BNWL-1285
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SUPPRESSION SPECTROMETERS
FOR NONDESTRUCTIVE
RADIOCHEMICAL ANALYSIS

J. A. Cooper

August 1970

AEC RESEARCH &
DEVELOPMENT REPORT

BNWL-1285

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BNWL-1285
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PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

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ABSTRACT

Ge(Li) Compton suppression spectrometers are evaluated relative to the specific needs of the analytical radiochemist. Their application to the analysis of radionuclides found in neutron activation, fission product, and environmental radionuclide analyses is discussed.

EVALUATION OF Ge(Li) COMPTON SUPPRESSION SPECTROMETERS FOR NONDESTRUCTIVE RADIOCHEMICAL ANALYSIS

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INTRODUCTION

This report evaluates Ge(Li) Compton suppression spectrometers as they apply to the work of the analytical radiochemist. A large number of sophisticated Ge(Li) Compton-suppression spectrometers⁽¹⁻¹⁹⁾ have been developed and evaluated relative to their basic performance parameters. However, they have not been evaluated relative to the specific needs of the analytical radiochemist. All of these spectrometers can provide an improved peak-to-Compton ratio for a pure radionuclide, but this is not always accompanied by an improvement in the sensitivity for the nondestructive measurement of radionuclides in complex mixtures.

SUMMARY

Ge(Li) Compton suppression spectrometers are evaluated relative to the specific needs of the analytical radiochemist. The Compton-summing spectrometers are shown to have a very limited range of applicability. They are not applicable to low-level counting and their application to high-level samples is questionable. The anticoincidence-shielded spectrometers with the source located outside the shield provide uniquely superior performance for decay scheme studies and high level samples which can't be subdivided; however, they are not applicable to low-level samples. The shielded spectrometers with the source positioned inside the shield appear to have the most general application to radioanalytical problems.

DISCUSSION

The sensitivity of a gamma-ray spectrometer is a measure of its ability to detect gamma rays in the presence of interference from natural radioactivity and the Compton continuum

from higher energy gamma rays. The sensitivity is of the greatest concern when this interference is greater than, or of the same order of magnitude as the peak. This general problem is illustrated in Figure 1, which shows a peak superimposed on a flat background before and after background suppression. The accuracy, or sensitivity, will be improved if the fractional error before background suppression (σ/N) is greater than the fractional error after background suppression (σ'/N'). That is, the background suppression device will improve the sensitivity if the ratio of the fractional error before to the fractional error after background suppression is greater than 1. However, since the full-energy peak detection efficiency is usually reduced along with the background interference, the magnitude of the improvement will depend not only on the background suppression, but also on the peak efficiency reduction factor (e), the initial peak-to-background ratio (R), and the improvement in the peak-to-background ratio (r).

Galloway⁽²⁰⁾ has derived the following equation relating these basic parameters to the ratio of the accuracy (A) before background suppression to the accuracy (A') after background suppression (A/A'):

$$A/A' = \left\{ [er(R + 2)] / (rR + 2) \right\}^{1/2}$$

The dependence of this ratio on e , r , and R is illustrated in Figure 2. The dashed line at A/A' equal to 1 divides the improved and reduced sensitivity regions. There will be an improvement in the sensitivity after background reduction for those parameter values for which A/A' is above the broken line. This figure shows that for large initial peak-to-background ratios (R), the ratio of A/A' approaches 1 for e values of 1. Thus, the improvement in sensitivity is rather insignificant for large initial peak-to-background ratios. On the other hand, the most dramatic improvements in sensitivity are obtained when R is small: i.e., when the peak of interest is difficult or impossible to detect because of Compton or background interference.

FIGURE 1. A Peak on a Smooth Background Before and After Background Suppression (20)

FIGURE 2. A Plot of the Ratio of the Fractional Error Before to the Fractional Error After Background Suppression Versus the Initial Peak-to-Background Ratio (20)

The importance of maintaining a high peak efficiency while reducing the background is also illustrated in Figure 2. For example, if $e = 0.1$ (i.e., if the peak efficiency is reduced by a factor of 10, as is often the case with the Ge(Li) Compton-summing systems), the background must be reduced by more than a factor of 100 before any improvement in sensitivity can be achieved ($r = 10$, $e = 0.1$). On the other hand, if the peak efficiency is not reduced at all (i.e., $e = 1$), then any reduction in the background will improve the sensitivity. However, the magnitude of the improvement will depend not only on the amount of background reduction, but also on the initial peak-to-background ratio. Thus, one of the most important factors to consider when evaluating Compton suppression techniques is the reduction in peak efficiency accompanying the Compton suppression.

