A HISTORY OF RADIATION DETECTION INSTRUMENTATION

Paul W. Frame*

Abstract—A review is presented of the history of radiation detection instrumentation. Specific radiation detection systems that are discussed include the human senses, photography, calorimetry, color dosimetry, ion chambers, electrometers, electroscopes, proportional counters, Geiger Mueller counters, scalers and rate meters, barium platinocyanide, scintillation counters, semiconductor detectors, radiophotoluminescent dosimeters, thermoluminescent dosimeters, optically stimulated luminescent dosimeters, direct ion storage, electrets, cloud chambers, bubble chambers, and bubble dosimeters. Given the broad scope of this review, the coverage is limited to a few key events in the development of a given detection system and some relevant operating principles. The occasional anecdote is included for interest.

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THE HUMAN DETECTOR

What the eye can see

That we can see radiation has been known for more than one hundred years. Roentgen himself, in his third and final paper dealing with x rays (1897), described "a feeble sensation of light that spread over the whole field of vision" when an x-ray tube on the other side of a door was discharged. Even so, Roentgen was not the first to make such a report. That honor belonged to G. Brandes (1896). While the mechanism is not completely understood, it has been assumed that the visual effects, known as radiation phosphenes, are due to the direct action of the x rays on the visual purple of the retina (Steidley et al. 1989).

An exposure to charged particles can also produce radiation phosphenes. For example, diffuse flashes and streaks of light were reported by astronauts in Apollo missions IX through XV (Tobias and Todd 1974). The diffuse flashes have been attributed to Cerenkov radiation produced by the charged particle component of

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cosmic rays traveling faster than light through the transparent media of the eye. The streaks and pinpoints of light appear to have been due to ionization within the retina itself.

Sight might not be the only human sense by which the body can detect radiation; it might even be possible to feel it! There have been reports (Steidley et al. 1979) that under dry conditions the ionization produced in air by an intense radiation source can make the hairs on the arms stand up and create a tingling sensation.

What the mind can conceive

While an over-reliance on the human senses for the detection of radiation would be misplaced, there is one human faculty that is indispensable: reason. With only this one instrument at his disposal, the Roman poet and philosopher Lucretius (ca. 95 to 55 B.C.) was able to describe how matter was composed of various combinations of different types of atoms which he called seeds: "In many elements in many ways, common to many things amongst them mixt. And oft it matterith greatly with what sort of other seeds these selfsame elements are linked, and in what order, and again, what mutual motions they do give and take. For selfsame seeds build sky and sea and earth, rivers and sun, likewise crops and trees and living creatures . . . entangled by their closelocked shapes, these [atoms] form strong roots of rock or iron's savage strength" (Lucretius 1946). That Lucretius did not predict the possibility of unstable atoms and radioactive decay can only be considered Becquerel's good fortune.

PHOTOGRAPHIC FILM

Principle of operation

Photographic devices for radiation detection and imaging typically employ a physical support (e.g., a glass plate, paper, cellulose acetate) coated on one or both sides with a photographic emulsion. In general, the emulsion consists of silver bromide crystals (ca. $1-2 \mu m$) suspended in a gelatinous matrix. Exposure to radiation promotes some of the electrons in the crystals to the conduction band. Now mobile, these electrons move to impurities in the crystals where they reduce the silver.

^{*} PTP MS 11, Oak Ridge Associated Universities, PO Box 117, Oak Ridge, TN 37831-0117.

For correspondence contact P. W. Frame at the above address, or email at framep@orau.gov.

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Later, during the development of the film (or plates), these specks of metallic silver serve as catalysts that speed up the chemical reduction of the crystals. It is the resulting conversion of the silver bromide to metallic silver that gives the film its dark color (for a more detailed discussion, consult Attix and Roesch 1966). After the film is removed from the developer, it is washed and transferred into a fixative that dissolves the unreduced silver bromide. Any residual fixative is then washed off. Now that the silver bromide has been removed, the film can be exposed to the light and analyzed.

