A COMBINED LSC/NaI SYSTEM FOR LOW-LEVEL ENVIRONMENTAL MEASUREMENTS

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ABSTRACT. We describe a novel compact multisample system, Kvartett, with four liquid scintillation detectors combined with a large NaI crystal unit for low-level measurements of pure beta and beta/gamma emitters. The sample vials of the four tightly packed liquid scintillation units, each with a single photomultiplier tube (PMT), are inside the inverted well of a large NaI crystal, which serves as an anticoincidence detector when β-active samples are measured, or as a γ detector for β/γ activity. In the latter case, the NaI pulse-height spectrum is recorded only for pulses that are in coincidence with pulses from each of the four liquid scintillation detectors, which reduces the γ background count rate by a factor of 400–600. The Kvartett offers higher counting efficiency than germanium crystals and lower or similar background. The disadvantage of this method is that it calls for chemical separation of the sample. A simpler version of Kvartett for pure β emitters uses a flat Geiger guard counter. Kvartett will be useful in environmental control, measurement of cosmogenic nuclides and various other related work.

INTRODUCTION

Weak beta- and gamma-emitting samples need long counting times and, often, the collection and processing of a large sample volume. Low-level counting systems used in such work should have high counting efficiency, low background and allow long counting times. Price and space requirements should also be considered. Earlier developmental work on low-level counting systems emphasized reducing background count rate and increasing counting efficiency. Multidetector systems that provide longer counting times became a practical possibility with cheaper electronics in the early 1970s. The system presented here, Kvartett, is the result of a transfer of the multidetector concept to a system with liquid scintillation (LS) detectors.

Initially, our system was intended only for the measurement of β activity, especially ^{14}C, but it also turned out to be a very sensitive γ-detection system. Germanium crystals have now almost completely replaced NaI detectors in measuring weak γ-active samples, despite their low detection efficiency, higher price and need for a continuous supply of liquid nitrogen. The advantage of the Ge crystals is their high energy resolution, which allows separation of γ lines of similar energy. In low-level measurements, high resolution of the Ge crystal is usually unimportant when a specific nuclide is being measured, except for the lower background it gives, because a narrow energy window is counted. Our unit employs a NaI detector, which has a much higher γ-counting efficiency compared to the Ge crystal.

LOW-LEVEL LIQUID SCINTILLATION COUNTING

The early liquid scintillation counting (LSC) systems used a single photomultiplier tube (PMT) for detecting the weak scintillations of the sample. An important improvement that lowered the background and increased the stability was the introduction of the two-PMT coincidence system. LSC systems soon were preferred in tracer work with pure β emitters. As most biologically important elements, such as carbon, hydrogen, phosphorus and sulphur, have only β-emitting isotopes, the need for these systems grew rapidly, and they were developed to a high technical standard. Inexpensive commercial LSC systems were applied to ^{14}C dating early in the 1960s, and dominated in new dating laboratories since the early 1970s, despite their somewhat higher back-

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ground compared to gas proportional counters. The LSC systems usually were used unmodified, but in some cases, lead and, in rare cases, an anticoincidence counter was added to the shield.

Commercial LSC systems were developed to serve the large tracer market, where background was usually not important. For a long time, commercial systems were not available for low-level measurements. However, some academic institutions promoted low-level LSC systems. Punning and Rajamae (1977) developed both single-PMT and two-tube LSC coincidence systems, with heavy shields and guard counters; this development was only moderately successful. At the 1983 Banff LSC conference, Pei-yun and Ting-kui (1984) presented an attractive system with a single PMT and a remarkably low background, long after single-PMT systems had been practically abandoned in LSC.

A breakthrough came in 1985 with the introduction of the low-level Wallac Quantulus system, developed through the initiative of Henry Polach. It has a rather thick lead shield, but its most important feature is the annular LS guard counter surrounding the conventional coincidence detector system. Packard has also introduced an attractive LSC system for low-level counting that is a modification of their conventional systems. It is simpler and cheaper than the Quantulus, but with a somewhat higher background. Their design reduced the background through analysis of pulses that follow within a μsec of the main pulse, and by addition of a plastic guard tube around the vial for detecting cosmic-ray particles (Noakes & Valenta 1989).

