OPTIMIZATION OF COUNTING CONDITIONS FOR CERENKOV RADIATION BY LSC

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ABSTRACT. We present the results of a search for a better medium than water for Cerenkov radiation measurements. Some liquids and solid materials with coefficients of refraction *n* higher than water were examined, but without a clearly successful result. However, we found that the relative counting efficiency for Cerenkov radiation seems to increase with energy faster than in a linear way.

INTRODUCTION

Cerenkov radiation is produced when a charged particle moves in matter with a speed greater than the speed of light for a given medium. In the case of electrons moving in water, the minimum kinetic energy for the effect (a threshold) is 260 keV (Elrik and Parker 1968). Cerenkov radiation in water or dilute acid solutions has been used for many years to determine beta activity in environmental samples (Elrik and Parker 1968; Francois 1973). Since the effectiveness of photon production is very low, just above the threshold, the method is considered suitable for radionuclides emitting electrons with a maximum energy above 500 keV (Francois 1973). The main advantages of the method are that samples are in a convenient form that enables further processing, have easily disposable organic wastes, exhibit no chemical quenching, and are low in cost. The disadvantages are that the spectrum information is lost, the method is applicable only to high-energy beta emitters, and the counting efficiency is usually much lower when compared to scintillation mode with a cocktail. The aim of this work is to optimize measuring conditions for the Cerenkov detection of beta emitters by searching for conditions that result in a higher efficiency.

EXPERIMENT

A 1414-003 Wallac Guardian liquid scintillation counting (LSC) spectrometer and standard 15-mL LSC diffuse (polyethylene) vials were used in our experiment. The main radioactive source was a solution of ^{90}Sr equilibrated with its daughter ^{90}Y. Some tests were also done with separated ^{90}Sr and ^{90}Y, as well as some others with ^{90}Sr during ingrowth of ^{90}Y. In each case, as reference we used an appropriate radioactive spike (i.e. a mixture of ^{90}Sr and ^{90}Y or each radionuclide separately) in 2 mL of 1M HNO_3 mixed with 10 mL of Wallac HiSafe 3 liquid scintillation (LS) cocktail. Different media were used in our tests for Cerenkov radiation. Besides pure liquids like water, CS_2, TBP, and ethyl acetate (pure and with about 3 g of dissolved transparent polystyrene), we also used solids: 1) Plexiglas® rods (3 mm diameter and 5 cm long) immersed in water inside 15-mL LSC vials, and 2) larger Plexiglas rods of similar length but of 15 mm diameter with a drilled well (3 mm diameter) where microliter amounts of radioactive solutions were placed.

^{90}Y was separated from ^{90}Sr using a chromatographic column filled with Sr-resin (Horwitz et al. 1992). A 3M HNO_3 solution of ^{90}Sr and ^{90}Y in radioactive equilibrium was passed through the column. Yttrium eluted from the column without retention, whereas strontium was retained by the resin. The column was then treated as a ^{90}Y generator. Yttrium was stripped off using 20 mL of 8M

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HNO_3 after the 2-week periods needed for its ingrowth. The purity of each ^{90}Y separation was checked every time by means of tracing the half-life of its decay.

Besides this study on efficiency, an approximate spectrometer energy calibration (for cocktail-added scintillation mode) was done by comparing the approximate channel number of the upper limit of each beta spectrum with the known maximum energy for beta radiation for that nuclide. The radionuclides used, their energies, and the approximate channel of the end of spectrum are presented in Table 1. In Figure 1, the plot of the calibration line obtained and the corresponding equation are presented. The equation is a way to trace the Cerenkov efficiency as a function of the mean beta energy of a mixture of ^{90}Sr and ^{90}Y during the ingrowth of the latter.

Table 1 Data used for the approximate energetic calibration of the LS spectrometer.

Isotope	E_{max} (keV)	Channel number
^{241}Pu	21.6	260
^{14}C	156	470
^{90}Sr	540	600
^{32}P	1700	740
^{90}Y	2270	800

