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SCINTILLATION COUNTING: AN EXTRAPOLATION INTO THE FUTURE*

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Abstract

Progress in scintillation counting is intimately related to advances in a variety of other disciplines such as photochemistry, photophysics, and instrumentation. And while there is steady progress in our understanding of luminescent phenomena, we are in the middle of a virtual explosion in the application of semiconductor technology to detectors, counting systems, and data processing. The exponential growth of this technology has had, and will continue to have, a profound effect on the art of scintillation spectroscopy. This paper will review key events in technology that have had an impact on the development of scintillation science (solid and liquid) and will attempt to extrapolate future directions based on existing and projected capability in associated fields. Along the way there have been occasional pitfalls and several false starts; these too will be discussed as a reminder that if you want the future to be different than the past, study the past.

Introduction

It is not an easy task to examine the early development of any technology and extrapolate from this the trends of the future. Clearly, the most important "trend setters" are applications; what are the needs that we can identify today? Application needs tend to push the technology with deliberate, measured steps and although these trends are somewhat less than exciting, they are fairly easy to predict. We usually know where we want to be next month or next year and can work to that end. However, the problem is more complex in scintillation counting by the recognition that it is not an independent discipline but rather a unique blend of chemistry, physics, optics, electronics and computer science. It is the unexpected advance in one of these related disciplines that

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has the potential of triggering new, and perhaps unexpected initiatives that force the technology to hop, skip, and jump down the uncharted research paths of the future. Here, a role reversal occurs when new fundamental developments create surprising applications.

With the relative uncertainty of future developments in basic science, is it possible, or even worthwhile to try, to predict the future direction of scintillation technology? For the short term, perhaps 5 to 10 years, I would say, yes. My thesis is that the informed practitioner of ten years ago had most of the technological input needed to forecast the situation of today. Had such predictions been made they would have, like a vector parameter, a direction and a magnitude. And while we might quibble over the length of the arrow, I believe the direction would be clear.

The role of soothsayer is not a happy one. Be optimistic, and you have your head in the clouds; be pessimistic, and you are an old fuddy-duddy set in your ways. You must also be prepared to suffer the ridicule of your future readers who can directly compare theory and practice. (Soothsayers should never put it in writing!) In spite of this, soothsaying is decidedly fun and since I've already been identified as an incurable optimist, this meeting should be an ideal place for me to give it a try.

My plan is simple. I propose to trace the development of scintillation counting using a rather broad brush. I hope to identify those benchmark developments that gave new direction and vitality to the field. A special objective is to locate in time the gestation period of these events and ask if they could have been predicted. Finally, I will attempt to correlate the emerging research of today with the anticipated needs of tomorrow and hopefully arrive at a realistic portrayal of the future.

During the development of the material for this paper, it was surprising to me that there was no real conceptual boundary between solid, liquid, plastic, gas, organic, and inorganic scintillation counting. Therefore, none will be made here. Also, space limits this discussion to only a few of the key developments in the field. Because of this, much good work will not be discussed. I apologize now if I have left out your very favorite publications. One final caveat. In some cases it has been difficult for me to document priority and/or publication and have therefore, been forced to occasionally rely on secondary sources. In these situations, your indulgence is appreciated. Happy "Ides of May."

Exploratory Phase

Around the turn of the century, the art (science?) of scintillation counting was born in the laboratory of Ernest Rutherford when he demonstrated that an atom has a nucleus. He observed the path of alpha particles as they passed through thin metal foils. He placed a zinc sulfide screen behind the foils and determined the scattered positions of the particles by watching for the flash of light they produced on the sulfide screen. The counting and localization of each flash in the darkened laboratory required great patience, and it was rumored that during this period, Rutherford selected his laboratory assistants more for their visual acuity and "staying power" than their intellectual prowess. This then was the practice of scintillation counting in the early 1900's. Rutherford, or surely one of his assistants, must have thought how nice it would be to have a "light-pulse counter" for this work.