In radiocarbon dating, as in other aspects of low-level measurement, the counting capacity, rather than the background count rate, is the limiting factor in accuracy (Theodórsson 1991a). Increasing the counting capacity through multidetector systems will be an important improvement.

MULTIDETECTOR SYSTEMS

In the past, the cost of the electronic apparatus represented a substantial part of the total cost of low-level systems. The introduction, in the early 1970s, of integrated circuits and microprocessors changed this radically. This led Theodórsson, in 1973, to propose a multicounter system with 5–10 Geiger detector elements in a single acrylic plate for the measurement of weak solid β-active samples. The system was developed jointly by the Science Institute of the University of Iceland and the Risö Research Centre in Denmark (Bötter-Jensen, Hansen & Theodórsson 1977; Theodórsson 1987).

Multicounter systems with gas-proportional sample detectors were developed for ^{14}C dating in Heidelberg (Schoch *et al.* 1980) and in Groningen (Tans *et al.* 1982), and demonstrated convincingly the advantage of the multidetector arrangement. Thus, it was a challenging task to adopt this powerful technique to low-level LSC. Theodórsson (1985) proposed such a system and reported its progress at the 13th International Radiocarbon Conference (Einarsson & Theodórsson 1989). Einarsson (1992) constructed and tested a multidetector LSC system for four samples.

COSMIC-RAY BACKGROUND REDUCTION

Cosmic rays are the source of much of the background count rate of most β- and γ-radiation detectors. This component is reduced with a thick shield of lead or iron and an anticoincidence (guard) detector system inside the main shield. Gas counters are generally used as guard detectors. They suppress primarily cosmic-ray pulses caused by direct hits of muons or protons in the sample detector, but have little effect on the contribution of the secondary radiation created by these particles in the shield (Theodórsson & Heusser 1991). Otlet *et al.* (1983) demonstrated a very high background reduction in a system with small gas-proportional counters inside a 10-cm-thick annular

NaI crystal guard counter. This background reduction was demonstrated further on gas-proportional counters studied under different shielding conditions in four low-level laboratories (Loosli, Forster & Otlet 1986), which confirmed the superiority of the NaI guard detector. It clearly eliminated a major part of the cosmic-ray-generated secondary radiation.

This large background reduction can be explained by the high γ-detection efficiency of the NaI crystal. A substantial part of the residual background of most low-level β- and γ-counting systems comes from secondary γ-ray showers produced by muons and protons in the main shield. The NaI crystal must detect only one γ photon of this shower to eliminate contribution to the anticoincidence channel of the system. The γ-detection efficiency of conventional anticoincidence gas counters is too low to reduce this component appreciably. Thus, we chose a large well-type NaI scintillation unit as a guard detector in our multidetector system.

KVARTETT, A NEW MULTIDETECTOR LSC SYSTEM

Kvartett has realized the goal of a simple and compact multidetector LSC system with low background, based on the single-PMT arrangement, intended primarily for ^{14}C dating. In conventional two-PMT systems, the coincidence technique reduces much of the phototube background contribution. In a single-PMT system, reduction of these pulses is more difficult. First, one must select a phototube with a low dark current and high quantum efficiency. High light-collection efficiency is also important to enhance the scintillation pulses from the sample compared to the phototube background pulses, so that the latter will be eliminated more effectively by the low-level discriminator. One should also select a scintillator with high light output. We were able to reach quite a low ^{14}C background using this technique.

Einarsson (1992) described Kvartett and its application to ^{14}C dating. We briefly describe the system here and emphasize its application to measuring other β- and β/γ-emitting samples. The core of the detection system is a compact unit with four LS detectors where a single PMT views the scintillations from each vial (Fig. 1). Use of 29-mm-diameter tubes, instead of 50-mm tubes used generally in LSC systems, reduces the size and weight of the system. An anticoincidence detector reduces cosmic-ray background contribution. There are two versions of the system. The simpler one, Kvartett/GM, is designed to measure pure β emitters, where very low background is less important. It has a single, flat Geiger guard counter on the top of the lead shield. The other version, Kvartett/NaI, has a large NaI well crystal guard counter, inside of which are the sample vials (Fig. 1). This can also be used as an ultra-low-level counting system for β/γ emitters, as discussed below. Both systems are shielded by 5 cm of lead. The total weight of the shield and detector system is ~300 kg for Kvartett/GM, and 500 kg for Kvartett/NaI. The sample volume may be either 3 or 6 ml.

