, from 0° to 70°.

(b) the measurement of a third series of alpha-spectra with the same source, utilizing an *Al* collimator 24.4 mg/cm^2 thick (bigger and bigger than the range of the Am^{241} α-particles in the *Al*, around 9.6 mg/cm^2)[24] and with a hole of 2.78 ± 0.06 mm in diameter, fitted to the detector window, distant $d_3 = 27.42 \pm 0.13$ mm from the source, also for $0° \leq \theta \leq 70°$.

(c) the measurement of alpha-spectra with an Am^{241} source electrodeposited on stainless-steel, utilizing the same collimator, distant $d_4 = 19.75 \pm 0.14$ mm from the source, for emission angles from 0° to 40°.

The Am^{241} emits α-particles of 5.4857, MeV in 85.2 % of the cases[20].

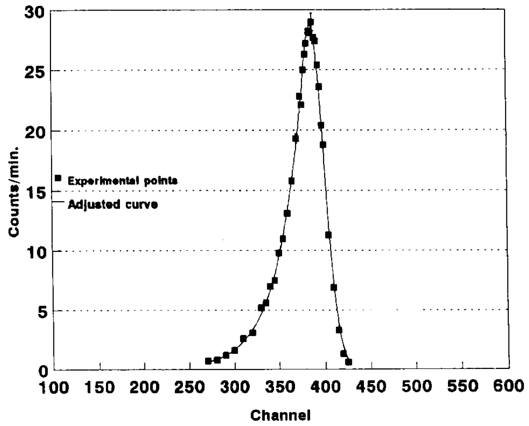

Figure 2: Alpha-spectrum of evaporated Am source obtained with the system, for $\theta = 30°$. Curve was fitted by the least-squares method. Error bars represent examples of the standard deviations of count rate.

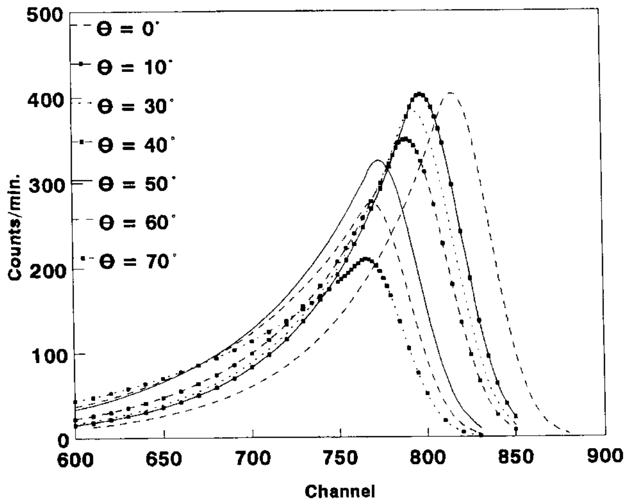

Figure 3: Alpha-spectra for several θ angles (*first series*), from evaporated Am^{241} source ($d_1 = 27.00$ mm), showing only the adjusted lines for better view.

Fig. 2 shows the experimental points of one of the obtained spectra (d_3, $\theta = 30°$); the solid line is a least-squares fit to the experimental data which consists of a gaussian modified in the left by an exponential[21]. Fig. 3 shows some of the adjusted curves obtained for the measurements made for $d_1 = 27.00$ mm and Fig. 4 shows the curves for the spectra obtained for $d_2 = 50.00$ mm.

The third series of α-spectra was obtained with the same source, and with the referred collimator. Fig. 7 shows the curves adjusted to the obtained spectra for some measured angles.

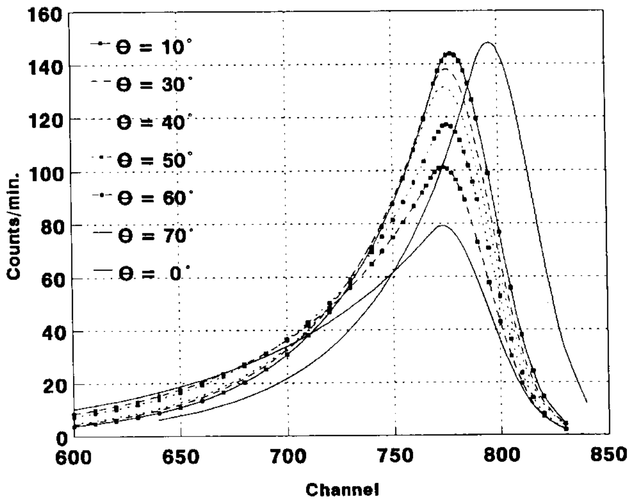

Figure 4: Alpha-spectra for several θ angles (*second series*), from evaporated Am^{241} source (d_2 = 50.00 mm), showing only the adjusted curves.

