Cherenkov Counting Efficiencies for β^--Emitters in Dry State in Glass Vials

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Cherenkov counting efficiencies for standardized β^--emitters in the dry state at the centre of air-filled glass vials were measured with a liquid scintillation spectrometer. Cherenkov counting efficiencies, which are plotted as a function of the average energy of β^--particles and the internal conversion electrons, give a straight line on log–log scale.

The use of Cherenkov radiation in determining radionuclides has a significant advantage that the sample radionuclide, which was measured by Cherenkov counting, is often suitable for any other experiments. A number of radionuclides have been measured and standardized by this counting technique. It should be emphasized that most of those radionuclides were measured in aqueous[1,2] or in non-aqueous media,[3,4] and that only a few Cherenkov counting efficiencies for radionuclides in dry state have been previously reported.[5]

In the course of study concerning standardization of ^{222}Rn by liquid scintillation counting, however, we observed that β^--particles from short-lived ^{222}Rn daughters (^{214}Pb and ^{214}Bi), which were distributed in the air space of the counting vial, gave appreciable Cherenkov radiation by interacting with the wall of the vial. In order to measure Cherenkov counting efficiencies for ^{214}Pb and ^{214}Bi, we determined those for standard pure β^--emitters and β^--γ-emitters as a function of the average energy of β^--particles and the internal conversion electrons. Based on the results obtained, we estimated Cherenkov counting efficiencies for ^{214}Pb and ^{214}Bi. Although most of the Cherenkov radiation observed in this study was produced by β^--particles in the glass wall of the counting vial, the Cherenkov counting efficiencies obtained were compared with those due to γ-rays. We have included a brief discussion of the data obtained by similar experiments.

The standard β^--sources were prepared as follows. About 0.05 mL aliquots of ^{32}P solution ($Na_2H^{32}PO_4$ in 0.1 M HCl) were spotted on a 0.65 µm-pore cellulose nitrate membrane filter paper (Advantec Toyo Co., Ltd., Tokyo) of 10 mm diameter and air-dried at room temperature. Upon completion of the measurement of Cherenkov radiation in the manner described below, a few drops of water and 2 mL of 2-methoxyethanol were added to the ^{32}P sample in the counting vial and stirred carefully for 2 min. Then, 10 mL of ACS II (Amersham Corp., Chicago, IL, USA) was added to the aqueous solution of ^{32}P. The disintegration rate of ^{32}P was determined by the modified integral counting method.[6]

Samples of ^{36}Cl, ^{60}Co and ^{137}Cs were prepared in a similar manner as described in the sample preparation of ^{32}P. Solutions used for sample preparation were: ^{36}Cl ($Na^{36}Cl$ in H_2O), ^{60}Co ($^{60}CoCl_2$ in 0.1 M HCl) and ^{137}Cs ($^{137}CsCl$ in 0.1 M HCl). The disintegration rate of ^{36}Cl sample was determined in a similar manner as described in the case of ^{32}P. ^{60}Co and ^{137}Cs samples were standardized by measuring γ-rays from these nuclides with an intrinsic Ge detector (Princeton Gamma-Tech Inc.) coupled to a 4096-channel analyzer, which was absolutely calibrated using a set of LMRI (Laboratoire de Metrologie des Rayonnements Ionisants, Saclay, France) γ-ray standard sources.

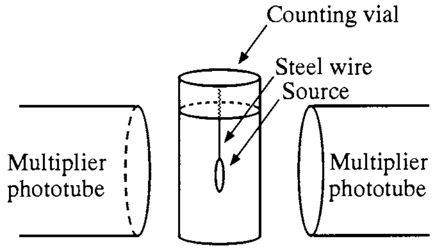

Figure 1. The experimental arrangement for measuring Cherenkov counting efficiency.

Samples of ^{32}P, ^{36}Cl, ^{60}Co and ^{137}Cs which were paralleled to the surfaces of PMTs at the center of the counting vials as shown in Figure 1, and measured with an Aloka liquid scintillation spectrometer, Model LSC-3500 (Aloka Co. Ltd. Tokyo, Japan) for 10 min in the coincidence mode. Most of the Cherenkov radiation is in the ultraviolet wavelength region. However, in spite of this, it can be measured with liquid scintillation spectrometers, because a typical spectral response of the current bialkali multiplier phototube is in the wavelength range of 185–650 nm.

The Cherenkov counting efficiencies for pure β^--emitters are usually expressed percent of the activities for the pure β^--emitters. However, in order to express the Cherenkov counting efficiencies for a variety of the radionuclides, the Cherenkov counting efficiencies were expressed percent of emission rates of the β^--particles and the internal conversion electrons in the present study.

Table 1 lists the average energy of β^--particles and the internal conversion electrons with the absolute intensity for the nuclides used in the present study. The energy and the absolute intensity of the internal conversion K-shell electrons, e_K, and the internal conversion L-shell electrons, e_L, from ^{137m}Ba were: e_K (0.624 MeV, 9.16%) and e_L (0.655 MeV, 1.73%).

