Experimental and Monte Carlo determination of mass absorption coefficients for ^{90}SR/^{90}Y beta particles in organic compounds

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Abstract
In order to investigate the β-particles absorption in organic compounds, an experimental arrangement was designed. A Monte Carlo transport code was also used to simulate this process. This allowed reporting for the first time μ (^{90}Y) values in several organic compounds for ^{90}Sr/^{90}Y isotopic sources. The obtained experimental and calculated values showed a good concordance. Methodological aspects of the μ (^{90}Y) determination concerning the range of the absorber's superficial density are discussed.

DETERMINACIÓN EXPERIMENTAL Y POR MONTE CARLO DE COEFICIENTES DE ABSORCIÓN MÁSICOS PARA PARTÍCULAS BETA DE ^{90}SR/^{90}Y EN COMPUESTOS ORGÁNICOS

Resumen
Se estudia la absorción de partículas beta en compuestos orgánicos en una instalación diseñada al efecto. El proceso de absorción es además simulado mediante el código de Monte Carlo. Se reportan, por primera vez, los coeficientes de absorción másicos para partículas beta de ^{90}Sr/^{90}Y en varios compuestos orgánicos. Se obtiene una buena correspondencia entre los resultados experimentales y los simulados. Se discuten aspectos metodológicos de la determinación del coeficiente de absorción másico, relacionado con el intervalo de densidad de absorción superficial.

Key words: *Monte Carlo method, organic compounds, adsorption, strontium 90, yttrium 90, beta particles, strontium compounds, yttrium compounds, experimental data*

Introduction

The knowledge of isotopic β-particles behavior in matter is of great importance due to its applications in nuclear and solid state physics and dosimetry studies[1]. The β-particles are continuous-energy electrons, which are emitted by radioactive decay. Over a limited penetration range, their observed transmission curves are of characteristic exponential shape (1), which implies that the attenuation of β-particles is of the same form as that of photons. When a β emitting radionuclide is counted through an absorber an exponential relationship of the type [2]:

$$I = I_0 e^{-\mu x} \quad (1)$$

is valid over a large range of absorber thickness, where I_0 - counting rate without absorber [s^{-1}], I – counting rate with absorber [s^{-1}], μ - mass absorption coefficient [$cm^2 \cdot g^{-1}$] and x - absorber thickness [$g.cm^{-2}$].

In the case of β-particles the mass absorption coefficient (μ) is sometimes, over a limited penetration range, a more useful and interesting parameter than the range for such studies. This parameter is nothing more than a mathematical craft of purely empirical origin. It measures the average number of absorbed particles that occur in a given mass per unit area thickness of material encountered [3].

There is an empirical relationship between the mass absorption coefficient, μ, and the maximum energy of the β^- emitter, E_m, of the form [2]:

$$\mu = K E_m^{-a} \quad (2)$$

K and a are constants and their values depend not only on the material of the absorber, but also on the geometry of the counting arrangement and the type of counter used.

In the literature, there are many reported values of μ for several metals (Pb, Cu, Zn, Al, etc.) [2], but there are practically none for light elements or organic compounds. Therefore, the studying of the absorption of β-particles (through μ determination) is a good first step towards the understanding of the β-particles radiation interaction with organic compounds.

The solution, using deterministic methods, to the β-particles transport problem, in an experimental array, can prove to be very complex. Therefore, the use of a computational transport code based on the Monte Carlo method[4] is a very useful tool in order to simulate the β-particles transport in the experiment.

Materials and Methods

Experimental Arrangement and Samples: The experimental arrangement shown in figure 1 was designed in order to experimentally measure and afterwards simulate (with the MCNP-4C transport code) the β^- particles attenuation in organic compounds. The detector was coupled to a NP-363 Gamma Müvek counting system.