Ge(Li) Compton-suppression spectrometers can be divided into two basic categories: (1) the all Ge(Li) Compton-summing systems,⁽¹⁰⁻¹⁶⁾ and (2) the anticoincidence-shielded Ge(Li) systems^(1-9,17) which can be subdivided into those systems with the source located inside the shield (ACS-in) and those with the source outside the shield (ACS-out). The all Ge(Li) Compton-summing spectrometers utilize two Ge(Li) detectors which are operated in a sum-coincidence mode, while the anticoincidence-shielded spectrometers reject diode pulses resulting from gamma rays which have deposited a fraction of their energy in both the diode and scintillator.

The reciprocal of the peak reduction factors ($1/e$) for some of those radionuclides of interest to the analytical radiochemist have been calculated for the ACS-in system and are listed in Tables 1, 2 and 3. The reduction factors were determined by estimating the difference between the normal and Compton suppressed count rates for the most intense peak in the normal spectrum. This was determined from the radionuclide decay scheme and the anticoincidence shield efficiency.⁽¹⁷⁾

TABLE 1. Inverse Reduction Factors for Radionuclides
Produced by Neutron Activation

<u>Radionuclides</u>	<u>E_y, keV</u>	<u>Inverse Reduction Factor, 1/e</u>
Na-24	1369	4
Mg-27	842	1.0
Al-28	1780	1.0
Si-31	1260	1.0
S-37	3090	1.0
Cl-38	1645	5
K-42	1524	1.0
Sc-46	889	7
Ca-47	1300	1.0
Sc-47	160	1.0
Ca-49	3100	1.0
Cr-51	320	1.0
Ti-51	320	1.0
V-52	1434	1.0
Mn-54	835	1.0
Mn-56	847	2
Fe-59	1095	1.0
Co-60	1173	7
Cu-64	511	20
Ni-65	1481	1.0
Zn-65	1115	1.0
Zn-69m	439	1.0
Ga-70	1040	1.5
Ga-72	835	20
Ge-75	265	1.0
Se-75	401	20
As-76	559	1.2
Br-82	554	65
Sr-85	514	1.0
Rb-86	1078	1.0

TABLE 1. (Contd)

<u>Radionuclides</u>	<u>E_y, keV</u>	<u>Inverse Reduction Factor, 1/e</u>
Sr-87m	388	1.0
Y-90	1750	1.0
Zr-95	724	1.0
Zr-97	747	1.0
Mo-99	740	1.0
Tc-99m	143	1.0
Ru-103	497	1.0
Ru-105	726	1.1
Rh-104m	555	1.1
Ag-108	633	1.0
Pd-109	88	1.0
Pd-109m	188	1.0
Ag-110m	658	50
Sn-113	393	1.0
In-114m	192	1.0
Cd-115	530	1.0
In-115m	335	1.0
Te-121	573	1.0
Te-121m	212	1.0
Sb-122	564	1.1
Sb-124	603	4
I-128	441	1.1
Ba-131	216	1.4
Cs-134	605	10
La-140	1596	10
Ce-141	145	1.0
Pr-142	1570	1.0
Nd-147	531	1.0
Eu-152	344	8
Eu-152m	963	5
Gd-153	99	1.0

TABLE 1. (Contd)

<u>Radionuclides</u>	<u>E_y, keV</u>	<u>Inverse Reduction Factor, 1/e</u>
Sm-153	103	1.5
Gd-159	363	1.0
Tb-160	966	8
Dy-165	95	1.0
Yb-169	198	20
Tm-170	84	1.0
Er-171	308	20
Yb-175	396	1.0
Hf-181	482	8
Ta-182	100	40
W-187	685	1.3
Re-188	155	1.1
Os-191	129	1.2
Ir-192	317	20
Hg-197	191	1.3
Pt-197	191	1.3
Au-198	412	1.0
Hg-203	280	1.0
Pa-233	312	1.3
Np-239	278	3
U-239	75	1.0

TABLE 2. Inverse Reduction Factors for Fission Products

<u>Radionuclides</u>	<u>Eγ, keV</u>	<u>Inverse Reduction Factor, l/e</u>
Zn-72	145	1.0
Ga-72	835	20
Ga-73	295	1.1
Ga-74	596	5
Ge-75	265	1.0
As-76	559	1.2
Ge-77	210	20
As-77	240	1.0
Ge-78	277	1.0
As-78	614	1.8
As-79	96	1.0
Br-80	618	1.0
Se-81	280	5
Br-82	554	65
Se-83	360	20
Br-83	530	1.0
Br-84	880	5
Kr-85	514	1.0
Rb-86	1078	1.0
Kr-87	403	1.4
K-88	191	1.4
Rb-88	898	7
Sr-91	728	1.4
Y-91	1210	1.0
Y-91m	551	1.0
Sr-92	1370	1.0
Y-92	934	4
Y-93	267	1.4
Zr-95	724	1.0
Nb-95	765	1.0

