PERGAMON

Applied Radiation and Isotopes 56 (2002) 907–916

Applied Radiation and Isotopes

www.elsevier.com/locate/apradiso

Cherenkov counting of yttrium-90 in the dry state; correlations with phosphorus-32 Cherenkov counting data

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Received 4 October 2001; accepted 15 November 2001

Abstract

We present data that illustrate some advantages of Cherenkov counting for the radioassay of ^{90}Y in the dry state and provide recommendations concerning sample counting geometry. Slightly higher detection efficiencies and figures-of-merit were obtained when counting ^{90}Y in the dry state in polyethylene plastic counting vials compared to the counting of ^{90}Y in 20 ml of water in borosilicate glass vials. The effects of polyethylene plastic counting vials and sample counting geometry are compared to similar data obtained in the Cherenkov counting of ^{32}P. Data are presented to interpret the effects of polyethylene plastic and borosilicate glass on Cherenkov counting efficiency and background counts. Applications of the Cherenkov counting of ^{90}Y and ^{32}P in the dry state in the biological and radiopharmaceutical sciences are foreseen as well as applications in the analysis of $^{90}Sr(^{90}Y)$ and ^{32}P in health physics and environmental monitoring. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Cherenkov counting; Yttrium-90; Phosphorus-32; Strontium-90; Radiopharmaceuticals; Health physics

1. Introduction

The applications of ^{90}Y in radiopharmaceuticals for medical research and cancer treatment are becoming more numerous and only a few examples are cited in this paper (Antoniw et al., 1996; Axworthy et al., 2000; Campbell et al., 2000, 2001; Cremonesi et al., 1999; Dietz and Horwitz, 1992; Erwin et al., 2001; Paganelli et al., 1999; Shoner et al., 1999; Watanabe et al., 1999; Wike et al., 1990; Wiseman et al., 2000). Also, the monitoring of ^{90}Sr levels in the environment remains a widespread concern, and ^{90}Sr analysis can be performed via assay of the daughter nuclide ^{90}Y ingrowth or decay after separation from the parent (L'Annunziata, 1971). Since the pioneering papers on Cherenkov analysis of ^{89}Sr and $^{90}Sr(^{90}Y)$ by Buchtela and Tschurlovits (1975) and Randolph (1975), the Cherenkov counting of these radionuclides in aqueous media has become common and the techniques used have been reviewed previously (L'Annunziata, 1979, 1987, 1998a). Some of the techniques involved are reviewed and tested by Scarpitta et al. (1999).

The methods used for the radioassay of ^{90}Y are reviewed by Coursey et al. (1993). Among these are liquid scintillation counting, Cherenkov counting, and Bremsstrahlung solid scintillation counting with NaI(Tl) detectors. They report (i) high liquid scintillation counting efficiencies for ^{90}Y ($\sim$99.7–99.75%) over a wide range of quench levels, (ii) moderate counting efficiencies over the range of 39–68% using Cherenkov counting depending on instrumentation, sample geometry and color quench level, and (iii) low counting efficiencies with Bremsstrahlung counting ($\sim$9.9–18%) depending on sample counting geometry and detector efficiencies. Cherenkov counting has certain advantages over liquid scintillation analysis, namely, (i) it is less expensive as no scintillation fluor cocktails are used, (ii) it is carried out generally in aqueous solution without the addition of any reagents that could destroy the

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PII: S0969-8043(01)00276-7

sample (e.g. radiopharmaceutical) leaving the sample suitable for furthers tests (e.g. spectroscopy, chromatography, electrophoresis, bioassays, etc.), and (iii) no interference is caused by other radionuclides in the sample with decay emissions that cannot produce the Cherenkov effect, such as, ^{3}H, ^{14}C, ^{35}S, ^{33}P, ^{45}Ca, etc. When interfering radionuclides are present in the sample, such as ^{137}Cs, ^{89}Sr and ^{60}Co among others, sometimes encountered in the analysis of environmental samples for ^{90}Sr(^{90}Y), Brajnik et al. (1994, 1995) demonstrated the possibility of discriminating against these by use of low-refractive index silica aerogel counting medium to increase the electron threshold energy required for the production of Cherenkov radiation.

The Cherenkov counting of radionuclides in the dry state can be more convenient, at times, such as in the analysis of radioisotope-labeled biochemicals or radiopharmaceuticals on filter material, electrophoresis gels, tissue homogenates, precipitates, etc. or swipes taken for the monitoring of surface contamination. The first report of Cherenkov counting of a radionuclide in the dry state was provided by Hülsen and Prenzel (1968), who analyzed the amounts of ^{32}P tracer in the green algae *Chlorella* by counting the Cherenkov photons. The dry algae was collected on glass fiber filter and deposited into glass or plastic vials. They used a conventional liquid scintillation analyzer and reported a detection efficiency of 13% for ^{32}P in the dry state for glass vials. The detection efficiency was reported to increase twofold when plastic vials were used. Further studies by Berger (1984) demonstrated the analysis of dry samples of ^{32}P in recombinant DNA procedures where enzyme-catalyzed reactions were carried out in volumes of 10 μl or less. In this case, the ^{32}P samples were deposited onto glass fiber filters, dried under heat lamps and then inserted into glass counting vials. Detection efficiencies of $25 \pm 1\%$ were reported with the glass vials assayed with a conventional liquid scintillation analyzer. As it is common to use microplate scintillation and luminescence counters in the biological sciences, tests performed on the Packard TopCount microplate scintillation and luminescence counters with ^{32}P in the dry state in 24- and 96-well plastic OptiPlates^{TM} have demonstrated Cherenkov photon detection efficiencies of 10% and 14%, respectively (Anonymous, 1996; L'Annunziata, 1998a). Examples of more recent studies in the biological sciences report the use of a conventional liquid scintillation analyzer for the Cherenkov counting of dry samples of ^{32}P on Whatman phosphocellulose paper (Luciani et al., 2000), purified and lyophilized 5′-nucleotide monophosphates (Lehmann and Bass, 1999), and precipitated covalent histone–DNA complexes (Angelov et al., 2000). Further studies by Morita-Murase et al. (2000) demonstrate a linear relationship between Cherenkov counting efficiencies and average energies of the emitted beta-particles and internal conversion electrons using standards of ^{32}P, ^{36}Cl, ^{60}Co and ^{137}Cs in the dry state. The radioisotope samples were spotted onto 0.65-μm-pore cellulose nitrate membrane filter paper of 10 mm diameter and air-dried. The filter paper was then suspended by wire from the glass counting vial cap to permit the paper to be situated geometrically in the center of the counting vial with the paper surface parallel to the faces of the two photomultiplier tubes of the liquid scintillation analyzer. They reported a 38.8% Cherenkov counting efficiency for ^{32}P in the dry state with air-filled glass vials using an Aloka Model 3500 liquid scintillation counter with the photomultipliers in the coincidence counting mode.