MEASURING BETA EMITTERS WITH KVARTETT

Although Kvartett was designed primarily for ^{14}C dating, it soon became evident that the system could be used for other low-level measurements. The most important pure β-emitting nuclides in environmental studies are ^{14}C, ^{90}Sr and ^{99}Tc, which, until recently, have been measured commonly by low-level Geiger or proportional counters with thin windows. ^{137}Cs can also be studied, as γ emissions are delayed with respect to β particles (Fig. 2). With the advent of better low-level LSC systems, these β emitters have increasingly been measured with LS counters because of their higher detection efficiency. A further advantage of the LSC system, compared to Geiger and proportional counters, is that the LS pulse-height spectra can check the reliability of the final result.

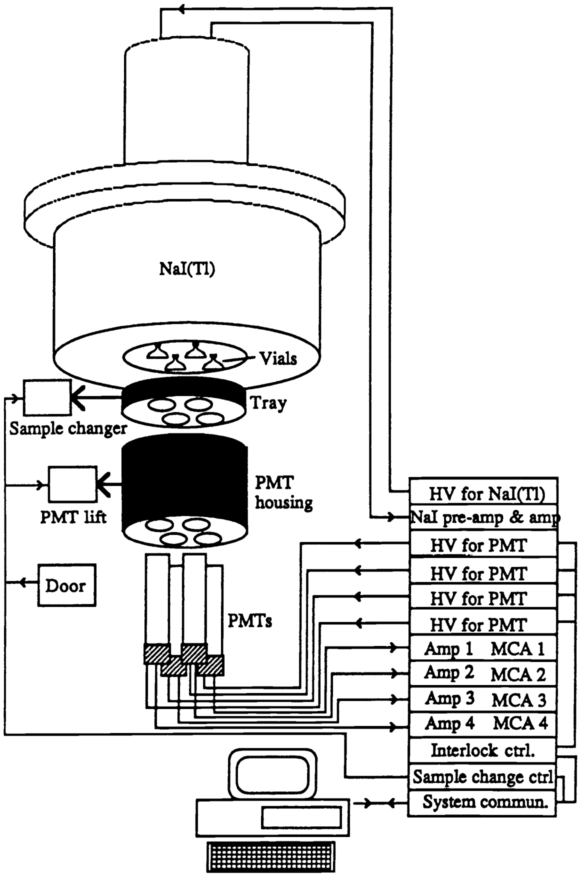

Fig. 1. Schematic diagram of Kvartett

The Kvartett, with its high counting capacity and low background, is ideal for environmental studies. To measure ^{90}Sr (usually through its daughter, ^{90}Y), ^{99}Tc and ^{137}Cs, they must first be separated chemically, which usually yields a sample in aqueous solution. A scintillation cocktail that can incorporate high water content with the highest possible light efficiency and least quenching should be used. Organic solutions of the sample will yield still better results.

MEASURING GAMMA EMITTERS WITH KVARTETT/NaI

To detect β/γ-emitting isotopes in Kvartett, the NaI crystal is used as the main detector, which sends pulses to a multichannel analyzer. When a pulse above a given threshold occurs in one of the four LS detectors, a pulse is sent the corresponding coincidence signal input of the analyzer, splitting the NaI spectrum into eight subspectra, a coincidence and anticoincidence spectrum for each sample. This coincidence technique decreases the background of the NaI crystal by a factor of about 400–600 in the energy range of 500–1500 keV.

