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RADIATION DETECTORS: NEEDS AND PROSPECTS

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ABSTRACT

Important applications for α - and γ -ray spectroscopy are found in prospecting, materials characterization, environmental monitoring, the life sciences, and nuclear physics. The specific requirements vary for each application with varying degrees of emphasis on either spectrometer resolution, detection efficiency, or both. Since no one spectrometer is ideally suited to this wide range of needs, compromises are usually required. Gas and scintillation spectrometers have reached a level of maturity, and recent interest has concentrated on semiconductor spectrometers. Germanium detectors are showing continuing refinement and are the spectrometers of choice for high resolution applications. The new high-Z semiconductors, such as CdTe and HgI₂, have shown steady improvement but are limited in both resolution and size and will likely be used only in applications which require their unique properties.

INTRODUCTION

The field of α - and γ -ray spectroscopy has continued to mature with continuing technical refinements and an increasingly broad range of applications. The primary driving force for early development in this area was generated by the critical spectroscopy needs of nuclear physics required for significant advances in this area of the physical sciences. With the maturation of α - and γ -ray spectroscopy, new applications have been found or refined in other areas of human activity. It is the intent of this paper to explore some of these developing needs, to consider the special demands which many of these applications make on spectroscopy systems, and to consider the capabilities, limitations, and prospects for meeting these needs with current and developmental spectrometer systems.

α - and γ -Ray Spectrometer Applications

In considering present and potential applications, it would be fruitful to first consider these need areas which have a major impact on human activity prior to considering some of the spectrometer applications in those areas.

Basic human needs include food, clothing, shelter, and medical care. In supplying these basic needs, additional factors such as protection of the environment must be considered. In considering the potential applications of detectors, it is worthwhile to consider such applications in the context of 1) the pursuit and development of these natural resources needed to support life, 2) the protection of the environment, and 3) the life sciences.

Natural resources, in most cases, are contained in the crusts of the earth. Critical resources include the fossil and nuclear energy sources and the large variety of minerals which are needed. Major needs exist in locating, identifying, and evaluating the quality of many of these distributed resources. It is now possible to perform this identification using many of the newly developed techniques of excitation and analysis of α -ray and γ -ray spectra. Many of these techniques, once the province of the laboratory, have been refined and hardened for field use applications such as prospecting and well logging. Due to the obvious needs which mankind has in this area, it is safe to assume that we have only seen the beginning of such applications.

One area of natural resource needs which is particularly critical involves the food supply. While significant penetration of spectroscopy has not yet occurred in this area, food chain applications involving radionuclide studies have been long known and continue to improve. More recent studies involving the use of spectroscopic techniques for elemental characterization into the part per billion range promise new studies in plant growth understanding and disease control.

The pursuit of energy includes the current use of nuclear fission and the potential future use of nuclear fusion. The control and monitoring of the nuclear fuel cycle places significant demand on spectroscopy. Obvious needs exist in the enrichment of nuclear materials, the fabrication of nuclear fuels, and fission reactor control and monitoring. Further application needs also exist for the bottom end of the fuel cycle in fuel reprocessing and nuclear waste disposal.

A particular need in the nuclear fuel cycle exists in the protection and control of the fissionable isotopes which are suitable a byproduct of fission power and which are for the fabrication of nuclear weapons. The misuse of such materials must be avoided. Current safeguards techniques used to prevent nuclear material diversion include the obvious physical protection measures, the measurement and accounting procedures for the control of special nuclear materials, and the detection of any intentional material diversion activities. The nondestructive techniques necessary for the accounting step are very demanding in terms of the needed accuracy and rely heavily on various α -ray and γ -ray spectroscopy techniques. Diversion detection presents its own set of problems in terms of the needed rapid detection of relatively small quantities of materials which are not really very radioactive.

Protection of the environment is important both in ensuring esthetic continuity and in pre

venting the buildup of hazardous conditions. The primary environmental concern is the addition of hazardous substances as a result of human activity. Such activities include energy production and use, manufacturing and farming, and human waste disposal.

Many of the emissions from these activities involve either radioactive materials or trace elements which are toxic even in small quantities. X- and γ -ray spectroscopy provide attractive alternatives for the monitoring and evaluation of these substances. γ -ray techniques for elemental analysis involve γ -ray spectroscopic measurement of the neutron-induced prompt γ -rays and delayed radioisotopic emissions. X-ray elemental analysis, using x-ray, electron beam, and charged particle excitation, also provides a powerful tool for such measurements.