TABLE 2. (Contd)

<u>Radionuclides</u>	<u>E_γ, keV</u>	<u>Inverse Reduction Factor, 1/e</u>
Nb-96	778	20
Zr-97	747	1.0
Nb-97	665	1.0
Nb-98	787	20
Mo-99	740	1.0
Tc-99m	143	1.0
Ru-103	497	1.0
Ru-105	726	1.1
Rh-105	319	1.0
Rh-106	512	2.0
Rh-107	305	1.0
Ag-111	342	1.0
Pd-112	617	2.0
Cd-115	530	1.0
Cd-115m	935	1.2
Sn-123m	160	1.0
Sn-125	1068	10
Sn-125m	325	1.0
Sb-125	427	1.0
I-128	441	1.1
Te-129m	455	1.0
I-130	538	20
Te-131	150	1.1
Te-131m	780	20
I-131	364	1.0
I-132	669	15
Te-132	230	1.0
Te-133m	432	20
Xe-133	81	1.0
Xe-133m	233	1.0

TABLE 2. (Contd)

<u>Radionuclides</u>	<u>E_y, keV</u>	<u>Inverse Reduction Factor, 1/e</u>
I-134	850	20
I-135	1280	1.5
Xe-135	250	1.0
Cs-136	818	15
Cs-137	662	1.0
Cs-138	1426	3
Ba-139	166	1.0
Ba-140	537	1.0
La-140	1596	10
La-141	1370	1.0
Ce-141	145	1.0
La-142	650	15
Ce-143	293	2.0
Ce-144	135	1.0
Nd-147	531	1.0
Nd-149	210	1.0
Pm-149	286	1.0
Sm-153	103	1.5
Sm-155	104	1.1
Eu-155	105	1.0
Eu-156	646	15
Eu-157	413	2
Gd-159	363	1.0

TABLE 3. Inverse Reduction Factors for Environmental Radionuclides

Radionuclide	E, keV	Inverse Reduction Factor, 1/e	Radionuclide	E, keV	Inverse Reduction Factor, 1/e
Be-7	477	1.0	Rh-106	512	2
Na-22	511	20	Ag-110m	658	50
K-40	1460	1.0	Sb-124	603	4
Sc-46	889	7	Sb-125	427	1.0
Cr-51	320	1.0	I-131	364	1.0
Mn-54	835	1.0	Cs-134	605	10
Co-57	112	1.0	Cs-137	662	1.0
Co-58	810	1.2	Ba-140	537	1.0
Fe-59	1095	1.0	La-140	1596	10
Co-60	1173	7	Ce-141	145	1.0
Zn-65	1015	1.0	Pr-144	695	1.2
Y-88	898	6	Ce-144	135	1.0
Zr-95	724	1.0	Eu-152	344	8
Nb-95	765	1.0	Eu-155	105	1.0
Ru-103	497	1.0	Bi-207	570	8

For example, ^{46}Sc has an 889 keV gamma ray in coincidence with an 1120 keV gamma ray. The gamma ray most likely measured under normal conditions would be the 889 keV gamma ray because of the detectors greater efficiency for lower energy gamma rays. However, when this radionuclide is measured in an ACS-in system, the intensity of the 889 keV peak will be reduced in the anti-coincidence spectrum by an amount determined by the efficiency of the anticoincidence shield for detecting the 1120 keV gamma ray. The efficiency of one anticoincidence shield for detecting 1120 keV gamma rays is 86%.⁽¹⁷⁾ Thus 86% of the 889 keV gamma rays would be stored in the coincidence spectrum and 14% in the anticoincidence spectrum yielding an inverse reduction factor for the measurement of ^{46}Sc in this particular ACS-in spectrometer of 7. Radionuclides with more than 2 gamma rays in cascade

will have still greater reduction factors because each gamma ray has the possibility of being detected in coincidence with more than one additional gamma ray.

In Figure 3, the number of radionuclides with a specific reduction factor are plotted versus the reciprocal of the peak reduction factor ($1/e$). [The reduction factors for all Ge(Li) Compton-summing system were calculated from the efficiency curves of Kraner and Chase.⁽¹⁴⁾] One survey included 85 radionuclides resulting from neutron activation with half lives greater than 2.5 min and the other included 40 of the most commonly found radionuclides in neutron activated material with half lives greater than 60 min. A reduction factor of 1 means the full-energy peak intensity is not reduced by the Compton reduction process and the sensitivity for the measurement of this radioisotope will definitely be improved. Those radionuclides with inverse reduction factors greater than 10 cannot have improved detection sensitivities with either an anticoincidence-shielded system or the Compton-summing spectrometers since neither can provide background reductions greater than 100 in the region where they might obtain a peak reduction of 10. The sensitivity improvement for radionuclides with inverse reduction factors between 1 and 10 is uncertain and will depend on how much the background is reduced and the original peak-to-background ratio.