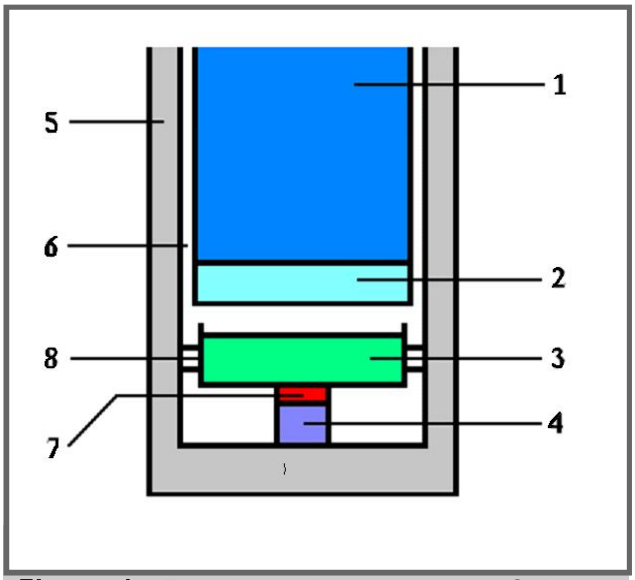

Figure 1. Experimental Arrangement Scheme: 1-Gamma Müvek ND-304 Scintillation Detector, 2-Detector's window (1 mg·cm^{-2}Al thin film + 1cm of air), 3-Sample, 4-Plastic base, 5-Lead shielding, 6-Air, 7- 5 μCi ^{90}Sr/^{90}Y Amersham source (E_m= 2.28 MeV) and 8-Cylindrical plastic container (only when the sample is liquid).

The Aluminium (E-3a Absorbers, Tracerlab. Inc.) and Carbone (nuclear reactor-grade graphite) samples were cylindrical whereas the polyvinyl acetate samples (plastic) were rectangular parallelepipeds (see table 1). Alkenes and Alcohols were purchased from Merck. The Eucalyptus essential oil was obtained in Cuba by the InSTEC. Its main components are shown in table 2.

Table 1. Chemical formulae and densities of the samples

Compound	Chemical Formula	Density ($g.cm^{-3}$)
Al	Al	2.7
Acetate	$C_4H_6O_2$	1.41
Graphite	C	1.674
Heptane	C_7H_{16}	0.68
Octane	C_8H_{18}	0.7
Decane	$C_{10}H_{22}$	0.73
Butanol	C_4H_9OH	0.809
Pentanol	$C_5H_{11}OH$	0.8085
Eucaliptus Oil	See Table 2	0.87

Table 2. Eucalyptus essential oil main components, its chemical formulae and relative percentages

Component	Chemical Formula	%
Citronelal	$C_{10}H_{15}O$	64
Citral	$C_{10}H_{15}O$	7
Citronelol	$C_{10}H_{15}OH$	10
Alfa-Terpineol	$C_{10}H_{15}OH$	2
Linalol	$C_{10}H_{15}OH$	2
Geraniol	$C_{10}H_{15}OH$	11
Cadineno	$C_{15}H_{24}$	4

Monte Carlo simulation: MCNP-4C is a computational general code that allows the simulation of the transport process for electrons, photons and neutrons using the Monte Carlo method. It is a numerical procedure that simulates sequentially probabilistic individual events starting from the statistical sampling of the probability distributions governing these events. This sampling is based on the selection of random numbers. In the particular case of the transport process for β-particles (electrons), multiple-scattering theories are used (Goudsmit-Saunderson theory for angular deflections [5], Landau theory of energy-loss fluctuations [6], Blunck-Leisegang enhancements of the Landau theory [7]). These theories rely on a variety of approximations that restrict their applicability. In particular, it is assumed that the energy loss is small compared to the kinetic energy of the electron. In order to follow an electron through a significant energy loss, it is necessary to break the electron's path into many steps (each step representing an 8.3% kinetic energy loss). These steps are chosen to be long enough to encompass many collisions (so that multiple-scattering theories are valid) but short enough that the mean energy loss in any one step is small (so that the approximations necessary for the multiple-scattering theories are satisfied). The energy loss and angular deflection of the electron during each of the steps can then be sampled from probability distributions based on the appropriate multiple-scattering theories. This subsumption of the effects of many individual collisions into single steps that are sampled probabilistically constitutes the «condensed history» Monte Carlo method. However, the representation of the electron's trajectory as the result of many small steps will be more accurate if the angular deflections are also required to be small. Therefore, MCNP further breaks the electron steps into smaller substeps. The sampling of Bremsstrahlung photons is based on the Bethe-Heitler-Born approximation results [8].