The life sciences have numerous opportunities for potential applications. At present, nuclear techniques provide a non-invasive means of gaining information on internal body conditions and functions. The preferential uptake of labelled pharmaceuticals by abnormal tissue as well as certain normal body organs has made possible a variety of imaging techniques. The most popular such application is the anger γ -ray camera in popular use throughout the world. An alternative imaging approach under development is the positron camera which depends on the simultaneous detection of the annihilation quanta from a positron emitting radio-pharmaceutical.

An alternate method for obtaining internal organ information is provided by localized body density measurements which depend on either scattering or absorption. The current sophistication of this technique is represented by the Computer Assisted Tomograph (CAT) scanners currently in use.

Other life science applications are also found in blood flow and organ function studies. These require the use of special probes (some intravenous) or large computer controlled detector arrays.

The physical sciences have continuing requirements. The needs of nuclear and high energy physics are well known to the spectroscopist. Of growing interest in materials science are those applications which permit elemental identification, particularly on a microscale, or those which permit the unravelling of atomic and molecular structure. Significant areas of advancement in metallurgy and materials science depend on such spectroscopy applications. Adaptations of this basic materials analysis tool are currently finding applications in manufacturing and quality control.

Spectrometer Requirements

Many of the spectroscopy applications considered earlier have been summarized in short form in Table I. Each of these applications places particular emphasis on certain detector parameters such as detector resolution, detector efficiency (which includes solid angle as well as intrinsic peak efficiency), and stability. These parameters are also shown in Table I and are considered in a matrix format with the listed applications. A qualitative estimate was made of the relative importance of specific detector parameters for each application. For each parameter judged important, a "Y" was entered in the matrix on Table I. Where a parameter was judged to be of either secondary or no importance, a "-" was entered.

In most applications, either detection efficiency, spectrometer resolution, or both were considered to be important. Few applications are insensitive to these parameters. In general (and the reader is invited to consider this on his own), an application which requires either resolution or efficiency leads to a lack of willingness to compromise these requirements to a significant extent unless other factors are overriding. Thus, high resolution spectroscopic applications usually will employ germanium or silicon detectors despite the inconvenience of cooling. Conversely, applications which require efficiency and can compromise on resolution will use a scintillation detector. Special applications may require some other particular characteristics which may obviate the use of a high-Z semiconductor detector or one of the gas counters. The choice of spectrometer type is determined is reached by considering the reali-

ties and driving forces of real-world problems rather than the developmental dreams of the spectroscopy researcher.

Given the importance of the parameters of both resolution and efficiency in certain applications, it is obvious that no one spectrometer type satisfies all of the needs simultaneously. Improvements are always needed in both resolution and efficiency, especially if these could be combined in the same detector. The ability of different known detector types to meet these expectations, both for the present and in the foreseeable future, will be considered in the next section.

Spectrometers: Status and Future

The most common spectrometers include: 1) the various gas and liquid ionization and proportional counters, 2) the scintillators, and 3) the semiconductor detectors. Each of these classes of detectors has peculiar operating characteristics and will be considered in terms of their ability to provide the parameters of efficiency, resolution, or other particular detector property.

Ionization and Proportional Counters

Counters in this category depend on transient charge collection in a liquid or gas media due to ionizing events. Such counters are straightforward and are generally reliable, but are limited in both efficiency and energy resolution. The efficiency limitation has been largely due to the low density of the gas detection phase used in such spectrometers. The resolution of these spectrometer types has been limited by the large average energy per ionization, problems in the uniformity in the charge collection and/or multiplication process, and impurity effects.

Significant advances have been made in achieving higher efficiency with recent improvements with liquid ionization detectors based on liquid argon or xenon. New purification processes have been largely responsible for this advance and an energy resolution of 34 keV FWHM has been achieved for the $\sim$ MeV beta particles from ^{208}Tl [1]. The ultimate resolution for such a detector is projected to be somewhat better than this, but cannot equal a good germanium spectrometer due to the problems of higher average energy per ion-pair and the collection of only one type of charge specie. This latter effect manifests itself as a geometrical dependence of pulse magnitude on location of ionization in the detection medium and would be especially troublesome for γ -ray spectroscopy applications.