Since the sum of the absolute intensity for the internal conversion M-, and N-shell electrons, e_M and e_N from ^{137m}Ba is estimated to be ca. 0.29%, e_M and e_N were ignored in the calculation of the Cherenkov counting efficiency. Similarly, the internal conversion electrons from ^{60}Co were also ignored, e_K/γ values are 1.736×10^{-4} for 1.173 MeV γ-rays and 1.294×10^{-4} for 1.332 MeV γ-rays.

Table 1. Cherenkov counting efficiency for the standard source

Source	Av. energy / MeV	Absolute intensity / %	Emission rate / s^{-1}	Counting efficiency / %
^{32}P	β^- : 0.695	100	2114 ± 3	38.8 ± 0.1
^{36}Cl	β^- : 0.2514	98.1	1841 ± 3	5.2 ± 0.03
^{60}Co	β^- : 0.0966	100	3416 ± 96	0.69 ± 0.02
^{137}Cs	β^- : 0.187	100		
	e_K : 0.624	7.78		
	e_L : 0.655	1.47		
	(Av. 0.2245^a)		1254 ± 32^a	4.98 ± 0.13^a

[a] For β^-- particles and internal conversion electrons.

On the other hand, glass wall of the counting vial and glass components of the detection system are exposed to γ-rays, Cherenkov radiation may be produced, if the energy of photoelectron and Compton electrons due to γ-rays is greater than the threshold energy of a medium. Therefore, we covered standardized sample of ^{60}Co and ^{137}Cs with plastic plates which have sufficient thickness to absorb β^--particles from the samples, placed at the center of the vial, and measured for 10 min with the liquid scintillation spectrometer. The Cherenkov counting efficiencies due to γ-rays, which are expressed percent of emission rates of the photons, were: 0.02 ± 0.004% for ^{137}Cs, 0.12 ± 0.004% for ^{60}Co. Based on the results and the fact that an approximate correlation exists between Cherenkov counting efficiency and logarithm of γ-ray,[7] Cherenkov counting efficiencies due to γ-rays for ^{214}Pb and ^{214}Bi were estimated to be 0.003 ± 0.0006% and 0.09 ± 0.003%, respectively. Because the average energy of Bremsstrahlungs due to the β^--particles measured in this study are less than 118 keV, and because Cherenkov counting efficiencies of which are less than 1.5×10^{-4}%, effect of Bremsstrahlungs on the Cherenkov counting efficiency can be neglected. Details of which will be reported elsewhere.

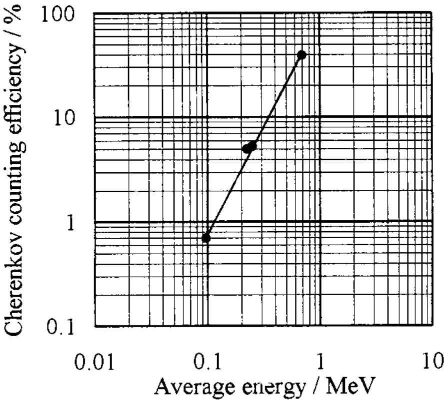

Figure 2. Cherenkov counting efficiencies as a function of the average energy of β^- – particles and the internal conversion electrons.

The Cherenkov counting efficiencies, which were corrected for those due to γ-rays and plotted as functions of the average energy of β^--particles and the internal conversion electrons (Table1), gave a straight line on log–log scale (Figure 2). The equation fitted to the experimental data, which calculated by the least squares method, is given by

$$\log Y = 2.039 \log X + 1.911$$

where Y is the Cherenkov counting efficiency expressed in percent and X is the average energy expressed in MeV.

Thus, Cherenkov counting efficiencies are estimated: 3.7 ± 0.2% for ^{214}Pb (av β^- 0.220 MeV) and 32.9 ± 1.3% for ^{214}Bi (av β^- 0.641 MeV). In quantitative determination of energetic β^--emitters, therefore, accurate correction for the Cherenkov radiation is essential.

The precision was calculated as the sum of the maximum estimates of probable errors. The principal errors are due to: uncertainties of standard source (< ±2.0%), standard statistical error due to measurement of γ-rays (< ±2.0%), standard statistical error due to measurement of Cherenkov radiation (< ±1.0%). Thus the total error adds up to ca. 3.0%. Cherenkov counting efficiencies were reproducible to ca. ± 4.0% on repeated runs. We took the nuclear data used from the reference work by Lederer and Shirley.[8]

There is a fairly large difference between the reported Cherenkov counting efficiencies for ^{36}Cl (6.6%[2]), ^{60}Co (5.6%[2]), ^{137}Cs (4.8%[2]) and ^{32}P (46.8% [2], 57%[1]) and those obtained in the present study (Table 1). This is mainly due to the fact that those radionuclides in earlier experiments were measured in aqueous media. The most comparable work is the determination by Berger,[5] who reported a 25% counting efficiency of Cherenkov radiation from ^{32}P on dry glass filters placed on the bottom of air-filled glass vials. However, it is readily apparent that the difference in physical state and geometry of the samples would introduce the discrepancy between the data by Berger and those obtained in the present study.

Before concluding, it is interesting to note that energetic β^--emitters which are present in air (i.e., ^{214}Pb and ^{214}Bi) are suggested to be determined by Cherenkov counting using liquid scintillation spectrometers.

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