Experimental considerations were taken into account. Some constructive elements that had no implications in the transport processes (such as the lead shielding) didn't need to be modelled. The other elements of the experimental arrangement were modelled as accurately as possible regarding position and composition.

The source was modelled as a disc-shaped surface that emitted isotropically in 2p towards the absorber. The theoretical energetic spectrum of emission of a $^{90}Sr/^{90}Y$ source was calculated individually using a FORTRAN code supplied by Chang [9].

Radionuclide ^{90}Sr ($T_{1/2}$ = 28.64, E_m = 525 keV) and ^{90}Y ($T_{1/2}$ = 64.10 h, E_m = 2.28 MeV) are pure β–emitters. As observed, the ^{90}Sr half-life is much longer than the ^{90}Y half-life, establishing a secular equilibrium. Therefore in the total population of electrons, one half has the

energetic distribution of the ^{90}Sr and the other one has the distribution of the ^{90}Y. Furthermore, as the distributions are independent, the probability to find an electron in the range included between E and E+dE shall be the sum of both distributions after normalizing and is the one used in the calculi. Figure 2 shows the normalized energetic spectrum of emission probabilities for the modelled source.

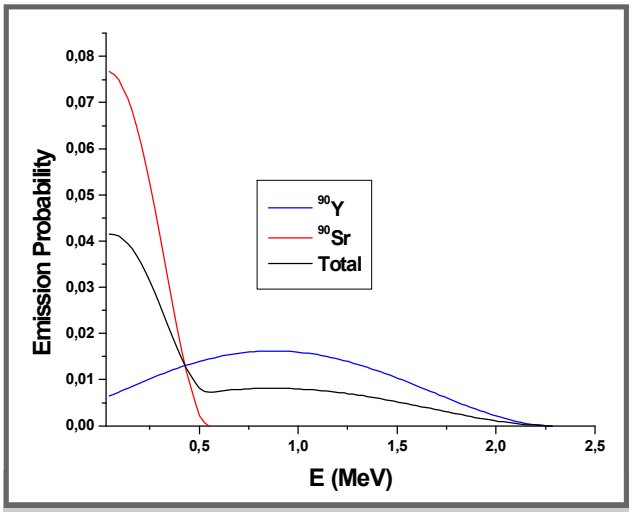

Figure 2. Energetic spectra of emission probabilities.

The MCNP transport code was set to EP (electrons + photons) mode, in order to simulate not only the electrons but also the Bremsstrahlung produced photons that could be counted by the detector. The output file (tally) used was of type 1. It corresponds to the number of electrons and photons that go through the detector's scintillator surface. As an approximation, the detection efficiency of the scintillator was set to 100%.

Results and Discussion

The results of the experimental measurements and the MCNP simulation for the Aluminium were separated in 3 zones accordingly to the absorber's superficial density: 0–70 mg.cm^{-2}, 100–550 mg.cm^{-2} and 550–950 mg.cm^{-2}. Using (1), if one plots

$-\ln\left[\left(I-I_f/I_0-I_f\right)\right]$ (i.e. μx) *versus* "x" the slope is equal to μ (cm^2·g^{-1}). Here, I_f is the background counting rate of the experimental arrangement.