Interesting results have also been obtained with the gas scintillation proportional counters [2-4]. In this counter, detection occurs in a low-field region of the gas. The collected charge is then accelerated through a high field region resulting in light scintillation. This light is then detected and amplified by a photo-multiplier tube. Such detectors have shown excellent energy resolution for low energy x-rays - $\sim$ 600 eV FWHM at 5.9 KeV and 85 eV FWHM at 149 eV [3]. More recent advances in this type of detector have resulted in efficient operation to over 100 KeV in energy with resolution for the 122 KeV line of ^{57}Co of $\sim$ 5 KeV [4]. Such performance must be taken seriously, particularly where large area or position sensitive applications are considered.

Scintillation Detectors

Scintillation detectors have been popular due to their reliability, reasonable cost, and attainable efficiency. In general, their primary drawback has been in terms of their resolution capability for spectroscopy applications.

Efficiency has not been a particular problem for scintillators. Improvements in efficiency have been sought by the use of alternate scintillator materials such as cesium iodide or calcium tungstate. A scintillator material which is receiving recent attention in this regard is bismuth germanate [5]. Perhaps the most important factor in determining energy efficiency, however, is in the attainable dimensions of the scintillation media. Single crystal sodium iodide detectors approaching one cubic meter have been made [6]. In addition, the use of polycrystalline scintillator

materials and crystalline pieces in a light coupling medium have reduced thermal sensitivity and increased the flexibility of geometrical shapes and sizes. Without question, the scintillators represent the most efficient x-ray spectrometers available now or for the foreseeable future.

The resolution capability of scintillators is largely limited by the multiplicity of the physical processes involved in generating an electrical signal. Loss mechanisms result at each stage of ionization, light emission, light collection, and photocathode emission with the result that an average energy of 300 eV is required for each photoelectron. Resolution loss occurs as a result of the statistical spread occurring in the production of a relatively small number of electrons and by the spread in each of the loss mechanisms due to non-uniformities. Additional spread in the signal amplitude can also be caused by non-uniformities in the electron multiplication process.

Each of these resolution factors continues to receive attention and continued refinement. However, no significant breakthroughs - accompanied by a step-function improvement in resolution - are anticipated in the foreseeable future. Of the anticipated refinements, improvements in scintillator and photocathode efficiency and uniformity are the most likely.

Semiconductor Detectors

Two of the semiconductor detectors, silicon and germanium, form the backbone of interest in semiconductor detectors for providing the highest resolution spectrometers. The primary advantages of these semiconductor detectors are their low average energy per ion-pair, small Fano factor, nearly perfect charge collection, and negligible noise contribution when cooled to liquid nitrogen temperatures. These properties of germanium and silicon make them unchallenged, either now or in the foreseeable future, by any other semiconductor for providing high resolution x-ray and y-ray spectroscopy.

Both germanium and silicon are relatively low atomic number elements and suffer a corresponding loss in photoelectric interaction efficiency. This factor, coupled with limitations on active volume caused by the problems in properly depleting and collecting charge carriers from larger P-N junction depletion regions, have limited the efficiency of individual detectors when compared with the scintillators. The result has been a search for new higher atomic number semiconductors for spectrometer applications. The results of one such study, which considered all possible binary compounds of the higher atomic number elements, was a rank-ordered listing of potential detector materials as shown in Table II [7]. The rank ordering was based on those compounds which had an energy bandgap between 1.5 and 2.2 eV (which permits room temperature operation) and was based on the charge collection capability of these semiconductors as represented by their minimum charge carrier mobility. To date, only two of these compounds, CdTe and HgI₂, have received significant development attention and study as spectrometers.

Prior to considering what has or can be accomplished with these new spectrometer materials, it is appropriate to consider those factors which impact semiconductor spectrometer performance. In the detection of x-rays and y-rays, ionization is produced by the photoelectric, Compton, and pair production interactions. For large detector volumes, incomplete energy transfer events (Compton or pair production) or energy loss events (Bremsstrahlung or energetic electron loss) are minimized by multiple capture events. However, as the physical detector dimensions are reduced, decreased possibility for full-energy event generation by multiple interactions results. In addition, as the detector dimensions approach the range of high energy photoelectrons (~1 mm), significant loss of photoelectric events occurs due to loss of ionizing electrons from the active volume. Thus, high atomic number is a sufficient criteria for producing efficiency in a detector since detector volume is perhaps of greater importance due to both the probability of multiple event capture and the increased solid angle for detection.