In the 1920's the concept of the photoelectric detector was well known, but these single-stage devices had insufficient sensitivity to detect feeble scintillation pulses. The electronics also left something to be desired. Can photoelectric gain stages be cascaded? Slepian (1) thought so and obtained a U.S. Patent on the idea in 1923. Things started moving and in 1930 the first effective photocathode (Ag_2O-Cs) was developed by Koller (2). 1936 was a good year; Gorlich (3) developed the Cs_3Sb photocathode and Zworykin et al. (4) developed the first PMT of note. It was this device that quickly evolved into the ubiquitous 1P21 tube. With the realization of an efficient light flash detector and the existence of the well characterized ZnS scintillator, who will be the one forever remembered for having the idea to couple the two? As best as I can tell, no one! I have been unable to turn up any evidence that this obvious experiment was ever performed before about 1947. It took 10 years to see this likely combination and test its experimental practicality.

Science

In 1947, word came (5) that H. Kallman, of the Kaiser Wilhelm Institute, had coupled a naphthalene crystal to a home-built PMT and thereby developed the first organic scintillation counter (6). This development initiated a flurry of research activity in national, university, and industrial laboratories alike. Every nuclear physicist and his brother were sticking anything that fluoresced against a PMT to see if it would "count, and it was the unusual radiochemist that did not smell like mothballs or some related compound. In 1948 P. R. Bell showed that anthracene had

twice the light output of napthalene and in the following ten years, hundreds of organic scintillators were described by Kallmann, Furst, Hayes, Ott, their co-workers and others (7,8,9). And although the search for improved organic scintillators still continues today, it is of interest to note that in the last 30 years only a very few good, new candidates have been identified (perhaps the best known being dimethyl-POPOP).

The intensity of effort on the study of organic scintillators was phenomenal; it is hard to believe that the development by Hofstadter of the thallium-activated sodium iodide detector in 1948 (10) would cause a shift in research emphasis that lasted for over 10 years. The first NaI detectors were small and not very efficient but they had the significant advantage that their stopping power for gamma radiation was greater than that of the organic scintillators. Initially, only a Compton distribution was observed experimentally. Then in 1950, McIntyre and Hofstadter (11) described the first photopeak response with NaI and the science of gamma-ray spectrometry was born.

The demonstration of the scintillation properties of NaI touched off a search for other crystal scintillators in a manner similar to that which occurred after Kallmann's experiments with naphthalene. In this case, however, the search was far less fruitful. Although a few materials have proved superior to NaI(Tl) in one or two performance parameters, overall, no other scintillating crystal has been able to generally replace it. It is a rare scientific occurrence when the first proves to be the best.

In the early 1950's, the research emphasis was clearly one of defining the essential characteristics of scintillating systems. Chemists were constructing delicious new solvents and scintillating materials and physicists were making careful measurements of system response. Although there were several obvious assay applications in the early years of scintillation history, only a few were actually developed. Part of the problem can be traced to the very limited availability of electronic counting equipment. Most of the equipment in use was of the home-built variety and was often tailored to a specific measurement task. Also, it was not very good. Photomultiplier tubes were small, insensitive, and noisy and the electronics were complicated and slow. Perhaps more important however, is the circumstance that the field was undergoing explosive growth and it is well known that under this condition, application often takes a back seat to exploration. All of this was soon to change.

In the early 50's, an important innovation occurred in the science of organic liquid scintillation counting that virtually divided the scintillation field into two separate disciplines. In 1953 it was demonstrated that for liquid counting, it was possible to combine the sample and the scintillator solution and thereby realize many unique measurement advantages. This advance is usually credited to Hayes and Gould (12) in a publication in Science. However, my ORNL chauvinism forces me to bring to your attention a description (13) of the internal counting of C-14 and S-35 labeled compounds in a terphenyl scintillator by Ropp and Davidson in an ORNL report some months before the Hayes and Gould paper. The work did not appear in the open literature, however, until 1956 (14). At any rate, the Hayes work was pivotal to the application of liquid counting as it shifted the organic research emphasis from scintillator photophysics to sample assay. Now sodium iodide was selected primarily for counting photon emitters while organic liquid scintillation became the method of choice for the assay of low energy beta emitters. And for the first time in scintillation technology, the advantages of 4π geometry, no sample self absorption, and high detection efficiency of low energy beta emitters could be realized experimentally. From this time on, the exploitation of organic and inorganic scintillation spectrometry diverged considerably.