In the zone of lower densities (figure 3a), the superposition of the two spectra (^{90}Sr and ^{90}Y) leads to the following well-known result: the experimentally measured and the MCNP calculated points are well above the extrapolated experimental points from the second zone (medium densities). This is explained by the fact that μ (^{90}Sr) has a much higher value than μ (^{90}Y) accordingly to (2). So, in fact, in the first zone, the obtained points are the result of the superposition of μ (^{90}Sr) and μ (^{90}Y) for Aluminium. On the contrary, in the zone of medium densities (figure 3b) the electrons from ^{90}Sr aren't energetic enough to arrive to the detector, in comparison with the ^{90}Y electrons that have no problem in getting to the detector as they are much more energetic (see figure 2). The ^{90}Sr electrons are absorbed by the thicker absorber and thus aren't counted.

The Aluminium absorption coefficient μ(^{90}Y) obtained from the second zone (of medium densities) by the experimental measurements and the MCNP simulations (table 3) agree quite well with the reported in the literature. This shows that the geometry of the experimental arrangement is adequate and that the approximations used in the MCNP simulation were valid.

Table 3. Aluminium absorption coefficient (mg.cm^{-2}) for ^{90}Y

Experiment	**5.61 ± 0.04**
MCNP	**5.63 ± 0.03**
Thontadarya [10]	**5.57 ± 0.05**

In the zone of higher densities (figure 3c) one observes that the experimentally measured and the MCNP calculated points are well above the extrapolated experimental points from the second zone (medium densities). This was expected, as (1) is only a restricted empirical relationship and it is known that for higher absorber's densities the electron's straggling path leads to non-compliance with (1). But, as we observed the results from the MCNP simulation we found out that the Bremsstrahlung photons produced by the incoming electrons were the main responsible for the increase of μ in this zone, as they were detected and counted as electrons and not photons. One can certainly not eliminate the electron's straggling path as cause for the increase of μ, but one can say with certainty that it isn't its main cause (see table 4).

