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Conference internationale sur les tendances nouvelles de l'analyse par activation, Saclay 2-6 octobre 1972

The Relative Sensitivity and Accuracy of NaI(Tl) and Ge(Li) Detectors for Minor Components

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A mixture of 233Pa, 140La, 82_{Br} , 64_{Cu} , 72_{Ga} , 60_{Co} , and 24_{Na} of known disintegration rates was counted on a NaI(Tl) and a large Ge(Li) detector. Both systems detected 233Pa when present at 0.2%, and accurate results were observed for 233Pa, 140La, and ⁶⁰Co when present at greater than 0.5% of the total disintegration rate. The accuracy of the results from either system was not significantly different since the mean ratio of the observed to true results was 0.99 ± 0.05 and 1.06 ± 0.06 , respectively. However, equal sensitivity and accuracy were achieved only when the counting interval for the germanium system was 1000 min, or 60 to 600 times that for sodium iodide.

Both sodium iodide and germanium detectors have been used in trace element analysis by the neutron activation method. A rigorous theoretical comparison of these two detector systems would require an evaluation of 1) the relative absolute counting efficiency, 2) resolution, 3) the accuracy and sensitivity of the computational method used to resolve the resultant complex spectra, and 4) the number of counts recorded in a reasonable counting interval from a practical irradiation interval.

A common assumption is that the superior resolution of the germanium detector is always sufficient to make it the better system. However, experimental evaluation of these two systems has shown that sodium fodide may be the system of choice under some circumstances. Voigt, et al. used the peak area computational method and evaluated the precision and sensitivity of the two detector systems for four different types of samples. They concluded that there appear to be situations in which better results are obtained by the sodium lodide system. (1) R. F. Coleman evaluated the precision of sodium fodide and germanium systems,

using both the least squares and the peak area computational methods.⁽²⁾ He found that the highest precision was obtained with the sodium iodide detector and the least squares computational method, especially if combined with group separations. The relative sensitivity and the accuracy of these two detector systems have not been previously reported and are the subjects of this study.

The sodium iodide counting system had a 4" dia by 2" thick Nal(Tl) detector with a resolution of 6% for the 662 keV gamma-ray from 137 Cs. This detector was in an 8" thick iron shield located in an underground counting room where line voltage was regulated to ± 0.2 volts, room temperature at 74 \pm 0.5° F and 222 Rn was less than 0.02 pCi/liter. Spectra were accumulated with a Nuclear Data ND-120 512 channel analyzer and written directly onto computer compatible magnetic tape. Spectra were accumulated until an integral count of 200,000 was obtained. As the samples decreased in counting rate, the counting interval varied from 1.6 to 15 min. All spectra were analyzed by means of a non-linear least squares computer program which corrects för shifts in zero energy intercept and conversion gain. $(3, 4, 5)$

The germanium detector was a 20 cc coaxial type Ge(Li) with a resolution of 2.3 keV at 1.33 MeV, a peak to Compton ratio of > 21, an efficiency of 3*.4%* relative to a $3'' \times 3''$ NaI(Tl) detector, and was located in a $4''$ thick lead brick shield. Spectra were accumulated on a Geoscience 4096 channel analyzer. This system was in an air-conditioned room, but gain stabilization was not used so the width of a peak in channel 4000 was increased by about 50% to 3.7 keV during the 1000 min counting interval used to accumulate all spectra. This variation in resolution did not affect the peak area analysis. The area and height of each peak were determined using the Argonne version (IBM 360 Fortran IV) of the computer program written by Gunnink and Niday. (6) In this program, peak areas are computed by a non-linear least squares method in which a Gaussian function is fit to the peak after subtraction of a baseline estimated from the data. This method has been called "classical" by Yule, and he further states that it always provides the most accurate estimate of peak areas. (7)

Experience has shown that the sensitivity and accuracy of these two detector systems vary with the nuclide being evaluated. For the sodium iodide system, a higher sensitivity is obtained for a nuclide with a single photopeak having an energy between 0.09 and 0.3 MeV than for a nuclide with higher energy or with multiple gamma rays over an energy range of 1 MeV or.more. In contrast, the germanium system was expected to be most sensitive for those nuclides with gamma-ray energies above about 0.5 MeV, or where the Compton contribution from higher energy gamma rays is reduced.