The previously cited studies on the Cherenkov counting of ^{32}P in the dry state provided the basis for our work with ^{90}Y. The increasing applications of ^{90}Y in the radiopharmaceutical, medical, and biological sciences sparked the interest of the writers to investigate the Cherenkov counting of ^{90}Y in the dry state. Also the possibility of analyzing swipe material for ^{90}Sr(^{90}Y) in the dry state without further sample preparation would be of interest to many persons in the field of radiological protection concerned with laboratory swipe tests. This paper reports results of tests performed to compare and evaluate Cherenkov counting of ^{90}Sr(^{90}Y) in the dry state and in aqueous media in plastic and glass counting vials. Although standards of ^{90}Sr(^{90}Y) were used in this study, the results of Cherenkov counting were interpreted as counts for all practical purposes derived exclusively from ^{90}Y beta-particles, because the optimum Cherenkov counting efficiency reported for ^{90}Sr is only $\sim 1\%$ (Rucker, 1991; Chang et al., 1996; Cook et al., 1998). The Cherenkov counting data of ^{90}Y are compared to data collected in the counting of ^{32}P under similar sample counting conditions. An interpretation of the results obtained and recommendations for the radioassay of ^{90}Y and ^{32}P by Cherenkov counting are provided.

2. Experimental

2.1. Radionuclides

A sample of ^{90}Sr(^{90}Y) was obtained from Tennelec, Inc. a subsidiary of Canberra Industries, Oak Ridge, TN 37830, USA and used as radionuclide standard for this study. The radionuclide was provided in the quantity of 3.7 kBq in 5 ml as $SrCl_2$ in dilute HCl solution. The absolute activity of the radionuclide was confirmed as 5.0 kBq by liquid scintillation analysis of six replicate samples in 10 ml of Ultima Gold LLT cocktail (Packard BioScience, Meriden, CT). A Packard Tri-Carb Model 3100TR liquid scintillation counter was used. The radionuclide activity was confirmed with 2.1% standard deviation for the replicate samples using the efficiency tracing technique previously described (Noor et al., 1996). The precision of the determination was suitable for this work. Most accurate disintegration rates for standardization can be obtained using the efficiency tracing with ^{3}H method described by Coursey et al. (1984, 1986, 1989), Grau Malonda (1999), Grau Malonda and García-Toraño (1982), Grau Malonda et al. (1985), and Günther (1996).

2.2. Instrumentation

A Packard Tri-Carb™ 3100TR liquid scintillation analyzer from Packard BioScience, Meriden, CT was used for Cherenkov counting. The instrument is equipped with dual photomultiplier tubes, and counting was carried out in coincidence mode with an 18 ns coincidence resolving time. The normal count mode (NCM) was employed. The analyzer is equipped with a 4000 channel multichannel analyzer calibrated to register pulse events over the linear energy scale of 0–2000 keV.

2.3. Counting vials

Cherenkov counting was carried out with 20-ml volume polyethylene plastic (1-mm wall thickness) and low-potassium borosilicate glass sample vials (Packard BioScience). Background counts were determined also with both plastic and borosilicate glass vials.

2.4. Sample preparation for Cherenkov counting

Aliquots of 0.1 ml of ^{90}Sr(^{90}Y) radionuclide standard were added to counting vials so that each vial would contain 98.98 Bq. Four sets of six polyethylene plastic vials and six low-potassium borosilicate glass vials were prepared to contain the ^{90}Sr(^{90}Y) standard. The following volumes of distilled water: 0, 1, 4, 8, 12, 16 and 20 ml were added to each set of the plastic and glass vials. The vials that received no additional water were allowed to air-dry. The air-dried samples were applied to the bottom of the vial, and no residue was visible in the vials after drying. The vials were capped and ready for Cherenkov counting.

2.5. Cherenkov counting

The pulse height discriminator settings were determined to define a suitable counting region for Cherenkov photons. This was carried out by counting plastic and glass vials containing 98.98 Bq of ^{90}Sr(^{90}Y) in 20 ml of distilled H_2O in the liquid scintillation analyzer to observe the Cherenkov photon pulse height spectra. No significant pulse events could be observed beyond 50 keV when either plastic or glass vials were used. This is in agreement with past observations (Passo and Cook, 1994; Noor et al., 1996; L'Annunziata, 1998a; Scarpitta et al., 1999). Therefore, a counting region of 0–50 keV was selected for the lower-level (LL) and upper-level (UL) pulse-height discriminators in order to count all of the pulse events originating from the Cherenkov photons. A 1-min counting time was selected for the four sets of samples containing ^{90}Sr(^{90}Y) in plastic and glass vials. This was adequate time considering the sample activities.

2.6. Background counting

Four background count determinations were made in the counting region of 0–50 keV (LL–UL) on the following blank samples: (i) empty polyethylene plastic vial containing only air, (ii) polyethylene plastic vial containing 20 ml of distilled water, (iii) empty borosilicate glass vial containing only air, and (iv) borosilicate glass vial containing 20 ml of distilled water. The samples were counted for 100 min in order to collect sufficient counts and produce measurable background pulse height spectra. The background spectra were then analyzed by SpectraWorks™ (Packard BioScience), spectrum analysis software, to analyze the pulse-height spectra and extract the background count rates in the region of 0–50 keV.