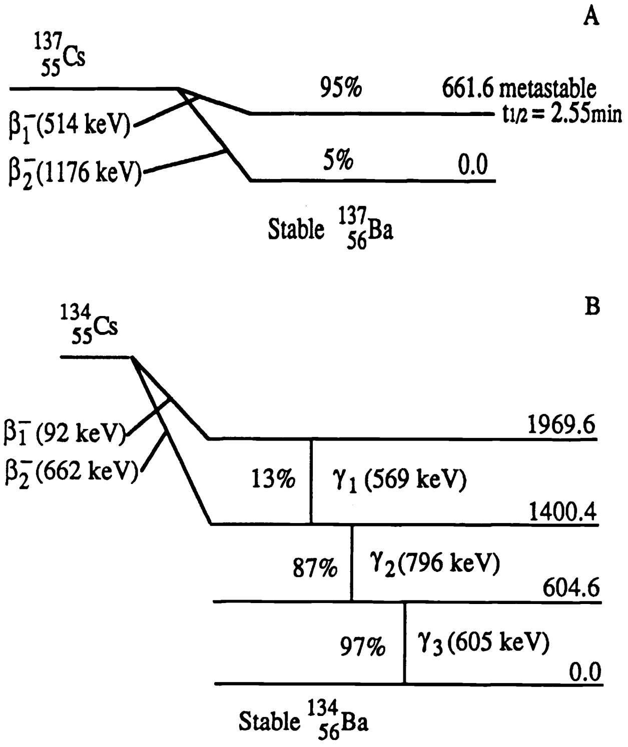

Fig. 2. A. The decay scheme of ^{134}Cs; 10% of the 662 keV γ are lost by internal conversion emitting 625 keV electrons. B. The decay scheme of ^{137}Cs; only the contributing γ lines are shown in the diagram.

BACKGROUND AND COUNTING EFFICIENCY OF KVARTETT

Measurements of ^{137}Cs and ^{134}Cs standard samples illustrate the performance of Kvartett for β and β/γ emitters. Figure 2 shows the decay scheme of these nuclides. For our study, we used a 0.7-ml aqueous standard solution with 2.3 ml of Opti-Fluor®. One can determine the concentration of ^{137}Cs in three ways, by measuring:

1. The total β spectrum in the LS detector in the energy range below 500 keV
2. The 624-keV internal conversion line in the LS detector
3. The coincidences between the 624-keV internal conversion line and the accompanying 33-keV X-ray line in the NaI crystal. This gives very low background but poor counting efficiency, and, thus, is more of academic interest.

Figure 3A shows the LS anticoincidence background spectrum, and Figure 3B, the spectrum of a standard sample with about 2.5 Bq of ^{137}Cs. Figure 4A shows the total background spectrum of the 15-cm-diameter NaI crystal of Kvartett, and Figure 4B, the part of this spectrum that is in coincidence with pulses from one of the LS detectors. The coincidence arrangement reduces the background count rate of the crystal by a factor of 400–600 in the 500–1500 keV range. Figure 5 shows the coincidence NaI spectrum with 7 Bq of ^{134}Cs in the LS detector. This spectrum shows the two main γ lines of ^{134}Cs, 604 keV and 796 keV, as well as the sum line.

Table 1 compares the counting efficiency (counts/disintegration) and background for ^{134}Cs and ^{137}Cs in Kvartett with the Ge-crystal system (27% relative efficiency) of the State Radiation Control Institute.

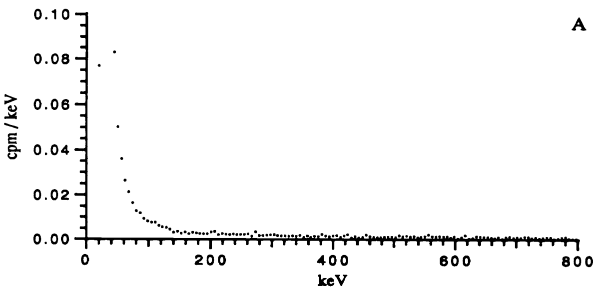

Fig. 3A. The LS anticoincidence background spectrum of Kvartett/NaI. Sample: 0.7 ml water + 2.3 ml Opti-Fluor®