Photographic detection of radiation prior to Roentgen

In 1858, in the second of a series of presentations to the French Academy of Sciences on the subject, Claude Félix Abel Niepce de St. Victor observed that "a design traced on a sheet of cardboard with a solution of uranyl nitrate . . . and applied on a sheet of sensitive paper prepared with silver bromide, imprints its image." Years later, in the 1884 edition of the Encyclopédie Chimique, Alfred Ditte commented that "the salts of uranium are capable, long after the exposure to sunlight, of shining invisible rays around themselves, at least the fact seems to result from the curious experiments of M. Niepce de St. Victor." That the 1903 Nobel Prize awarded for the discovery of radioactivity did not go to Niepce has been described by Paul and Josette Fournier (1990) as a case of collective amnesia. Henri Becquerel did receive the award, and his one documented comment concerning Niepce's experiments (1903) left no doubt as to who he thought deserved it: "Uranium is in quantity so weak on these [photographic] papers that to be able to produce an appreciable impression on the plates that the author [Niepce] used would require several months of exposure. M. Niepce therefore could not have observed the radiation of uranium."

Becquerel might have been right. Chemical vapors from the uranium, not radiation, could have produced the images on Niepce's photographic paper. It is even possible that Becquerel felt a bit of sympathy for Niepce. After all, the action of chemical vapors on his own photographic plates might have explained Becquerel's somewhat embarrassing initial report (1896) that calcium sulfide, as well as uranium, was a strong source of invisible radiation.

The early days of photographic imaging

Wilhelm Roentgen was the first to knowingly employ photography for the detection and imaging of radiation. Without doubt, the most famous of his x-ray images is the one that he took of the hand of his wife Bertha. But it was not elation or a sense of history that Mrs. Roentgen felt upon seeing it. When shown the "ghostly shadow" of the bones of her hand, she shuddered with a vague premonition of death (Glasser 1958).

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In the early days of x-ray photography, extremely long exposure times were required, often an hour or more. Unfortunately, the results were often faint and lacking contrast. To compensate, attempts were made to boost the contrast and image density by making paper prints from the negative image roentgenograms. Faster films were sought out, thicker emulsions were used, double emulsions were tried, and plates were "sensitized" with various types of solutions. One partial improvement, pioneered by Michael Pupin of Columbia University, involved the use of intensifying screens. By placing the photographic plate in contact with a screen of calcium tungstate, the direct action of the x rays on the plate was supplemented with the light emitted by the fluorescent screen. Exposure times were reduced, but at a price: the resulting images, like all the fluoroscopic images of the time, were grainy. Furthermore, any non-uniformities in the intensifying screen were reflected in the images.

At first, glass plates were preferred to paper or film, although the latter was the medium of choice in dentistry where its flexibility made it convenient for positioning in the mouth. Eventually, however, the tide would turn in favor of film, and WWI served as the catalyst. Until then, the glass used in the production of photographic plates had been manufactured in Belgium, but the war in Europe effectively halted the supply. Out of necessity, physicians began a switch to film, a switch that accelerated in the 1920's when safety film (cellulose acetate) became commercially available and significantly faster emulsions were developed (Fuchs 1956).

Film for dosimetry

The first to employ film as a personnel dosimeter seems to have been Rome Wagner, a Chicago manufacturer of x-ray tubes. In his address at the 1907 meeting of the American Roentgen Ray Society, Wagner described how he tried to protect himself from accidental exposures in the course of inspecting malfunctioning x-ray units: "The thing is to know whether you have exposed yourself during the day . . . I concluded that I would not take chances with any ray that would affect a photographic plate, so I carry one in my pocket, and in the evening, after the day's work, I develop this film to see whether I have been exposed." Of course, protection and detection are two different things. Within six months, Wagner died of radiation-induced metastatic cancer of the liver (Brecher and Brecher 1969).

Photographic dosimetry experienced its first major leap forward during World War II as a result of the urgent need to monitor enormous numbers of Manhattan District workers. Film was selected because it was reliable and it required far less handling and servicing than the only alternative, a pocket chamber. The first obstacle to overcome was the over-response of the film (Dupont 502 dental film packets) to low energy photons. The solution that Ernest Wollan and his small group of health physicists came up with was to incorporate a 1 mm thick cadmium filter into the badge to flatten out the response (Pardue et al. 1944). Their second problem was to devise a reproducible method to quantify the darkening of the processed film. Prior to WWII, the methods employed to evaluate film dosimeters were imprecise at best, e.g., at one facility an acceptable exposure was indicated if a newspaper could be read through the processed film (Andrews 1988). The method that Wollan and his coworkers came up with was to measure the blackening with a commercial spectrometer. What was then called "blackening," we refer to today as the optical density: the log of the ratio of the intensity of the light transmitted through an unexposed film to that transmitted through the exposed film. The idea was not new; this same approach had been used decades earlier by radiologists to measure the intensity of x rays (Fig. 1).