2.7. ^{90}Y Cherenkov counting efficiency

The contribution of ^{90}Sr to the Cherenkov counting efficiency was ignored, because of the very low detection efficiency ($\sim$1%) achievable. Therefore, counting of Cherenkov photons from ^{90}Sr(^{90}Y) beta-particles could be considered due solely to ^{90}Y emissions for all practical purposes. The Cherenkov counting detection efficiency for ^{90}Y was thus calculated as follows:

$$\%E = \frac{\mathrm{CPM}}{\mathrm{DPM}_{^{90}\mathrm{Sr}(^{90}\mathrm{Y})}/2} \times 100, \quad (1)$$

where $\%E$ is the Cherenkov counting efficiency of ^{90}Y for a ^{90}Sr(^{90}Y) sample in secular equilibrium, CPM is the Cherenkov photon count rate, and $\mathrm{DPM}_{^{90}\mathrm{Sr}(^{90}\mathrm{Y})}$ is the disintegration rate of the ^{90}Sr(^{90}Y) standard in secular equilibrium. The latter term is divided by two, as the DPM due to ^{90}Y is only one-half the disintegration rate of the ^{90}Sr(^{90}Y) sample when in secular equilibrium.

3. Results and discussion

3.1. ^{90}Y Cherenkov counting efficiency as a function of sample volume and vial type

The ^{90}Y counting efficiency varied with sample volume as expected (Coursey et al., 1993; L'Annunziata, 1997, 1998a). This was more significant with sample volumes in the range of 0–4 ml as illustrated in Fig. 1. Sample geometry has a less significant effect on Cherenkov counting efficiency when polyethylene plastic vials were used. In addition, plastic vials provided invariably an increased ^{90}Y Cherenkov counting efficiency and an optimum counting efficiency of approximately 7% over glass vials. The effect of sample geometry and vial type on ^{90}Y Cherenkov counting efficiency is similar to that found previously in the Cherenkov counting of ^{32}P as illustrated in Fig. 2. The air-dried samples gave the lowest ^{90}Y counting efficiencies as expected (66.9% and 51.3% for plastic and glass vials, respectively). The highest ^{90}Y Cherenkov counting efficiency recorded was 74.0% for a 16 ml sample volume in plastic vials of which approximately 1% is due to ^{90}Sr Cherenkov photons. The optimum detection efficiency for ^{90}Sr(^{90}Y) obtained in this work was similar to the value of 71.7% reported by Passo and Cook (1994) for a more narrow counting region of 0–30 keV.

Cherenkov counting efficiencies were always highest when using polyethylene plastic vials as compared to borosilicate glass vials regardless of the sample state, air-dried or in aqueous solution. These results may be best interpreted by consideration of the indexes of refraction of the various media involved and the consequent Cherenkov threshold energies. A more detailed treatment is provided by L'Annunziata (1998a), and only the relevant equations are used here. The threshold condition for the production of Cherenkov radiation in a transparent medium is derived from the equation

$$\beta n = 1, \tag{2}$$

where β is the relative phase velocity of the particle, that is, the velocity of the particle divided by the speed of light in a vacuum and n is the index of refraction of the medium. Only charged particles that possess

$$\beta > 1/n \tag{3}$$

produce Cherenkov photons in transparent media. Where beta-particles or electrons are concerned, the value of β is a function of the electron or beta-particle energy according to the equation

$$\beta = \left[1 - \left(\frac{1}{E/511\,\mathrm{keV} + 1}\right)^2\right]^{1/2}, \tag{4}$$

where E is the beta-particle energy in keV. From Eqs. (2) and (4) the minimum or threshold energy that beta-particles must possess for the production of Cherenkov radiation as a function of the index of refraction of the medium is derived as

$$E = 511\,\mathrm{keV}\left[\left(1 - \frac{1}{n^2}\right)^{-1/2} - 1\right]. \tag{5}$$

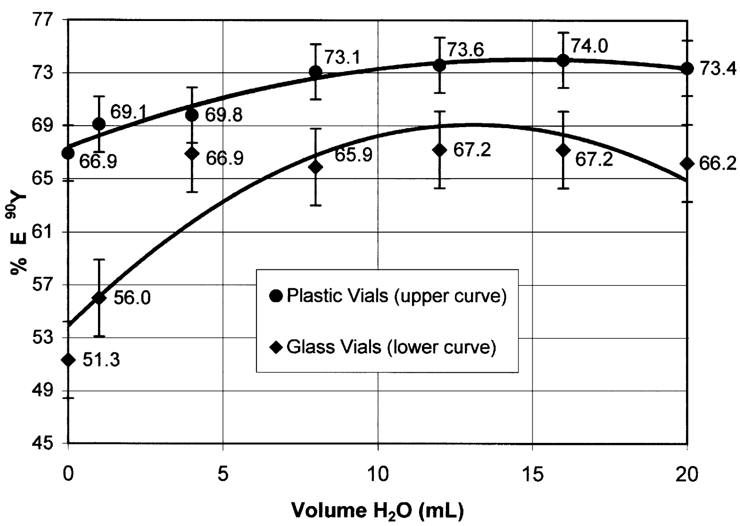

Fig. 1. Cherenkov counting efficiency for ^{90}Y as a function sample volume and counting vial type. The upper and lower curves provide data illustrating the performance of polyethylene plastic and borosilicate glass vials, respectively. The counting efficiencies at 0 ml of water were obtained with samples in the dry state. The error bars represent the standard deviation of replicate measurements. The mean counting efficiency values are identified beside each data point.

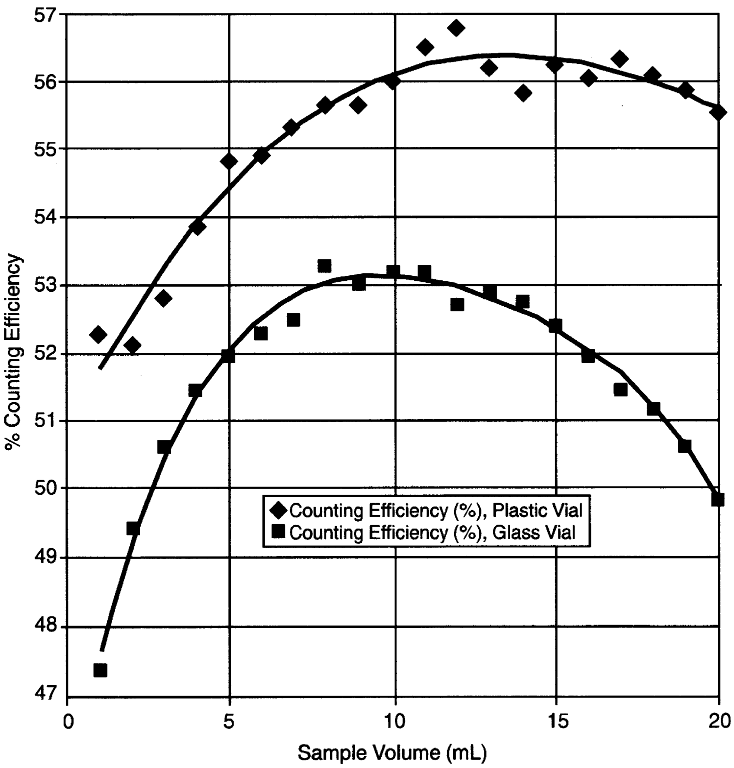

Fig. 2. Effect of sample volume on the Cherenkov counting efficiency of ^{32}P in water with polyethylene plastic and borosilicate glass counting vials measured with a Packard Tri-Carb 2300TR liquid scintillation analyzer (From L'Annunziata, 1997, reprinted with permission of the Packard BioScience Co., Meriden, CT).