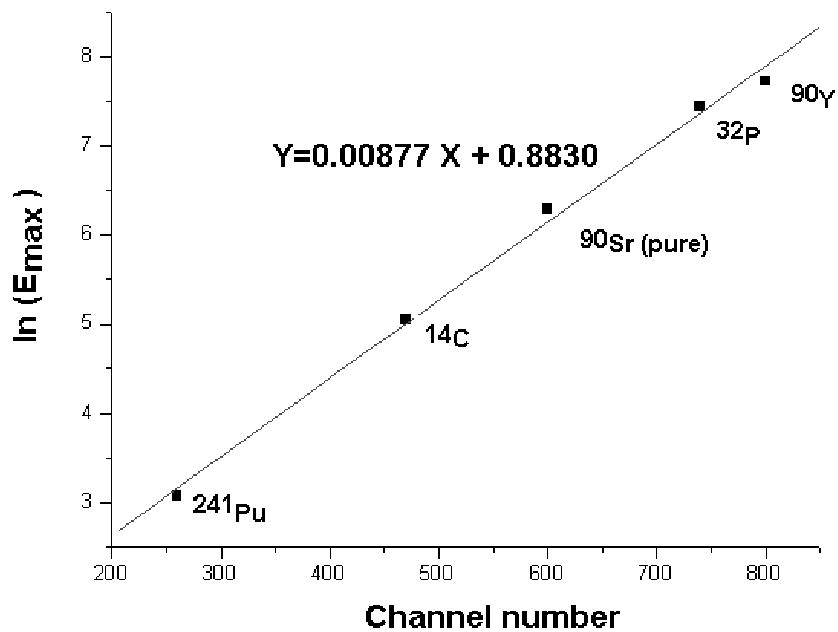

Figure 1 Approximate energetic calibration for Wallac Guardian spectrometer (E_{max} expressed in keV).

Coefficients of refraction for liquids were determined using an Abbe refractometer. For mixtures like Plexiglas rods immersed in water, the effective coefficient of refraction was calculated as a weighted (by mass factor) mean of 2 appropriate coefficients.

RESULTS

It was found that the relative (compared to standard LSC measurement) Cerenkov counting efficiency was almost constant in a broad range from 1 to 10 mL of a given solution within the 15-mL LSC vial. Efficiency increases from 27% for 1 mL to 36% for 10 mL of water (see Figure 2).

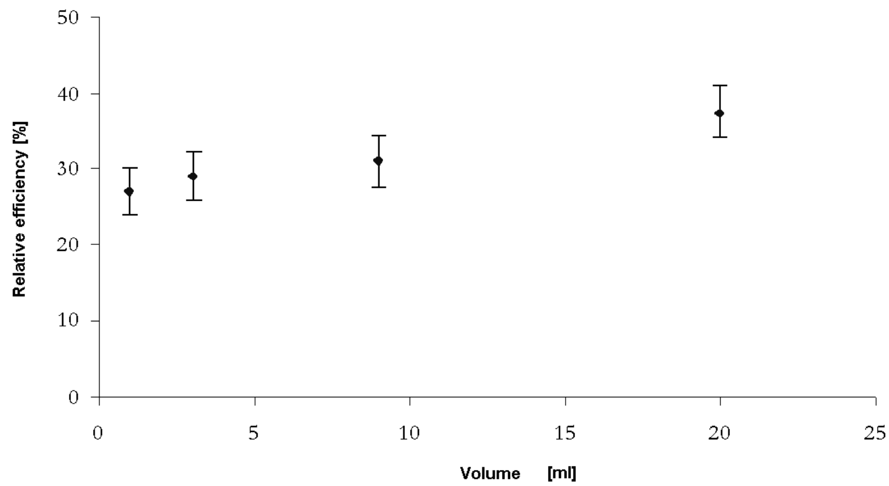

Figure 2 Relative efficiency for Cerenkov radiation as the function of LSC vial volume (for equilibrated ^{90}Sr-^{90}Y solution in water).

The dependence of the counting efficiency on the refraction coefficient *n* of the given medium and the average energy of beta particles was checked. The results are presented in Figure 3. Polystyrene dissolved in ethyl acetate or polystyrene elements immersed only in water produced a similar or slightly higher efficiency compared to pure water. This is perhaps due to some of the scintillation properties of polystyrene (Elrick and Parker 1968).