Once ionization is created in the detector,

it must be collected with an efficiency approaching 100% for reasonable resolution. The efficiency of this collection is affected by both the mobility of the charge carriers, μ , and their lifetime in the excited state, τ . Carrier mobility is a fixed physical property for a given semiconductor material and the best available values are also shown in Table II for the rank-ordered listing of potential high-Z semiconductors. Carrier lifetime, on the other hand, is largely a function of the crystal and detector technology and is usually limited by the presence of impurities and crystal defects.

The product of $\mu\tau$ is usually used as a quality factor indicator for charge collection in a semiconductor detector. The requirements on $\mu\tau$ are usually set by the minimum detection efficiency and resolution requirements levied on a given detector for practical applications. The high-Z detector material study referred to earlier assumed minimal performance standards of 2% photopeak efficiency at 662 keV with 2% FWHM resolution at this same energy [7]. Based on these minimum performance standards, the minimum acceptable values of $\mu\tau$ for different detector materials were determined as shown in Table III. $\mu\tau$ values smaller than those indicated would result in further detector performance degradation in terms of either attainable efficiency, resolution, or both.

Table IV indicates the best reported values of $\mu\tau$ for HgI₂ and CdTe with representative values for Ge and Si [7-10]. All other semiconductor detector materials have $\mu\tau$ products less than HgI₂ or CdTe. There has been continuing improvement in $\mu\tau$ for both materials over the years. Values of electron and hole $\mu\tau$ for CdTe are now marginally acceptable while hole $\mu\tau$ for HgI₂ is still about an order of magnitude too low. Whether further significant improvement in $\mu\tau$ can be achieved is unknown at present, but any optimism for achieving thicker HgI₂ detectors based on improving $\mu\tau$ product should be tempered by the limitations of charge collection time dictated by the small hole mobility.

R. Whited and M. Schieber recently published an excellent review article complete with "best" CdTe and HgI₂ spectral performance for ⁶⁰Co, ¹³⁷Cs, ⁵⁷Co, and ²⁴¹Am [10]. The best reported resolution performance for these detector materials is summarized in Table V. In general, significantly better resolution can be obtained with HgI₂ at the higher energies, but at a greatly reduced efficiency. Clearly, Ge has the highest resolution capability.

Lower energy behavior of the high-Z semiconductor detectors is considerably improved largely due to the enhanced energy absorption on one surface of the detector. This asymmetrical charge generation requires charge collection by only one type of carrier resulting in an improvement in charge collection efficiency of several orders of magnitude for HgI₂. The improvement noted for CdTe at lower energies is also enhanced by the ability to reduce the required detector volume which results in lower detector noise contribution.

Clearly, the performance of HgI₂ is of greatest interest in the low energy regime. As indicated in Table V, resolutions for x-ray energies, while not equal to silicon, are potentially adequate for those applications where liquid nitrogen cooling requirements may cause significant difficulties [11]. The primary advantage that the high-Z semiconductors may have - that of a high specific sensitivity per unit volume - may also be of overriding importance in applications such as insertable medical diagnostic probes [12]. In general, the tradeoffs involved in using any of the high-Z semiconductors in place of either a scintillation, germanium, or silicon spectrometer must be weighed carefully in terms of the gains in operating convenience at the loss of either efficiency or resolution or both.

While significant attention has been given to the limitations and prospects of the high-Z detectors in this section, it would be worthwhile to also consider the advances being made in germanium detector technology. The improvements in detector stability with the advent of high purity germanium are well known. However, the surface sensitivity of germanium detectors have continued to present problems in assembling detector matrices and in achieving a truly rugged detector. To compensate for this, an

encapsulation technique adapted to high purity germanium has been developed at the Lawrence Livermore National Laboratory which has demonstrated detector stability over a four year period. Using these detectors, it has been possible to assemble a working 200 element detector array for blood-flow studies in the brain.

An alternate successful approach for detector stabilization has also been reported by Hansen and coworkers [14]. In this approach, the sensitive surface of the germanium is coated with hydrogenated amorphous germanium. The results to date are quite promising.

Continued improvements in the assembly of larger germanium matrices have been reported [15]. These matrices are able to retain the excellent resolution of germanium spectrometers while considerably improving detector efficiency for specific applications. Further engineering developments have also been considered for providing alternate cooling for the detector independent of the use of liquid nitrogen. It is this general area of device improvement and system engineering that will provide significantly enhanced spectrometer performance and convenience for a wider future range of applications of germanium and silicon spectrometers.