When water is the medium, where $n = 1.332$, the threshold energy for the production of Cherenkov photons is calculated according to Eq. (5) as 263 keV. Thus, only beta-particles that possess energy in excess of 263 keV produce Cherenkov photons in water.

If we count ^{90}Y in the dry state, we have only air as the medium plus the container walls of the plastic or glass counting vials. Air has a very low index of refraction ($n = 1.00027712$) at the sodium D line at STP (standard temperature and pressure) as provided by Lide (2001). According to Eq. (5) beta-particles would have to exceed the threshold energy of 2.12×10^4 keV or 0.0212 GeV for the production of Cherenkov photons in air. Consequently, no Cherenkov photons are produced by the ^{90}Y beta-particles in air ($E_{max} = 2,280$ keV). Gases as Cherenkov detection media are limited to the measurement of high-energy particles in the MeV and GeV regions. For example, Iodice et al. (1998) calculated the threshold energy for the production of Cherenkov photons by electrons or positrons in CO_2 at STP ($n = 1.00041$) to be 0.017 GeV.Since no Cherenkov photons can be attributed to the gaseous medium surrounding the ^{90}Y in the dry state, there remains only the counting vial wall material to consider. The indexes of refraction of low-, medium- and high-density polyethylene plastics vary over the range of $n = 1.50-1.54$ while the values for borosilicate glasses vary over the range of $n = 1.468-1.487$ according to data from Bolz and Tuve (1975). If we select median values of $n = 1.52$ for polyethylene plastic and $n = 1.477$ for borosilicate glass, the threshold beta-particle energies for Cherenkov radiation production in the plastic and glass counting vials are calculated according to Eq. (5) as 167 and 183 keV, respectively. Thus, the lower threshold energy for the production of Cherenkov photons in polyethylene plastic counting vials compared to borosilicate glass vials, will contribute somewhat to the higher Cherenkov counting efficiency of ^{90}Y and ^{32}P encountered with polyethylene plastic counting vials. This is particularly relevant when counting these radionuclides in the dry state.

Increasing the wall thickness of the plastic vial would increase the path length of travel of the beta-particles in the lower-threshold energy medium. This should increase the number of Cherenkov photons produced. As described by Sundaresan (2001), the number of photons dN emitted as Cherenkov radiation per path length of particle travel dx in the medium is defined by the expression:

$$\frac{\mathrm{d}N}{\mathrm{d}x} = \alpha z^2 \int_{\beta n \succ 1} \mathrm{d}\omega \left(1 - \frac{1}{\beta^2 n^2}\right), \tag{6}$$

where α is the fine structure constant (1/137), z is the number of units of charge on the particle ($z = 1$ for beta-particles), and ω is the photon frequency. From the above equation, the number of photons N emitted in a path length L over the frequency range between ω_1 and ω_2 is

$$N = \alpha z^2 L \int_{\omega_2}^{\omega_1} \sin^2\theta \, \mathrm{d}\omega, \tag{7}$$

where θ is the angle of photon emission with respect to the direction of beta-particle travel. Sundaresan (2001) evaluates the above integral approximately by assuming the integrand to be essentially constant as a function of ω to become

$$N \approx \alpha z^2 L \sin^2\theta(\omega_1 - \omega_2) \tag{8}$$

and considering only the visible range of wavelengths from $\lambda_1 = 400\,\mathrm{nm}$ to $\lambda_2 = 700\,\mathrm{nm}$, the above equation provides the number of photons emitted N per path length L as

$$N/L \approx 490 \sin^2\theta \,\mathrm{cm}^{-1} \tag{9}$$

Consequently, from Eq. (9) the number of Cherenkov photons emitted could be increased and the Cherenkov counting efficiency of radionuclides in the dry state improved by increasing L, the polyethylene plastic counting vial wall thickness, that is, the effective particle path length of travel.

The polyethylene plastic vials used in this experiment had a wall thickness of only 1 mm. Thicker-walled (2 mm) plastic vials are available commercially (Maxi-Vial™, Packard BioScience). These, as well as others, could be tested for increased Cherenkov counting efficiency. The large difference between the beta-particle threshold energies for the production of Cherenkov photons in water (263 keV) and that of polyethylene plastic (167 keV) is very convincing evidence for the higher Cherenkov counting efficiency for ^{90}Y and ^{32}P in the dry state in polyethylene plastic counting vials. Another factor that could contribute to the higher Cherenkov counting efficiencies measured with plastic counting vials is the higher UV transparency of plastic compared to the borosilicate glass.

Lower Cherenkov detection efficiencies occur when counting small samples of ^{90}Y or ^{32}P that rest at the lower part (0–4 ml) of the counting vial for either dry or wet samples as illustrated in Figs. 1 and 2. The effect of sample counting geometry for small sample volumes (0–4 ml) is due to larger numbers of Cherenkov photons produced at the bottom of the counting vial further away from the center of the photomultiplier tube faces. When counting samples in the dry state (e.g. biological samples, filter or swipe material with ^{90}Sr(^{90}Y) or ^{32}P, etc.), inserting a 1-cm thick polyethylene plastic disk of suitable diameter at the bottom of the vial would elevate the dry sample and consequently should reduce this geometry effect as well as increase the amount of plastic medium in the vial to enhance the production of Cherenkov radiation.