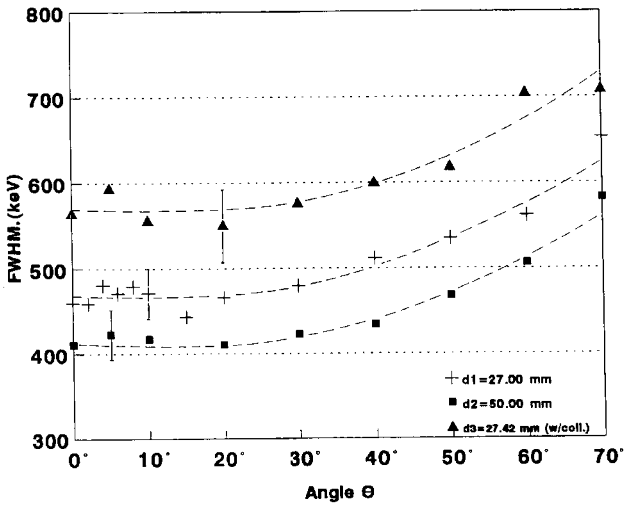

Figure 5: Resolutions of alpha-spectra (evaporated Am^{241} source). Bars represent examples of FWHM standard deviations calculated from the fitting process.

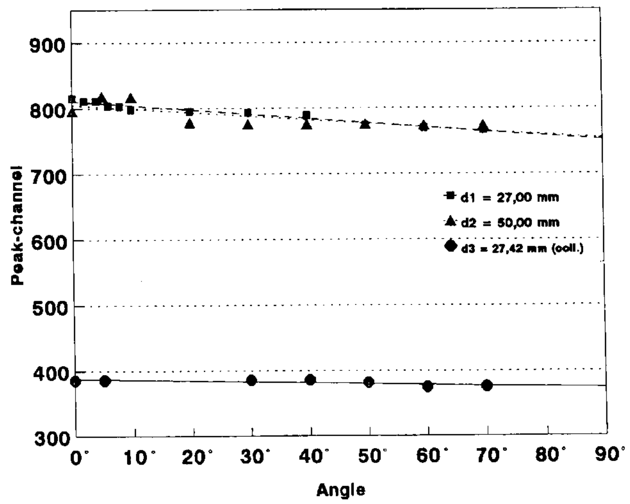

Figure 6: Shift of the peak-maximum of alpha-spectra for several θ angles (Am^{241} evaporated on mylar). FWHM standard deviations, calculated from the fitting process, are too small in the scale.

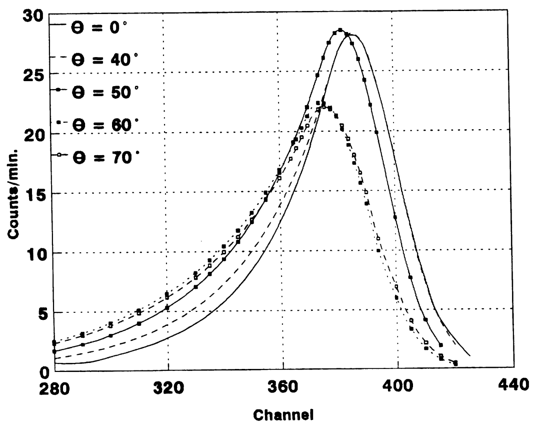

Figure 7: Alpha-spectra for several θ angles (*third series*), from evaporated Am^{241} source (d_3 = 27.42 mm), showing only the adjusted curves.

Finally, we have also measured, by comparison, the alpha-spectra using an Am^{241} sample electrodeposited on stainless steel. The electronics were the same utilized at the third series (see section II.1). The obtained spectra, in this case, showed almost symmetrical - half-width at half maximum (HWHM) at right and at left sides are roughly the same -, and without significant change on the FWHM of the peak at the measurement angular range, within the limits of precision (Fig. 8). Moreover, this width was at least 37% smaller than that achieved for the evaporated source, at a bigger distance.