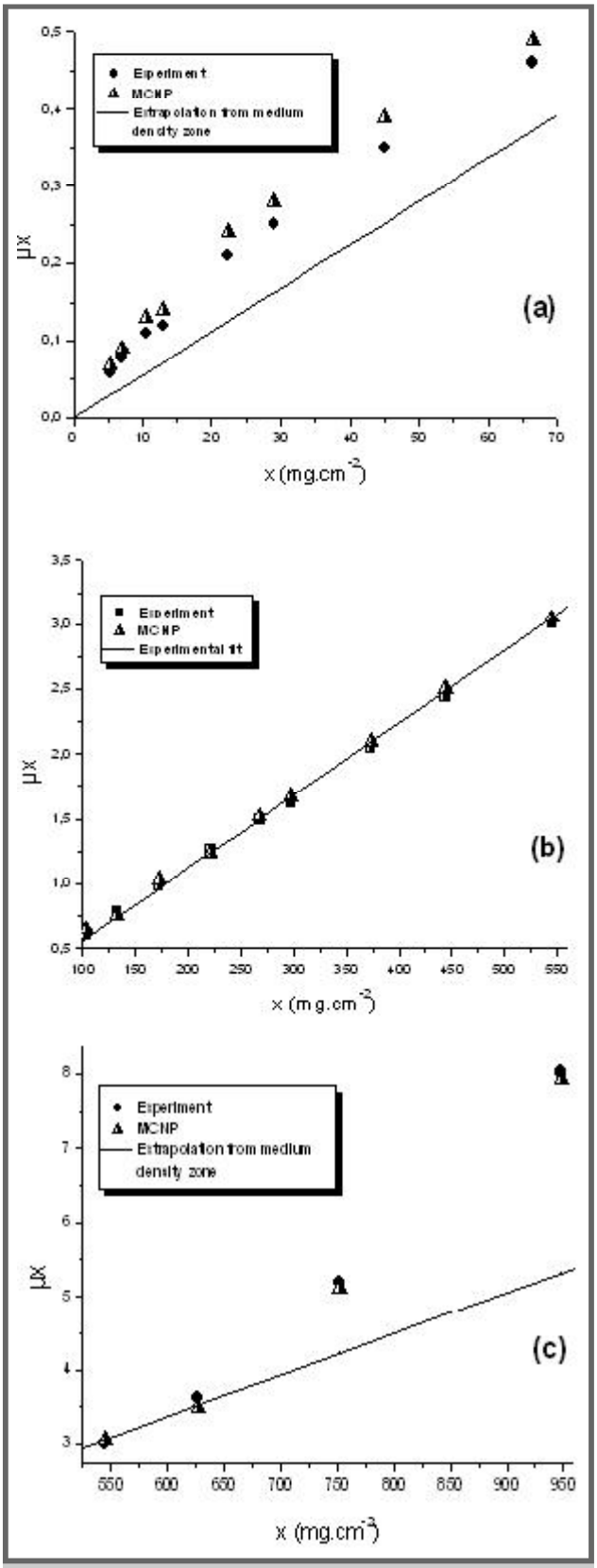

Figure 3. Aluminium μx versus x for different densities. (a): 0 -70, (b): 100 – 550 and (c): 550 – 950 mg.cm^{-2}).

Table 4. MCNP results concerning the responsibility in the increase of μ (^{90}Y) due to the electron's straggling path and to Bremsstrahlung

Superficial density (mg.cm^{-2})	Straggling (%)	Bremsstrahlung (%)
752	21	79
948	14	86

The experimental and MCNP calculated mass attenuation coefficients for the studied organic compounds are reported in table 5. Unfortunately, the comparison with previously reported values was impossible.

Table 5. Experimental and MCNP calculated absorption coefficients of several organic compounds (cm^2·g^{-1}) for ^{90}Y source and range of superficial densities involved

Compound	Experiment	MCNP	Range of superficial densities (mg.cm^{-2})
Acetate	4.87 ± 0.02	4.90 ± 0.03	100-400
Graphite	5.89 ± 0.03	5.86 ± 0.03	350-400
Butanol	5.07 ± 0.04	5.03 ± 0.03	125-375
Pentanol	4.72 ± 0.07	4.78 ± 0.03	125-375
Heptane	4.28 ± 0.05	4.33 ± 0.03	100-375
Octane	5.19 ± 0.04	5.15 ± 0.03	100-375
Decane	4.73 ± 0.04	4.76 ± 0.03	100-375
Eucaliptus Oil	3.97 ± 0.03	4.00 ± 0.03	100-300

The pattern of behavior followed by the Aluminium was also observed for the organic compounds. That is to say the superposition of the spectra (^{90}Y + ^{90}Sr) for light absorber's densities (0-100 mg.cm^{-2}), the μ measuring of ^{90}Y alone for medium densities (100-400 mg.cm^{-2}) and the non-compliance with (1) due to the presence of Bremsstrahlung photons counted as electrons and the electrons' straggling paths for such high densities (above 400 mg.cm^{-2}).

The μ (^{90}Y) values obtained from the intermediate zone (100-400 mg.cm^{-2}) by the experimental measurements and the MCNP calculations agree quite well. This shows that the geometry of the experimental arrangement is adequate and that the approximations used in the MCNP simulation were also valid for the studied organic compounds. This good concordance will allow the subsequent calculus with the MCNP transport code of absorbed doses, an important parameter when studying irradiation damages in more complex organic compounds (biological tissues, DNA, etc.).

The result concerning the Bremsstrahlung photons was obtained thanks to the MCNP simulation. This is interesting if one wants to irradiate a sample of one of the organic compounds found in table 5 with electrons only, since one knows that the superficial density of the absorber should not exceed 400 mg.cm^{-2}.

Conclusions

The designed experimental arrangement allow report, for the first time, μ (^{90}Y) values for ^{90}Sr/^{90}Y isotopic sources in several organic compounds and in more complex organic macromolecule (Eucalyptus essential oil). On the other hand, Monte Carlo simulation permits to estimate the strong β–particles absorption dependency on sample superficial density in organic compounds.

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