The results of this survey of 85 radionuclides show that two-thirds of these radionuclides will definitely have improved sensitivities with the ACS-in system. On the other hand, the Compton-summing spectrometers cannot improve the sensitivity for over half of these radionuclides, and those with inverse reduction factors between 1 and 10 are in the uncertain category. This same trend is also evident for the more selective surveys shown in Figure 4, which include those radionuclides

FIGURE 3. Frequency Distribution of the Inverse Full-Energy Peak Reduction Factors for Radionuclides Produced by Neutron Activation

FIGURE 4. Frequency Distribution of the Inverse Full-Energy Peak Reduction Factors for Radionuclides Measured in Neutron Activated Geological and Biological Material

commonly measured in neutron activated geological and biological material. The fission products listed in Table 2 show a similar trend, with two-thirds of these radionuclides having inverse reduction factors of 1 and therefore improved detection sensitivities with the ACS-in system.

The detection of radioactivity in low-level environmental samples requires not only a reduced Compton continuum but also a maximum absolute peak efficiency and a minimum interference from natural background radiation. The only Ge(Li) detector system which fulfills all these requirements for most radionuclides in environmental and biological samples is the ACS-in spectrometer. A frequency distribution for the 30 most commonly measured radionuclides in environmental samples is presented in Figure 5. Again, almost two-thirds of the radionuclides, when measured on the ACS-in system, will have essentially no reduction in the full-energy peak efficiency. Thus, any reduction in the Compton continuum and natural background will improve the detection sensitivity for these radionuclides. If the coincidence spectrum is also stored,⁽¹⁷⁾ there will be no loss in the sensitivity for the measurement of the other one-third. However, it becomes questionable whether this sensitivity can be improved for radionuclides with inverse reduction factors greater than 2 in low-level samples.

The major disadvantage of the Compton-summing spectrometers is the large reduction in the full-energy peak intensity which accompanies the Compton reduction. In addition, the absolute efficiency and resolution of these systems have not been equal to the state-of-the-art normal spectrometers which might be placed inside a Compton suppression shield. It is clear that they are not applicable to low-level counting and their application to high-level samples is questionable. They may be of practical value for the measurement of a few radionuclides in

FIGURE 5. Frequency Distribution of the Inverse Full-Energy Peak Reduction Factors for Radionuclides Measured in Environmental Samples

a few specific examples, but are generally of little use to the analytical radiochemist in their present state of development and hold little promise for the future.

Those systems in which the source must be highly collimated and placed outside the shield are seriously limited by the 100-fold or more reduction in their absolute detection efficiency. In addition, the maximum Compton reduction is presently only about 10. Although these systems are not of value for the measurement of radionuclides in low-level environmental samples, they are the best detection systems for the ultra-high intensity samples such as irradiated fuel elements and studies of decay schemes. The sensitivity for these problems in which the count rate is limited by the spectrometer's electronics depends on

the intrinsic efficiency of the primary detector and not its absolute efficiency. Thus, the over 100-fold reduction in the geometrical efficiency for the ACS-out system is of little consequence for these samples. However, this is not the case for the all Ge(Li) Compton-summing spectrometers and the ACS-in spectrometers because the rate of data accumulation is further restricted by the secondary detector. Although the count rate capabilities of the first diode in a Hick and Pepelnick(15) type Compton-summing spectrometer may be the same as a normal diode, the ultimate rate of data accumulation is further limited by the efficiency of the second diode to detect the gamma rays scattered from the first diode. The count rate of the ACS-in system is limited by the count rate capabilities of its shield and coincidence circuitry.

The ACS-in spectrometers are restricted to source intensities of less than about 1 μCi . However, more intense sources can often be subdivided to provide a source of the proper intensity. In addition, more intense sources could be counted if the shield electronics and coincidence circuitry were improved.

An additional feature of the ACS-in system is its high inverse Compton reduction factors for radionuclides which decay by emission of coincident gamma rays. For these radionuclides, there exists a very high probability that the diode response to a gamma ray (either full energy or Compton) will be cancelled by the subsequent detection of the coincident gamma rays by the anticoincidence shield. This effect is more often than not an asset rather than a liability. Because of this feature, the ACS-in system is in some cases capable of providing an 80-fold reduction in the Compton interference without affecting the peak intensity of noncoincident gamma rays. (17)

ACKNOWLEDGEMENTS

I wish to thank R. W. Perkins, L. A. Rancitelli, and N. A. Wogman for their many helpful comments.

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