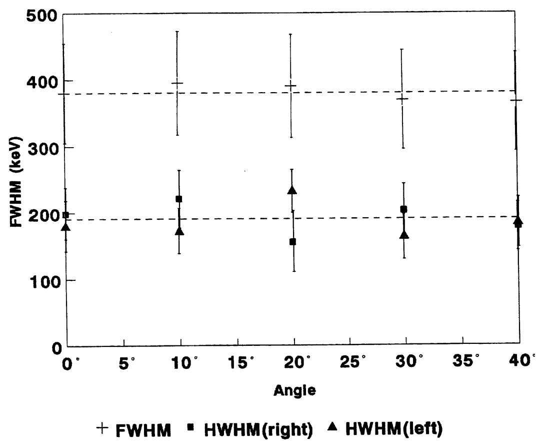

Figure 8: Resolutions of the spectra for several θ angles from electrodeposited Am^{241} sample (d_4 = 19.75 mm). HWHM is the half width at half of maximum, measured at right (HWHM right) or at left (HWHM left) of the peak. Bars represent HWHM standard deviations calculated from the fitting process.

Results Analysis

We can verify here the effects described before such as the peak shift to lower energies, the progressive broadening of the spectra and the increase of the lower energies "tail" with the angle θ, in qualitative agreement with Lewis and Landau's theories.

Figs. 5 and 6 present, respectively, the change of the spectra width η_T (FWHM, keV) and of the peak-maximum channel with θ, for the evaporated source. Although the quite big electronic noise is included in η_T ($\sim$ 300 keV for the first two series and 450 keV for the third one), we must notice that it was kept stable through all these series of measurements at the various emission angles. We can also observe, in Figs. 3 and 4, that the peak asymmetry grows with θ, making evident the increase, with the emergence angle, of the fraction of particles which bears progressive degradation of their energy and which are counted in smaller channels, due to the ionizations and nuclear collisions caused in their trajectories in the source material, whose extension also increases with θ.

In fact, in small diameter-sources, with thickness t, alpha-particles emitted from an deepness χ, with regard to the surface of the source, and detected at emergence angle θ, will travel in its inner volume, at least, an average distance $d_s = \chi/cos\theta$. As the d_s value increases with θ, the maximum energy the α-particles emerge is reduced, which causes the peak shift observed at the spectra. In the worst case, when $\chi = t$ the minimum thickness crossed by a particle emitted from the bottom of the sample might change from t to 2.92t, between 0° and 70°, which is similar to the making of frontal measures with sources of increasing thickness. On account of this the agreement with the results quoted at item I[15–18].

On the other hand, it is to be noted that the spectra width η_T, when d = 50.00 mm, is systematically lower than the peak broadening when d = 27.00 mm, for all values of θ. We can understand such fact taking into account that, according to the equation (2), in the absence of collimators and in vacuum, we have:

$$\eta_T^2 = \eta_D^2 + \eta_E^2 + \eta^2 + \eta_S^2 \qquad (3)$$

At a distance d = 50.00 mm, the measurement angular incertainty $\Delta\theta$ falls to 8° and this makes the tracks of the alpha particles which reach the detector active area more uniform in length, both in the source material and in the detector dead layer as well as in the silicon active volume, reducing the total energy straggling.

Moreover, when one takes into account the measurements made in several angles θ, for the same distance between source and detector and same series, the influence of η_E (noise of the electronic system), η_D and η shouldn't change significantly, so that only the η_S change contributes to the increase of the peak full width. If we admit η_E symmetrical, then the assymetry of the distribution is due to the particles which suffer single heavy collisions, mainly in the source material, and are counted in lower channels. Thus, if η_T and η_G represent, respectively, the measured FWHM and the FWHM of the gaussian portion of the spectra (twice the HWHM of the higher energy side of the peak), then $\eta_T - \eta_G$ will be a measurement of the peak assymetry at half of maximum.

Fig. 9 shows ($\eta_T - \eta_G$) as a function of 1/cos θ. The good linear fit is in agreement with the Lewis' hypothesis that the spectra assymetry is caused by *single* electrostatic interactions with sample nuclei, which number must increase linearly with the particles mean path lenght $t/cos\theta$.