3.2. ^{90}Y counting backgrounds and figures-of-merit for plastic versus glass vials

As seen from the data in Table 1, dry samples of ^{90}Y in plastic vials provided figures-of-merit of the same order of magnitude as samples in 20 ml of water in glass counting vials. The background count rates were lower with plastic counting vials than glass regardless of whether the vials were empty or filled with water. This would be expected as natural radionuclides that could produce Cherenkov photons, such as ^{40}K present in glass, would be essentially absent from plastic vials. An interesting observation here is that lower background count rates were obtained when the vials contained water compared to the dry empty (air-filled) vials. Data provided by the background pulse-height spectra

Table 1
Cherenkov counting of ^{90}Sr(^{90}Y) in the dry state and in water

Medium	Volume[a] (ml)	^{90}Y counting efficiency (%)		Background (cpm)		FOM[b]	
		Glass[c]	Plastic[d]	Glass[c]	Plastic[d]	Glass[c]	Plastic[d]
Air[e]	20	51.3	66.9	22.2	20.4	119	219
Water	20	66.2	73.4	21.4	16.1	205	334

[a] Volume of the medium (air volume for dry samples or water volume).
[b] Figure-of-merit calculated as the % counting efficiency squared divided by the background count rate (cpm), i.e. E^2/B.
[c] Borosilicate glass vials.
[d] Polyethylene plastic vials.
[e] The medium for air-dried samples.

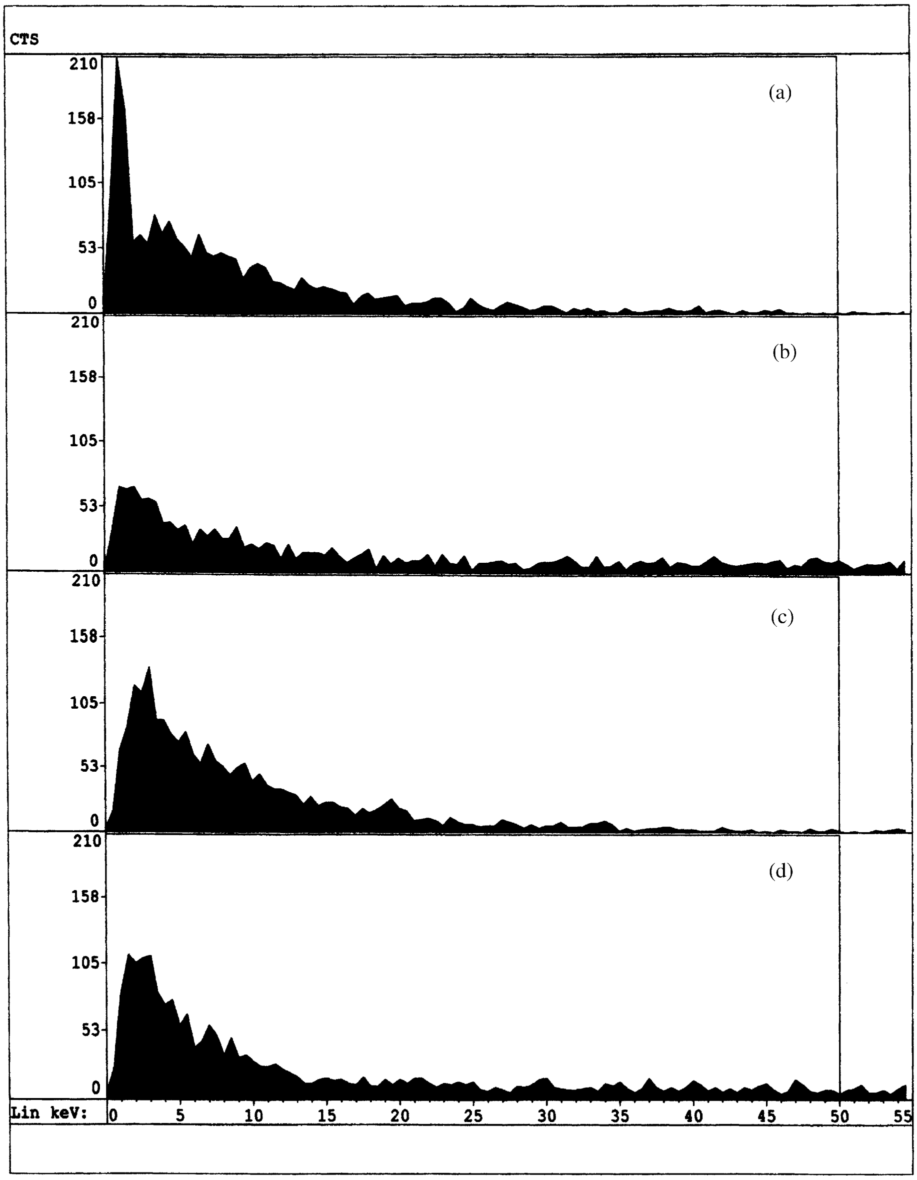

Fig. 3. Pulse height spectra (counts versus keV) of the four types of background count data collected with 100-min count times plotted with SpectraWorks™ (Packard BioScience). The four types of background blanks are described in descending order with the total counts collected in the region 0–50 keV provided in parenthesis as follows: (a) empty (air-filled) polyethylene plastic vial (2041 total counts), (b) polyethylene plastic vial filled with 20 ml of distilled water (1611 total counts), (c) empty borosilicate glass vial (2219 total counts), and (d) borosilicate glass vial filled with 20 ml of distilled water (2135 total counts). The vertical line at 50 keV represents the upper-level pulse height discriminator setting.

discussed subsequently provide information to interpret these results.

The pulse-height spectra of the four types of background count data are illustrated in Fig. 3. These show significant differences in background photon intensities for empty (air-filled) plastic and glass vials and water-filled vials. The higher background counts obtained with empty plastic and glass vials particularly in the 0–4 keV region, as illustrated in Fig. 3, may be due to high-energy cosmic radiation interactions, which could lead to air luminescence. It is known that particles of relatively high mass and charge, such as alpha-particles, produce luminescence via principally N_2 excitations in air (Takiue and Ishikawa, 1979; Takiue, 1980; Homma et al., 1987; Murase et al., 1989a, b; L'Annunziata and Kessler, 1998). Water-filled vials would prevent air luminescence and, at the same time, water could attenuate possibly some Cherenkov photons produced in the vial wall material.