CONCLUSIONS

Numerous applications are evolving in energy, environmental, and life science needs which utilize the x and γ -ray spectrometers developed for nuclear and atomic physics. In general, these spectrometer applications fall into the categories of environmental monitoring, materials characterization, and spatial radioisotope imaging. Most applications stress either efficiency, resolution, or both, with only a handful of applications at present which stress some other detector property such as operating temperature or high specific sensitivity per unit volume. The scintillators will continue to satisfy those applications where efficiency is paramount or where cost is a factor. Silicon detectors will continue to dominate the dispersive x-ray analysis field, though some of the gas scintillation proportional counters may be of interest in this area. Germanium detectors will continue to be the high resolution spectrometers of choice for the foreseeable future. Continuing system engineering improvements for the germanium spectrometers will improve their versatility, reliability, and reduce their dependence on liquid nitrogen supply. In addition, considerable improvement in efficiency will be possible

with bigger and better designed arrays. High-Z semiconductor detectors, which have limitations in both resolution and efficiency, will likely find use in those applications which require the unique properties of these detector materials.

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TABLE I: Relative Importance of Different Parameters
for Representative Application Areas.
The "Y" Entry Indicates Significant Importance.

APPLICATIONS

<u>Detector Parameters</u>	Prospecting and Mtls. Charac.		Monitoring and Environment			Life Sciences		Physics	
	n - γ	X-Ray	Reactor Control	Emissions And Waste	Nuclear Safeguards	Imaging	Probes	Nuclear	High Energy
Resolution	-	Y	-	Y	Y	-	-	Y	-
Efficiency	Y	-	-	Y	Y	Y	Y	-	Y
Stability	-	-	Y	Y	Y	-	-	Y	-
Special Properties	-	-	Y	-	-	-	Y	-	-

TABLE II

RANK ORDERED LIST OF POTENTIAL
DETECTOR MATERIALS

<u>SEMICONDUCTOR</u>	<u>Z_{max}</u>	<u>E_g</u>	<u>μ_e</u>	<u>μ_h</u>
1. AlSb	51	1.62	*1200	*700
2. InP	49	1.35	4700	150
3. ZnTe	52	2.25	340	110
4. CdTe	52	1.50	1050	80
5. WSe ₂	74	1.36	100	80
6. CdSe	48	1.74	50	50
7. BiI ₃	83	1.7	*680	*20
8. Cs ₃ Sb	55	1.6	500	10
9. HgI ₂	80	2.2	94	4

*THEORETICAL

TABLE III

MINIMUM DETECTOR THICKNESS AND $\mu\tau$
PRODUCT FOR 2% IPE AND 2% FWHM @ 662 keV

<u>Semi. Mass Range</u>	<u>Minimum Thickness</u>	<u>$\mu\tau_{\min}$</u>
-30 (Ge)	4.2 mm	$1.5 \times 10^{-3} \text{ cm}^2/\text{Volt}$
-50 (CdTe)	2.3	4.4×10^{-4}
-80 (HgI ₂)	1.1	1×10^{-4}

TABLE IV

"BEST" REPORTED $\mu\tau$ PRODUCT FOR THE
SEMICONDUCTOR DETECTOR MATERIALS OF INTEREST

<u>SEMICONDUCTOR</u>	<u>$\mu_e \tau_e$</u>	<u>$\mu_h \tau_h$</u>
Ge, Si	$>1 \text{ CM}^2/\text{VOLT}$	$>1 \text{ CM}^2/\text{VOLT}$
CdTe	2.6×10^{-3}	4×10^{-4}
HgI ₂	5×10^{-4}	3×10^{-5}

TABLE V

"Best" Reported Spectrometer Performance for
Different Radioisotopes for the Semiconductor
Spectrometers of Interest

<u>SEMICONDUCTOR</u>	<u>^{55}Fe</u>	<u>^{241}Am</u>	<u>^{57}Co</u>	<u>^{137}Cs</u>	<u>^{60}Co</u>
Ge	-	<500 eV	<1 KeV	<1.5 KeV	<1.8 KeV
Si	<150 eV	<500 eV	-	-	-
HgI ₂	295 eV	1200 eV	6 KeV	4.5 KeV	21 KeV
CdTe	1100 eV	1700 eV	4.6 KeV	8.3 KeV	14 KeV