Although the evolutionary path of these scintillation techniques was now different, they shared many of the hardware technical limitations. Fortunately for both applications, important advances occurred in both light detectors and electronics in the mid 50's. PMT's were made bigger, faster, and less noisy, and in 1955 the multi-alkali photocathode was described (15) that gave rise to a whole new generation of devices.

The electronics situation also changed markedly. On a fundamental level, new design techniques were used to realize faster circuitry that was so badly needed. The practice of pulse-height analysis changed from "eyeballing" a scope to the motor-driven single channel analyzer. This latter device, when used with a matched NaI crystal and PMT, elevated gamma-ray spectroscopy to a new level of sophistication. A little further down the road, the "innovative" idea of the multiple single channel analyzer gave birth to the first primative multichannel system. During this period, the electronics needs for liquid scintillation counting were being addressed but from a somewhat different vantage point. Here, the emphasis was less on linearity and pulse-height analysis and more on light collection efficiency, high gain with low noise, and lower backgrounds. It was fortunate that much of the basic instrumentation for NaI pulse counters (linear amps, discriminators, scalers) had direct application to liquid counting.

An instrumentation turning point for liquid scintillation was the production of the Packard Tri-Carb counter (16). This device used two phototubes in a coincidence detection mode [Morton and Robinson (17) and Kallmann and Accardo (18)] that substantially increased the signal to background response. One tube, the "monitor", was used to activate the coincidence gate while the other tube, the "analyzer", yielded the pulse-height information for analysis and counting. So successful was this device that for years it was the only liquid scintillation counter in widespread use. It can still be seen in many laboratories in one or more of its many design iterations.

Technology

By 1960, scintillation science gave way to scintillation technology. Word had spread of the capabilities of this technique and investigators from a variety of disciplines were anxious to examine chemical and physical phenomena via radiotracers and scintillation counting. New parameters became paramount - sample preparation, instrument calibration, standardization, data handling, etc. Many of these problems ran up against basic limitations in the capabilities of the instrumentation systems in use. The solution to these, and problems not yet envisioned, came into being 35 years ago at the Bell Laboratory; it was called a transistor. This device was a small, low-power amplifier that could replace the large and power-hungry vacuum tube. The creation of the transitor was almost simultaneous with the concept of the stored program digital computer and the synergism between these two technologies created a revolution whose impact is still difficult to appreciate. The transistor-computer research/industrial complex spawned an almost complete conversion from tube to solid-state instruments. At first, transistor systems were carbon copies of tube designs. But very quickly the technology became more adventurous and the impact on scintillation counting more profound. Low noise pre-amps became common. Circuit speed and complexity grew geometrically. The concept of the pulse-height-to-time converter coupled with computer type ferrite memories led to the first viable multichannel analyzer. Scintillation counters were more stable, noise was lower, and standardization and calibration became something you did in the morning and only checked later in the day. New device types led to new design approaches. Integrated circuits reduced size and power requirements and the truly portable scintillation spectrometer became a reality.

In the computation area, momentous changes were also taking place. Gamma-ray spectrum stripping programs became common. First they were implemented on a computer as an adjunct to the spectrometer and later became a part of it. Many types of corrections and standardization routines were handled with relative ease. In liquid scintillation counting, the onerous problems of quench correction and/or dual tracer calculations yielded to the computational power of the instrument. These were heady times!

Now the scintillation field entered a period of rapid and diverse growth that attempted to meet application needs generated in virtually every discipline of experimental science and engineering. For this reason, it is not possible to further define the main development lines in the broad perspective of this paper; scintillation counting was simply everywhere.

The gamma-ray spectroscopists were heavy into neutron activation analysis, decay scheme studies, and an almost endless refinement of equipment and techniques. Industrial applications became employed widely. The assay of radionuclides was commonplace and no one could be without his copy of the Heath catalog of gamma-ray spectra. As was usual, the concerns of the liquid scintillation community were somewhat different. Sample preparation was a general concern and procedures were developed to handle almost any problem - be it solid, liquid, or gas. The use of aqueous samples had high priority and the variety of suggested cocktails grew monthly. Suspension counting, emulsion counting, and total combustion of the sample were researched vigorously. The various aspects of color and fluorescence quenching received a great deal of attention. As noted above, the computational capabilities of the counting instrument led to several new approaches. Tritium counting efficiency was always a practical concern and new light collection optics, noise reduction techniques, and improved sample processing strategies were the main avenues of attack.