3.3. Self-absorption

Self-absorption, that is, absorption of beta-particle radiation by the sample, should be considered when counting samples in the dry state. Fortunately, the lower-energy beta-particles are the first to be absorbed by the sample depending on sample thickness and density. In view of the beta-particle threshold energy for Cherenkov photon production in polyethylene plastic of 167 keV, the sample thickness and density should not be capable of absorbing beta-particles of energy in excess of 167 keV or even reduce significantly the energy of beta-particles escaping from the dry sample to energies below the threshold energy of 167 keV. If the sample thickness is such that self-absorption would occur at any appreciable extent, then an internal standard might be required to determine the degree to which self-absorption occurs. Several empirical formulae are available for calculating the average range for electrons or beta-particles in matter. These may be used to calculate the average range of beta-particles from samples in the dry state including the following formula of Flammersfeld (1946) described by Paul and Steinwedel (1955) and employed by Grau Malonda and Grau Carles (1998):

$$R = 0.11(\sqrt{1 + 22.4E^2} - 1) \quad \text{for } 0 < E < 3\,\text{MeV}, \tag{10}$$

where R is the beta-particle range in $\mathrm{g\,cm^{-2}}$ and E is the beta-particle energy in MeV, or that reported by Glendenin (1948) and described by L'Annunziata (1998b):

$$R = 0.407E^{1.38} \quad \text{for } 0.15\,\text{MeV} < \text{E} < 0.8\,\text{MeV}. \tag{11}$$

Both of the above formula provide similar values and in agreement with those obtained from the range–energy curve of Friedlander et al. (1964). For 167 keV beta-particles, the range is calculated to be 0.030 or $30\,\mathrm{mg\,cm^{-2}}$. To put this in perspective, commercial aluminum foil of 0.0025 cm thickness would have a surface density of only $6.75\,\mathrm{mg\,cm^{-2}}$ or commercial borosilicate glass fiber filter material of 0.0035 cm thickness, commonly used in biological research, would have a surface density of only $7.88\,\mathrm{mg\,cm^{-2}}$. Both of these materials are well within the range of 167 keV beta-particles where self-absorption might be ignored. In any case, tests for self-absorption using radionuclide of known activity are recommended.

3.4. Color quench

Chemical quench is absent in Cherenkov counting; however, color quench can be significant. If dry or aqueous samples are colored, which is sometimes encountered with dissolved biological samples, a color-quench correction curve based on a quench indicating parameter (QIP) derived from the sample pulse height spectra, could be used (L'Annunziata, 1998a). Alternatively, colored samples can be bleached to remove color. This is a good alternative, if the sample chemistry does not have to be preserved for subsequent tests, such as, radiopharmaceutical structural analysis, chromatography, electrophoresis, etc.

4. Conclusions and recommendations

The Cherenkov counting of ^{90}Y in the dry state is possible with only a slight reduction in detection efficiency compared to counting in aqueous solution. Polyethylene plastic counting vials provide significantly higher Cherenkov counting efficiencies than borosilicate glass vials. Dry samples of ^{90}Y in polyethylene plastic counting vials provide figures-of-merit of the same order of magnitude as samples containing 20 ml of water in glass counting vials. Evidence is provided to show the advantageous effects of polyethylene plastic vials for the Cherenkov counting of ^{90}Y and ^{32}P. The higher Cherenkov counting efficiencies obtained with plastic vials can be due to the higher index of refraction of plastic providing a lower beta-particle energy threshold for the production of Cherenkov photons, and the higher UV transparency of plastic over glass vials. The significantly higher index of refraction of polyethylene plastic ($n \cong 1.52$) compared to that of water ($n = 1.332$) attributes to plastic counting vials the property of reducing the beta-particle threshold energy for the production of Cherenkov photons from 263 keV in water to 167 keV in plastic.

The Cherenkov counting of radionuclides, such as ^{90}Y and ^{32}P among others, in the dry state or in aqueous solution should be carried out in polyethylene plastic counting vials. Lower Cherenkov counting efficiencies in 20-ml counting vials are encountered when samples are dried on the bottom (floor) of the vial or as small volumes (0–4 ml) in aqueous solution. When counting samples in the dry state, such as filter material, electrophoresis gels, and swipe material, etc., elevating the sample above the 4 ml volume level of a 20-ml counting vial should improve the detection efficiency. This may be accomplished by inserting a 1-cm thick polyethylene disk into the bottom of the vial before adding the dry sample.

When counting samples in the dry state in polyethylene plastic vials, the sample thickness should be kept well within the range limits of 167 keV beta-particles to avoid the deleterious effects of self-absorption. If self-absorption is suspected, the degree of self-absorption could be determined experimentally with radionuclide standard.

Acknowledgements

The authors wish to thank Tom Gerber for his assistance in providing background count rates for empty- and water-filled polyethylene plastic and borosilicate glass counting vials.

References

Angelov, D., Charra, M., Seve, M., Côté, J., Khochbin, S., Dimitrov, S., 2000. Differential remodeling of the HIV-1 nucleosome upon transcription activators and SWI/SNF complex binding. J. Mol. Biol. 302 (2), 315–326.

Anonymous, 1996. Cerenkov counting performance on the TopCount™ microplate scintillation and luminescence counter. TopCount Topics, TCA-024, Packard BioScience Company, Meriden, CT.

Antoniw, P., Farnsworth, A.P.H., Turner, A., Haines, A.M.R., Mountain, A., Mackintosh, J., Shochat, D., Humm, J., Welt, S., Old, L.J., Yarranton, G.T., King, D.J., 1996. Radioimmunotherapy of colorectal carcinoma xenografts in nude mice with yttrium-90 A33 IgG and Tri-Fab (TFM). Br. J. Cancer 74 (4), 513–524.

Axworthy, D.B., Reno, J.M., Hylarides, M.D., Mallett, R.W., Theodore, L.J., Gustavson, L.M., Su, F.-M., Hobson, L.J., Beaumier, P.L., Fritzberg, A.R., 2000. Biological sciences–medical sciences—cure of human carcinoma xenografts by a single dose of pretargeted yttrium-90 with negligible toxicity. Proc. Natl. Acad. Sci. (USA) 97 (4), 1802–1807.

Berger, S.L., 1984. The use of Cerenkov radiation for monitoring reactions performed in minute volumes: examples from recombinant DNA technology. Anal. Biochem. 136, 515–519.