Fig. 1. Ernest Wollan's film badge from the Metallurgical Laboratory (ca. 1943). An example of the first mass produced film badge. Wollan was the first to use the title "health physicist."

Nevertheless, as the following recommendations from NBS Handbook 51 (1952) indicate, a purely visual evaluation of film darkening was used long after WWII, at least for low doses: "When visual comparison with control films exposed to known amounts of radiation . . . indicates more than one-fifth of the permissible value, the film densities should be measured with a quantitative densitometer." It might be worth mentioning that there are alternatives to the optical evaluation of film, e.g., the silver content of the processed film can be determined by x-ray fluorescence analysis.

Perhaps the first major development in photographic dosimetry after WWII was the proposal by Tochilin and coworkers (1950) to use multiple filters (aluminum and copper) to estimate the effective energy of the photons to which the film was exposed. Even so, the use of filtration to estimate radiation energy had been pioneered 50 years earlier by the Frenchman Louis Benoist (1902). His "penetrometer," designed to measure the quality (energy) of x rays, consisted of a thin silver disk in the center of a ring of aluminum steps of different thicknesses. The step that produced the same reduction in the x-ray intensity as the silver disk when imaged with film or a fluoroscope was a measure of the radiation quality or energy in "B units."

To be sure, some innovative ideas never panned out. Take the case of the self-developing film dosimeter patented (1954) by Edwin Land of the Polaroid Corporation. The film was contained in a small circular packet along with all the necessary chemical reagents. Whenever or wherever the wearer needed to determine the severity of their exposure, the container containing the chemicals was ruptured and the packet was placed inside the mouth. By squeezing the packet against the roof of the mouth with the tongue, a uniform distribution of the chemicals was ensured over the film. The packet was then left in the mouth for a specified time before being removed and evaluated. The wonderful thing about this was that the processing could be performed indoors or outdoors, in any weather, without any special equipment, at a controlled standardized temperature, 98 degrees Fahrenheit.

CALORIMETERS

The earliest recorded use of calorimetry in the radiological sciences was an attempt by Friedrich Dorn in 1897 to measure the energy emitted from an x-ray tube. He did so by exposing metal plates inside a sealed vessel to the x-ray beam and measuring the increased pressure of the gas in the vessel. It was a logical next step to use calorimetry in the analysis of radioactive materials and the first to do so were Pierre Curie and Albert

Laborde (1904) who in 1903 used the technique to quantify the energy emission rate of radium. These gentlemen would have been pleased to know that the same method would be used half a century later by Wilfred Mann for comparing the radium standards at the U.S. National Bureau of Standards (Mann 1954).

Calorimetric measurements are usually performed by one of four general methods (Posey 1963): measuring the rate of temperature increase in a material with a known heat capacity; measuring the temperature gradient along a heat path (e.g., metal bar) under steady state conditions; measuring the temperature increase in a continual stream of cooling fluid; and measuring liquid evaporation rates. Aside from the fact that the equipment is rugged and easy to use, the undeniable attraction of calorimetry is that it is an absolute measurement of the energy emission rate, a quantity every bit as fundamental as the decay rate. It is especially well suited to the measurement of high activity pure alpha and pure beta emitters (e.g., ⁹⁰Sr in radioactive thermoelectric generators) when it would be impractical to remove the sources from their encapsulations. If calorimetry can be said to have a lukewarm reputation, it is due to the fact that the methodology does not lend itself to routine work.