By 1970, I believe it would be safe to say that things were pretty much under control. Revolution gave way to evolution and it is difficult to suggest anything during the past 15 or so years that has had a radical effect on the status or direction of scintillation counting (though not necessarily gamma or x-ray spectroscopy). Are we now at a dead end? Is there really nothing creative left to do? I think not!

Directions for the Future

This brief outline of scintillation counting has taken us through three quarters of a century - from embryonic science to sophisticated art. During each decade we could see creative developments from various disciplines coming to focus on the problems in scintillation counting. For many of these developments, several years passed before application was realized; for others, the impact was immediate and dramatic. But now that the field can be considered sufficiently mature, we may well ask if the 80's is the harbinger of the beginning of the end. I recall a comment of the Nobel Lauriate, Isidor Rabi:

> I don't think that physics will ever have an end. I think that the novelty of nature is such that its variety will be infinite - not just in changing forms but in the profundity of insight and the newness of ideas . . . (19).

If we accept this assessment of physics, and extend it to scintillation counting, the most obvious question is what are the disparate neoteric concepts that will come together in the next 10 years to shape new research and applications? Although I know implicitly that contributions will come from a plethora of sources, I sense that major effects will result primarily from technological advances in microelectronics, computer science, and laser research.

The revolution in microelectronics is far from being over. Each day we are learning new ways to exploit the potential of the integrated circuit by designing chips whose performance may yet be improved by an order of magnitude. The small size of these devices is clearly advantageous but it is the increased performance, reliability, and lower cost of this technology that provide the push for increased application. The most intensive demand for increased packing density, speed, and numbers of elements is in the area of computer design. It is here that microelectronics meets its natural mate. In 1980, the demand for integrated circuits reached a world value of about $6 billion. This tremendous financial driving force virtually guarantees continued development of a host of new and different circuit configurations, many of which will be used in scintillation systems. Although the majority of integration design is in the area of digital circuits (and this will be discussed shortly) a fair effort is maintained in the production of improved linear systems. It is these that will find application in the more conventional linear pre-amps, amps, voltage regulators, and servo systems of future scintillation instrumentation. Here, innovative new approaches will

produce distinctive changes in photodetectors and I believe we may see in the near future the first solid-state, multi-stage, non-vacuum photomultiplier device.

These detectors will exhibit high speed, high sensitivity and very low noise and will be easily programmed electrically for optimum S/N. A further enhancement will be the incorporation within the device of a pre-amp as part of its basic struct re. Some advantages of these detectors (that we do not have now with PMT's) will be that they can be fabricated into unusual sizes and shapes, they will be able to be "tuned" for spectral sensitivity, and they could be quite inexpensive. Since hybrid forms of tube-solid state detectors have already appeared in the literature, I would rate the prospect of this development as being very good.

Linear devices will certainly have an impact on scintillation counting but it will be the areas of digital circuits and computer science that will cause the experimental techniques of today to become reminiscences of a simpler age. The most striking characteristic of the microelectronics industry is the continuous decline in the cost of a unit circuit function and the attendant increase in chip complexity. In 1960, there was no commercial production of integrated circuits. Today, units are being produced with 100,000 logic gates on a single chip! These chips are now being fabricated into microprocessors, memory elements, and control systems. The chips, when hung together in an appropriate manner, produce computers whose power we see in evidence every day. An imminent development is the powerful "one chip" computer that combines all functions on one piece of silicon real estate. Can the one chip multichannel analyzer be far behind?

It would be an error to say that computers are only now becoming part of scintillation systems; nothing could be further from the truth. Primative systems were in routine (?) use for scintillation counting in the early 60's. However, I feel that the on-board computer systems that we have today will be considered just as primitive when compared with what will be available in the 1990's.