Bolz, R.E., Tuve, G.L. (Eds.), 1975. CRC Handbook of Tables for Applied Engineering Science. 2nd Edition, CRC Press, Inc., Boca Raton, FL, pp. 140, 163.

Brajnik, D., Korper, S., Medin, G., Stariè, M., Stanovik, A., 1994. Measurement of ^{90}Sr activity with Cherenkov radiation in a silica aerogel. Nucl. Instrum. Methods A 353, 217–221.

Brajnik, D., Medin, G., Stanovik, A., Stariè, M., 1995. Determination of high energy β-emitters with a Ge spectrometer or Cherenkov radiation in an aerogel. Sci. Total Environ. 173/174, 225–230.

Buchtela, K., Tschurlovits, M., 1975. A new method for determination of ^{89}Sr, ^{90}Sr and ^{90}Y in aqueous solution with the aid of liquid scintillation counting. Int. J. Appl. Radiat. Isot. 26, 333–338.

Campbell, A.M., Bailey, I.H., Burton, M.A., 2000. Analysis of the distribution of intra-arterial microspheres in human liver following hepatic yttrium-90 microsphere therapy. Phys. Med. Biol. 45 (4), 1023–1034.

Campbell, A.M., Bailey, I.H., Burton, M.A., 2001. Tumour dosimetry in human liver following hepatic yttrium-90 microsphere therapy. Phys. Med. Biol. 46 (2), 487–498.

Chang, T.-M., Chen, S.-C., King, J.-Y., Wang, S.-J., 1996. Rapid and accurate determination of $^{89/90}$Sr in radioactive samples by Cerenkov counting. J. Radioanal. Nucl. Chem. Articles 204, 339–347.

Cook, G.T., Passo Jr., C.J., Carter, B.D., 1998. Environmental liquid scintillation analysis. In: L'Annunziata, M.F. (Ed.), Handbook of Radioactivity Analysis. Academic Press, San Diego, pp. 331–386.

Coursey, B.M., Gibson, J.A.B., Heitzmann, M.W., Leak, J.C., 1984. Standardization of technetium-99 by liquid scintillation counting. Appl. Radiat. Isot. 35, 1103–1112.

Coursey, B.M., Mann, W.B., Grau Malonda, A., García-Toraño, E., Los Arcos, J.M., Gibson, J.A.B., Reher, D., 1986. Standardization of carbon-14 by $4\pi\beta$ liquid scintillation efficiency tracing with hydrogen-3. Appl. Radiat. Isot. 37, 403–408.

Coursey, B.M., Lucas, L.L., Grau Malonda, A., García-Toraño, E., 1989. The standardization of plutonium-241 and nickel-63. Nucl. Instrum. Methods A 279, 603–610.

Coursey, B.M., Calhoun, J.M., Cessna, J.T., 1993. Radioassays of yttrium-90 used in nuclear medicine. Nucl. Med. Biol. 20 (5), 693–700.

Cremonesi, M., Ferrari, M., Chinol, M., Stabin, M.G., Grana, C., Prisco, G., Robertson, C., Tosi, G., Paganelli, G., 1999. Three-step radioimmunotherapy with yttrium-90 biotin: dosimetry and pharmacokinetics in cancer patients. Eur. J. Nucl. Med. 26 (2), 110–120.

Dietz, M.L., Horwitz, E.P., 1992. Improved chemistry for the production of yttrium-90 for medical applications. Appl. Radiat. Isot. 43 (9), 1093–1101.

Erwin, W.D., Spies, S.M., Kelly, M.E., Rao, P., Eckersberg-Rhodes, T.E., Nannapaneni, M., Groch, M.W., 2001. Correlation of marrow dose estimates based on serial pretreatment radiopharmaceutical imaging and blood data with actual marrow toxicity in anti-CD20 yttrium-90 monoclonal antibody radioimmunotherapy of non-Hodgkin's B-cell lymphoma. Nucl. Med. Comm. 22 (2), 247–255.

Flammersfeld, A., 1946. Eine Beziehung zwischer Energie und Reichweite für Beta-Strahlen kleiner und mittlerer Energie. Naturwissenschaften 33, 280–281.

Friedlander, G., Kennedy, J.W., Miller, J.M., 1964. Nuclear and Radiochemistry, 2nd Edition. Wiley Inc., New York, pp. 106–107.

Glendenin, L.E.Jan., 1948. Determination of the energy of beta particles and photons by absorption. Nucleonics 2, 12–32.

Grau Malonda, A., 1999. Free Parameter Models in Liquid Scintillation Counting. Editorial Ciemat, Madrid, pp. 416.

Grau Malonda, A., García-Toraño, E., 1982. Evaluation of counting efficiency in liquid scintillation counting of pure β-ray emitters. Int. J. Appl. Radiat. Isot. 33, 249–253.

Grau Malonda, A., Grau Carles, A., 1998. The anisotropy coefficient in Cerenkov counting. Appl. Radiat. Isot. 49 (9–11), 1049–1053.

Grau Malonda, A., García-Toraño, E., Los Arcos, J.M., 1985. Liquid scintillation counting efficiency as a function of the figure of merit for pure beta-particle emitters. Int. J. Appl. Radiat. Isot. 36, 157–158.

Günther, E.W. 1996. A simple method for transferring the tritium calibration of an LSC system to other radionuclides. In: Cook, G.T., Harkness, A.B., MacKenzie, A.B., Miller, B.F., Scott, E.M. (Eds.), Liquid Scintillation Spectrometry 1994. Radiocarbon, Tucson, 1996, pp. 373–379.

Homma, Y., Murase, Y., Takiue, M., 1987. Determination of ^{222}Rn by air luminescence method. J. Radioanal. Nucl. Chem. Lett. 119 (6), 457–465.

Hülsen, W., Prenzel, U., 1968. "Dry" counting of ^{32}P in liquid scintillation counters. Anal. Biochem. 26, 483–484.

Iodice, M., Cisbani, E., Colilli, S., Crateri, R., Frullani, S., Garibaldi, F., Giuliani, F., Gricia, M., Lucentini, M., Mostarda, A., Pierangeli, L., Santavenere, F., Urciuoli, G.M., De Leo, R., Lagamba, L., Leone, A., Perrino, R., Kerhoas, S., Lugol, I.C., Mazaev, B., Vernin, P., Zaccarian, A., 1998. The CO_2 gas Cherenkov detectors for the Jefferson Lab Hall-A spectrometers. Nucl. Instrum. Methods Phys. Res. Sect. A 411, 223–237.