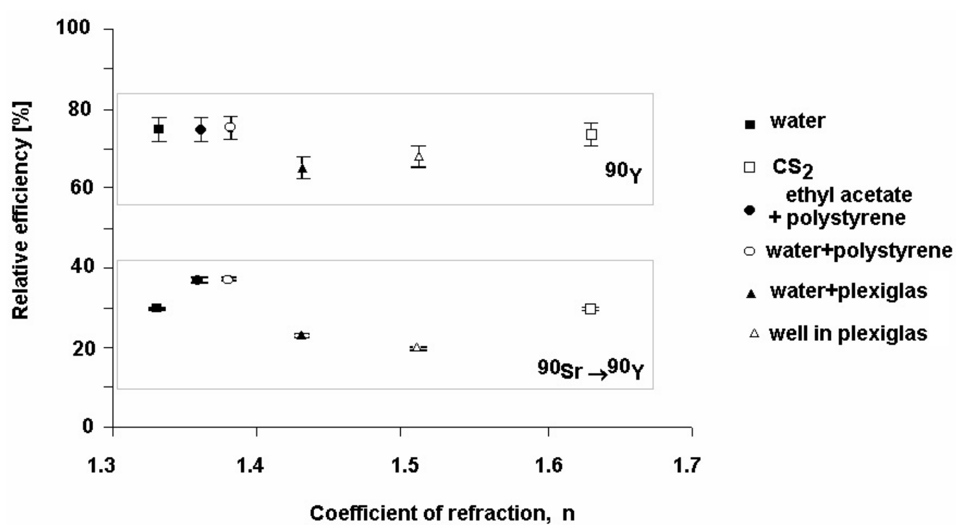

Figure 3 Relative efficiency for Cerenkov counting for different materials as a function of the refraction coefficient *n* for equilibrated ^{90}Sr-^{90}Y and pure ^{90}Y.

The Cerenkov radiation for all media examined was always detected by the spectrometer. It was not so obvious a priori, since the frequency of Cerenkov radiation (which is almost monochromatic just above the threshold, making the band wider with the increase of energy) is determined by the position of an absorption band for a given material (Jackson 1975). The resulting counting efficiency is strongly affected by the position of this band versus the frequency sensitivity characteristic of the phototube. During our studies, it appeared that organic liquids like TBP or CS_2, although they have high values of *n*, are not so suitable. This is likely due to their high chemiluminescence properties, which seem to interfere with the counting rate in a randomized way. The lack of efficiency increase for materials of higher *n* shows that the positions of absorption bands for these materials apparently do not fit well with the characteristics of the phototube.

The Cerenkov efficiency for water was studied as a function of its mean beta energy. Different mean energies of beta radiation were obtained by 1) using a spike of pure ^{90}Sr and measuring it several times during the ingrowth of ^{90}Y up to the secular equilibrium level, and 2) by using pure ^{90}Y spikes. The observed relation for Cerenkov counting efficiency as a function of mean beta energy is displayed in Figure 4. It seems that this efficiency increases in a non-linear way (perhaps parabolic), reaching more than 70% (for water) of the efficiency of cocktail scintillation counting for mean beta energies of about 0.8 MeV (for pure, separated ^{90}Y).

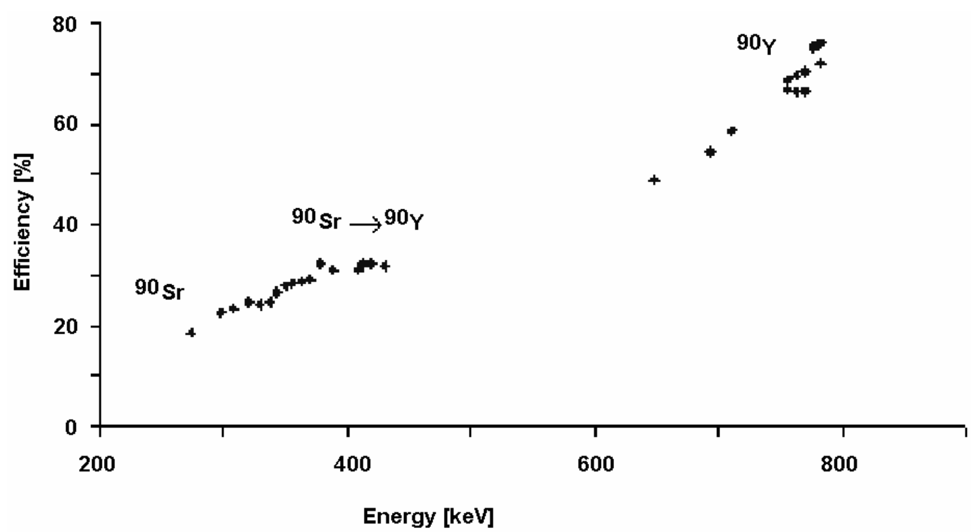

Figure 4 The counting efficiency for Cerenkov radiation as a function of the mean energy of beta radiation

CONCLUSIONS

Relative counting efficiency for Cerenkov radiation increases with the mean energy of beta radiation faster than in a linear way. Although some high *n* materials were examined, unfortunately no material was much better than water when used as a medium for beta measurements by means of Cerenkov radiation. However, polystyrene dissolved in ethyl acetate and pieces of polystyrene immersed in water showed a slightly higher counting efficiency than that of water, at least for an equilibrated strontium-yttrium source. Some improvements can perhaps be achieved in the future after introducing wavelength shifters, but the simplicity of the system will be lost.

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