A mixture of seven nuclides was prepared which contained three components with relatively long half lives (140 La, 233 Pa, and 60 Co) at 0.2 to 0.4% of the total disintegration rate and four components with relatively short half lives $(^{24}\mathrm{Na}$, $^{64}\mathrm{Cu}$, $^{72}\mathrm{Ga}$, and $^{82}\mathrm{Br}$). The disintegration rate of each component was accurately known and was adjusted to give a total integrated counting rate of about 100,000 CPM. The three minor components were chosen because they were representative of the three spectral shapes which are commonly encountered, viz., 233Pa, x-rays, and photopeaks at low energy; 60Co, high energy simple photopeak spectrum; and 140 La, a complex, multiple photopeak spectrum over an energy range of more than I MeV; The same samples were counted several times over a period of seven days on each detector. During this time, the activity of the samples decreased by a factor of ten- About 6 months after the first experiment

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was completed, the entire study was repeated and this report includes the data from both.

The spectra shown in Figs. 1 and 2 for the NaI(Tl) and Ge(Li) detectors, respectively, are representative of those obtained. For each, the lower curve is that of the minor components counted alone, and the photopeaks from the 233 Pa, $^{140}\rm{La}$ and $^{60}\rm{Co}$ are all clearly visible. The upper curve in Figs. 1 and 2 is the mixture of all seven nuclides. The great power of the least squares method is illustrated by Fig. 1 , since the minor components were measurable. although none of their photopeaks were visible in the spectrum of the mixture. In contrast, the more abundant photopeaks of the minor components are visible in the upper curve of Fig. 2, although the values for the peak-to-baseline (P/B) ratio range from only 0.005 to 0.05. The photopeaks for $^{140}\rm{La}$ at 1596, 815, and 2521 keV could not be used because of interference from *^^Ga* photopeaks at 1592, 812, and 2508 keV. These interferences are not given in the compilation by Pearson and $Bewers.$ (8)

The accuracy of the determination of 233 Pa , 140 La , and 60 Co when present at $^\circ$ 0.2 to 4% of the total disintegration rate is summarized in Fig. 3. The solid and open symbols are the results from the germanium and sodium iodide systems, respectively. The circle, square, and triangle are the values from 233 Pa, 140 La, and $60Co$, respectively, while the error bars are the standard deviation obtained from the counting data.

 $^{233}\rm{Pa}$ was detectable by both systems when present at 0.2% of the total disintegration rate. . However, the unusually high value from the germanium system (1.59 x true) results from a deliberately low estimate of the baseline by the computer program which was introduced to avoid loss of otherwise significant peaks. For both systems, all nuclides were accurately determined when present as low as 0.5 to 1.0% of the total. The accuracy of the results for all nuclides from the sodium iodide and germanium systems did not differ significantly since the mean ratio (observed/true) was 0.99 ± 0.05 and 1.06 ± 0.06 , respectively. In order to achieve equal accuracy and precision for these two systems, the $\frac{3}{3}$ counting periods for the germanium detector were 60 to 600 times as long as those for the sodium iodide system.

Except for those situations where the amount of activity is not limited by the available neutron flux, the long counting intervals required by the germanium detector are so limiting that the NaI(Tl) detector least squares system is the one of choice for routine activation analysis. The presence of 24 Na limits both systems equally in the analysis of almost all. biological materials , and chemical separations are required. If purely instrumental methods must be used, then only the germanium system is suitable for samples containing more, than 20 nuclides. However, simple photopeak area methods are not suitable for samples of this type, and the least squares technique must be used to eliminate the large number of interferences. $(9,10)$

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Fig. 1.

Fig. 2.

Sodium iodide detector spectra. The upper curve is a spectrum of the mixture when the minor components range from 0.2 to 0.4% of the total disintegration rate. The lower curve is that from the three minor components.

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Solid state detector spectra. The upper curve is a spectrum of the mixture when the minor components range from 0.2 to 0.4% of the total disintegration rate. The lower curve is that from the three minor components. The center curve is the spectra when the minor components are all at about 1% of the total disintegration rate.

Fig. 3. Accuracy of the results for each of the three minor components for each detector. The solid and open symbols are results from the germanium and sodium iodide detectors, respectively. The circle, square, and triangle are 233 Pa, 140 La, and 60 Co respectively.