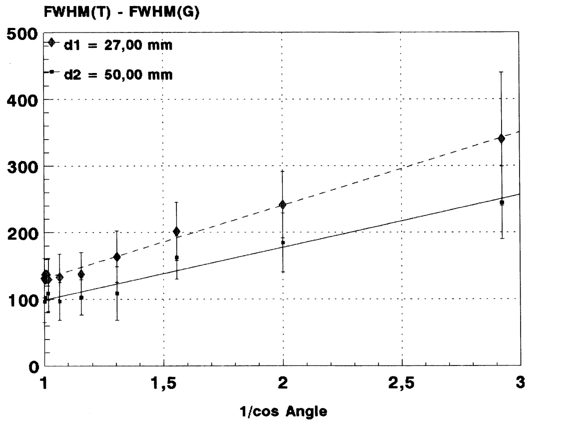

Figure 9: Change of η_T (FWHM(T)) - η_G(FWHM(G)) (see text) with $1/cos\theta$ for Am^{241} evaporated on mylar *(first and second series)*. Curve is a linear fitting of each set of data. Errors were obtained by propagation of deviations calculated from spectra fitting.

Concerning to the spectra obtained with the electrodeposited source, the smaller value of η_T indicates that the layer thickness and non-uniformities should be much lower than that of the evaporated sample, which was later confirmed by the measurements of the absolute activity of the sources ($(1.31 \pm 0.04) \cdot 10^{-3}$ μg/cm^2, for the electrodeposited source and (10 ± 4) μg/cm^2 for the evaporated one[19]). Thus, the particles energy loss in the source material must have been very little, and there may have been a few nuclear collisions and ionizations in the source, so that the spectrum low energies tail was kept inside the enlargement due to the electronic noise. Such comparative results agree with those of Aggarwall[18] and Vitale[25].

According to all the quoted results, in order to determine the FWHM, the corrections of the energy of the most frequent α-particles (5.4857 MeV) in relation to the energy losses in the detector Al window and dead layer (55 keV)[24], and to the nuclear collisions in the detector bulk (14 keV)[26] were made, despising the losses in the source material itself.

The results presented at this paper, for an Am^{241} evaporated source, are also very similar to those obtained by Budtz-Jørgensen and Knitter[22] with vacuum evaporated UF_4 deposits of several thicknesses, utilizing a Frisch-grid ionization chamber. In such work, as well as in the one described here, the chosen angular range ($cos\theta \geq 0.2$ and $\theta \leq 70^0$, respectively) guarantees that the results are free from backscattering and self-absorption interference[23].

V. Conclusions

From the results, we can conclude that:

a) The energy spectrum of alpha-particles emitted by 10 μg/cm^2 sources evaporated directly on mylar shows a low energy "tail" increasing together with the emission angle, with progressive deterioration of the resolution (a growth of up to 40% of the peak FWHM from 0 to 70°), in the present case, may occur), due to the increasing energy dispersion which the particles emerge from the source. This occurs due to the increase of the particles path in the source, as the emission direction deviates from the normal one. Regarding to electrodeposited sources, in which the thickness is usually smaller ($< 0.01\mu$g/cm^2, in the present case) this effect is less perceptible, and it may become "masked" by the enlargement produced by the electronic noise of the system.

b) It is to be noted that, for bigger distances between source and detector, in low geometry measurements, the spectrum FWHM diminishes, as a consequence of the reduction of η_D, η and η_S.

c) The alpha-spectrum peak (or peaks) of such sources shifts to lower energies as θ increases, due to the particles energy loss which increases as the path inside the sample becomes longer. Such fact may cause errors when identifying the nuclide in mixed samples, as well as inaccuracy when determining the alpha-particles energy, in non-frontal measurements.

Therefore, in the alpha-particles-spectrometry with SSB detectors, we can see that accuracy is greater in measurements made in the surroundings of the normal to the source plane. The increase with θ of the imprecision of the energy measurements depends fundamentally on the source preparation technique, and it can be small in thin samples, such as those produced by electrodeposition, or significant in thicker and less uniform deposits, such as those ones produced by direct evaporation, without the use of a spreading agent.

Finally, it is possible to analyze the applications of the described method, which open the possibility of making α-particles energy loss and energy straggling measurements in radioactive isotopes as a function of the material thickness, by measuring the same source in different emission angles, with no need of producing samples of increasing thicknesses, which are difficult to be evaluated and reproduced. The agreement of the results with Landau's theory also suggests this possibility.

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