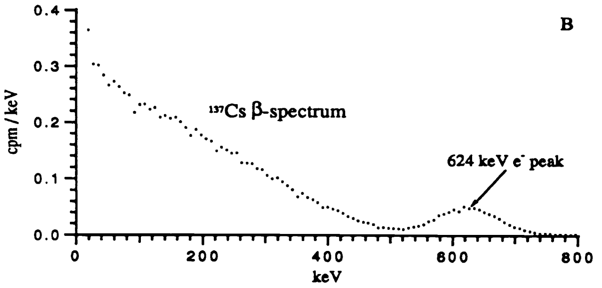

Fig. 3B. ^{137}Cs LS anti-coincidence spectrum of Kvartett/NaI. Sample: 2.5 Bq in 0.7 ml water + 2.3 ml Opti-Fluor®

TABLE 1. Comparison of Kvartett/NaI and a Ge Detector in ^{134}Cs and ^{137}Cs Measurements

	Kvartett		Ge crystal	
Energy (keV)	Detection sensitivity(c/dis)* (%)	Background (cpm)	Detection sensitivity(c/dis) (%)	Background (cpm)
^{134}Cs				
604, γ	11.0	0.13	2.2	0.6
790, γ	9.4	0.09	1.7	0.5
Sum line	4.4	0.06		
Total in peaks	24.9	0.28	3.9	1.1
Total spectrum	59.9	1.81		
^{137}Cs				
140–530, β	54	0.7		
530–765, e-peak	8.5	0.25		
664, γ			2.0	0.5

*c/dis = counts/disintegration

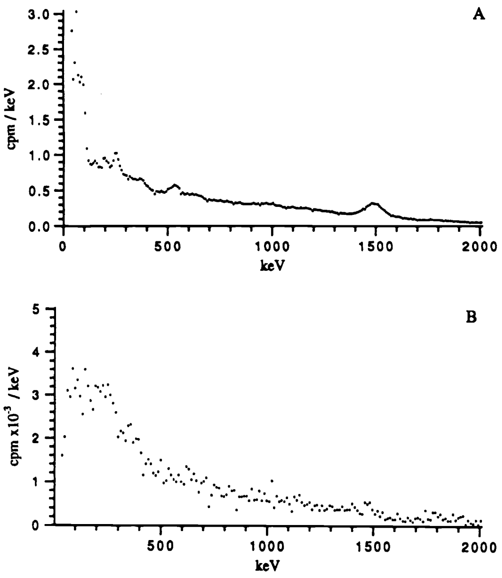

Fig. 4. Kvartett/NaI γ background. A. Total spectrum. B. Coincidence spectrum

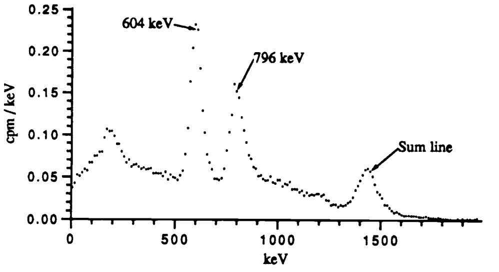

Fig. 5. Kvartett/NaI ^{134}Cs-coincidence spectrum. Sample: 7 Bq in 0.7 ml water + 2.3 ml Opti-Fluor®

DISCUSSION AND CONCLUSION

We have studied the possibility of monitoring ^{137}Cs in the ocean environment with Kvartett. Typical ^{137}Cs concentration in the sea around Iceland is ~3 Bq m^{-3}. A 100-liter sea sample with this ^{137}Cs concentration co-precipitated with AMP and counted for 24 h in the Ge system will give a precision of 7%. Using the Kvartett for these measurements can reduce considerably the sample size because of higher detection efficiency. A sample size of 20 liters, which is enough for Kvartett, is much easier to handle and process, but a subsequent chemical separation of Cs from AMP is necessary. With the same counting time as for the Ge crystal (24 h) and measurement over the total LS spectrum, Kvartett yields a precision of 2.2% for the 20-liter sample (and four samples can be measured simultaneously). We obtained a similar increase in sensitivity and precision in ^{134}Cs measurement.

Kvartett is a useful addition to the family of systems for the measurement of low-level β and β/γ activity. We have reported the first results of measurements with the prototype of Kvartett/NaI. Much work is needed to exploit the system's full potential. We expect that the γ background can be decreased by an order of magnitude by increasing the thickness of the lead shield to 10 cm, and by adding an external guard counter system, as suggested by Theodórsson (1991b).

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