L'Annunziata, M.F., 1971. Birth of a unique parent-daughter relation: Secular equilibrium. J. Chem. Educ. 48, 700–703.

L'Annunziata, M.F., 1979. Radiotracers in Agricultural Chemistry. Academic Press, London, pp. 105–112.

L'Annunziata, M.F., 1987. Radionuclide Tracers, Their Detection and Measurement. Academic Press, London, pp. 241–264.

L'Annunziata, M.F., 1997. Cerenkov counting of ^{32}P—instrument performance data. Packard BioScience Counter Intel. Tri-Carb LSC Application Note 002, p. 6.

L'Annunziata, M.F., 1998a. Cherenkov counting. In: L'Annunziata, M.F. (Ed.), Handbook of Radioactivity Analysis. Academic Press, San Diego, pp. 453–505.

L'Annunziata, M.F., 1998b. Nuclear radiation, its interaction with matter and radioisotope decay. In: L'Annunziata, M.F. (Ed.), Handbook of Radioactivity Analysis. Academic Press, San Diego, pp. 1–74.

L'Annunziata, M.F., Kessler, M.J., 1998. Radiotracer liquid scintillation analysis. In: L'Annunziata, M.F. (Ed.), Handbook of Radioactivity Analysis. Academic Press, San Diego, pp. 209–329.

Lehmann, K.A., Bass, B.L., 1999. The importance of internal loops within RNA substrates of ADAR1. J. Mol. Biol. 291 (1), 1–13.

Lide, D.R. (Ed.), 2001. CRC Handbook of Chemistry and Physics, 81st Edition, CRC Press, Boca Raton, FL, pp. 10–220.

Luciani, M.G., Hutchins, J.R.A., Zheleva, D., Hupp, T.R., 2000. The C-terminal regulatory domain of p53 contains a functional docking site for cyclin A. J. Mol. Biol. 300 (3), 503–518.

Morita-Murase, Y., Murakami, I., Homma, Y., 2000. Cherenkov counting efficiencies for β^--emitters in dry state in glass vials. Chem. Lett. 10, 1158–1159.

Murase, Y., Homma, Y., Takiue, M., Aburai, T., 1989a. Determination of air luminescence spectra for alpha-emitters with liquid scintillation spectrometers. Appl. Radiat. Isot. 40 (4), 219–294.

Murase, Y., Homma, Y., Takiue, M., 1989b. Effect of air luminescence counts on determination of ^{222}Rn by liquid scintillation counting. Appl. Radiat. Isot. 40 (4), 295–298.

Noor, A., Zakir, M., Rasyid, B., Maming, L'Annunziata, M.F., 1996. Cerenkov and liquid scintillation analysis of the triple label ^{86}Rb–^{35}S–^{33}P. Appl. Radiat. Isot. 47(7), 659–668.

Paganelli, G., Grana, C., Chinol, M., Cremonesi, M., De Cicco, C., De Braud, F., Robertson, C., Zurrida, S., Casadio, C., Zobolli, S., Siccardi, A., Veronesi, U., 1999. Antibody-guided three-step therapy for high grade glioma with yttrium-90 biotin. Eur. J. Nucl. Med. 26 (4), 348–357.

Passo Jr., C.J., Cook, G.T., 1994. Handbook of Environmental Liquid Scintillation Spectrometry. Publ. PMC0387 12/94, Packard BioScience, Meriden, CT, pp. 9–8.

Paul, W., Steinwedel, H., 1955. Interaction of electrons with matter. In: Siegbahn, K. (Ed.), Beta- and Gamma-ray Spectroscopy. North-Holland, Amsterdam, pp. 22.

Randolph, R., 1975. Determination of strontium-90 and strontium-89 by Cerenkov and liquid scintillation counting. Int. J. Appl. Radiat. Isot. 26, 9–16.

Rucker, T.L., 1991. Calculational method for the resolution of ^{90}Sr and ^{89}Sr counts from Cerenkov and liquid scintillation counting. In: Ross, H., Noakes, J.E., Spaulding, J.D. (Eds.), Liquid Scintillation Counting and Organic Scintillators. Lewis Publishers, Chelsea, MI, pp. 529–535.

Scarpitta, S., Odin-McCabe, J., Gaschott, R., Meier, A., Klug, E., 1999. Comparison of four ^{90}Sr groundwater analytical methods. Health Phys. 76 (6), 644–656.

Shoner, S., Matthews, D., Zheng, M., Appelbaum, F., 1999. Comparative biodistribution of yttrium-90 and iodine-131 labeled anti-CD45 antibody in a mouse model. J. Labelled Compd. Radiopharm. 42 (Supp/1), S747.

Sundaresan, M.K., 2001. Handbook of Particle Physics. CRC Press, Boca Raton, p. 81.

Takiue, M., 1980. Simple and rapid measurement of α-rays on smear samples using air luminescence. Health Phys. 39, 29–32.

Takiue, M., Ishikawa, H., 1979. α-ray measurement due to air luminescence employing a liquid scintillation spectrometer. Nucl. Instrum. Methods 159, 139–143.

Watanabe, N., Sawai, H., Endo, K., Shinozuka, K., 1999. Labeling of phosphorothioate antisense oligonucleotides with yttrium-90. Nucl. Med. Biol. 26 (2), 239–243.

Wike, J.S., Guyer, C.E., Ramey, D.W., Phillips, B.P., 1990. Chemistry for commercial scale production of yttrium-90 for medical research. Appl. Radiat. Isot. 41 (9), 861–865.

Wiseman, G.A., White, C.A., Stabin, M., Dunn, W.L., Erwin, W., Dahlbom, M., Raubitschek, A., Karvelis, K., Schultheiss, T., Witzig, T.E., Belanger, R., Spies, S., Silverman, D.H., Berlfein, J.R., Ding, E., Grillo-Lopez, A.J., 2000. Phase I/II ^{90}Y-Zevalin (yttrium-90 ibritumomab Tiuxetan IDEC-Y2B8) radioimmunotherapy dosimetry results in relapsed or refractory non-Hodgkin's lymphoma. Eur. J. Nucl. Med. 27 (7), 766–777.