In the next ten years, the reduced size and cost of computers combined with their sheer number crunching ability will create scintillation measurement systems of unbelievable power. This power will extend far beyond the usual chores of "housekeeping" and data reduction. For liquid scintillation counting, I think you can expect to see these computers being combined with cutting-edge technology in artificial intelligence and computer science. It is not hard for me to imagine the counter of 1990 to start off

the day by running a series of diagnostic studies to determine that it is operating properly. The built-in single chip multi-channel analyzers will be tested for linearity and stability and power supplies, amps, etc. will be ramped through their operating limits. The optical coupling efficiency of the measurement chamber will be checked perhaps using a diode laser. Any unusual circumstances will be brought to the attention of the operator.

When it is time to measure samples, coded vials will be loaded (by robot?) into the machine. The operator will have the option of indicating a preset level of error and/or a default counting time. The instrument will initiate the measurement cycle by taking a background and will process this data in a variety of ways. During the measurement of each sample, the spectrum of the response will be analyzed by pattern recognition techniques to determine the isotopes probably present (along with uncertainty), the effects of both color and fluorescence quenching, sample changes during the course of the count, volume corrections, and may indicate the presence of chemiluminescence. Finally, the true disintegration rate of each nuclide present will be calculated along with the probable measurement error. In some cases of diagnostic interest, the instrument will look into its memory and transform the counting data into a meaningful interpretation of the clinical situation.

When it happens, and it occasionally will, that the instrument makes an error in the work-up of the data, the operator can "instruct" the instrument about the failure and the instrument will "learn" to avoid that problem in the future. In essence, the instrument will re-program itself!

It is now the end of the work day and the instrument will go into another diagnostic self check routine. The unit will test itself all the way down to the sub-assembly level and if a problem is found, it will attempt self-repair by interconnecting previously unused circuits. Should this not be possible, a repair order will be generated (with the necessary charge number!). Of course, the unit will direct the technician to the defective module. All is now quiet in the lab; the only thing that can be heard is the humming of the microfiche generator where all of the data, algorithms, and diagnostics of the day are being recorded for archival storage.

The situation for NaI counting will be similar in concept if not detail. A special application will be in tomography and other types of multidimensional scans. The results will be used to look into both brains and barrels. Key features will remain nuclide

identification (probably isotope ratios too) and self analysis and correction. On the basis of very subtle changes in peak shapes and relative spectral ratios, the requirement for standard measurement geometries for accurate assays will be minimized. I believe it is hard to overestimate the cleverness that will be built into these machines.

Another field that appears to me will have a major impact on scintillation counting is laser spectroscopy and research. Never before has the photochemist had such an exquisite tool as the laser. And much of what was described above for instrument operation will be implemented by self-contained laser diode devices in the instrument. The laser will provide a whole new picture of excitation and emission dynamics of organic scintillator systems and these will lead to whole classes of new and improved cocktails. The use of lasers will be particularly useful for probing the various quenching mechanisms and may eventually lead to the development of quench resistant systems.

A spin-off activity of laser research should also have consequences for scintillation counting. The tunable dye laser has proved to be invaluable for a variety of optical studies. A basic limitation, however, is the available range of really good dye molecules that can be used. There is a great on-going research activity in the search for improved laser dyes. The good molecules exhibit the same general characteristics as good scintillators - high fluorescent efficiency (or low intersystem crossing and non-radiative de-excitation). The search for new and better dye materials will certainly have fallout effects in terms of possible new scintillators. And I would suggest that several good publications are just waiting to be written about new liquid scintillator fluors that are based on laser dye work already completed. I predict that now, after a 15-20 year dry spell, several good, new scintillators will be found.

Conclusion

Although a lot of what I have said may seem fanciful and not with much foundation, I believe there is ample evidence that much of it will come to pass. And if it does, many of you may be asking yourselves about the role you will play in this revolution. But we are not microelectronic technicians or computer engineers. What can we hope to contribute to the field when it seems to be veering considerably off the path of our expertise? The answer, I believe, resides in the fact that we are <u>not</u> microelectronics or computer engineers but rather scintillation scientists.

And the finest computers and detectors in the world will not obviate the need to understand the photo physics and chemistry of scintillation counting. It is true that as tools evolve, so man evolves. And we certainly need to be aware of the new capabilities and future potentials. But the way we will most directly participate in the revolution is to carry our experience, understanding, and training in one hand and our new tools in the other. If I have had even a small effect on your perception of the future, the purpose of this paper will have been met.

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