COLOR DOSIMETERS

The first commercial radiation dosimeter seems to have been the "chromoradiometer." Developed by Guido Holzknecht in 1902, it employed a yellow disk that turned darker following an exposure to x rays. The disk was placed on the portion of the body being x-rayed, and after the exposure its color was compared with a standard scale of various shades of yellow. Each shade corresponded to an incremental exposure of "1 H" (H for Holzknecht), the minimum exposure that would result in a discoloration. One H unit was roughly equivalent to one third of the exposure that would result in erythema. Unfortunately, the change in color was rather subtle and it could be affected by temperature and humidity. For these reasons, and the fact that many in the medical community resented that Holzknecht would not reveal the chemicals he used in his device, the chromoradiometer was not very popular. Over the years, this mystery substance has variously been reported as potassium chloride and sodium carbonate, or hydrogen chloride and sodium carbonate (Glasser 1956; Hudson 1932).

More popular, especially among dermatologists, was the dosimeter developed by Raymond Sabouraud and Henri Noiré (1904), and a very similar device developed by Léonard Bordier (1906). These dosimeters employed small disks (called pastilles) of barium platinocyanide, which would change from a green to a dark

yellow-orange when exposed to x rays. The Sabouraud-Noiré pastilles, positioned half way between the tube's target (anticathode) and the area being exposed, had a simple function. When the pastille color changed from its original light green ("teinte A") to a dark yellow ("teinte B"), the physician knew that a dose appropriate for the treatment of ringworm had been delivered. Bordier's pastilles, placed directly on the patient's body, used the following scale of four colors: teinte I (epilation after 20 days); teinte II (erythema); teinte III (dermatitis); and teinte IV (ulceration and necrosis) (Fig. 2).

Also popular was Robert Kienbock's "quantimeter," which employed strips of silver bromide paper placed on the part of the body being x-rayed (Kienbock 1905). The degree of darkening was compared with a standard scale of 10 shades called X units. This, of course, was a photographic rather than colorimetric dosimeter.

While the relative responses of these dosimetric systems depended on the quality of the x rays, it was commonly assumed that Sabouraud-Noiré's teinte B was more or less equivalent to five Holzkhnect (5 H), or ten Kienbock (10 X) units. These types of dosimeters continued to be used into the 1930's (Hudson 1932), but they ultimately fell victim to the advance of technology, e.g., the Victoreen condenser R chamber that could make accurate and energy-independent measurements of x-ray intensity.

The major virtue of colorimetric dosimeters is their simple mode of operation. That simplicity, and the fact that they can provide an immediate estimate of high doses, has not been lost on the military. In the 1950's, faced with a possible need to determine the survivability of troops that had received significant exposures from a



Fig. 2. Dr. Hampson's Roentgen Radiometer (ca. 1910-1925). Note the six small half-moon shaped pastilles coated with barium platinocyanide. Their color change is determined by reference to the graded scale of 25 shades (0-24) on the rotating disk.

nuclear explosion, the U.S. Army deployed the color dosimeter shown in Fig. 3. Each dosimeter employed five glass vials filled with a radiation sensitive solution: brom-creosol purple dissolved in chloroform. An exposure to gamma radiation turned the solution from purple to yellow (Taplin et al. 1951; Taplin 1956). Although this device used a somewhat different reaction, the basic idea can be traced to the observation by Hardy and Wilcock in 1903 that an iodine containing chloroform solution turned purple when exposed to radium or x rays.

IONIZATION CHAMBERS

The beginnings

Within a couple of months of Wilhelm Roentgen's discovery of x rays, J.J. Thomson (1896a, b) demonstrated that this new type of radiation could make a normally insulating material, such as air, conductive. Together with Ernest Rutherford (1896), he hypothesized that x rays ionized the air by stripping small negatively charged particles, now called electrons, from the molecules of the air. Rutherford and Thomson recognized that if this ionization occurred between two conductive plates given opposite charges, the electric field could prevent a recombination of the ions, and the resulting decrease in the charges on the plates could serve as a measure of the x-ray intensity. Their next step was to modify the system so that a fixed potential was maintained between the plates during the exposure to the x rays. The intensity of the latter was then measured via the chamber's current. An early and crucial observation was that this current would "saturate" at sufficiently high potentials. In other words, all the ion pairs produced in the air of the chamber were being collected and a further increase in the applied high voltage would not change the current. At first their



Fig. 3. U.S. Army colorimetric dosimeter (1950's).

ion chamber was employed to measure the intensity of x-ray beams, but Rutherford soon extended this technique to the analysis of uranium and other radioactive materials (1899).

The ion chamber is a particularly important instrument for the health physicist because the accuracy of its response is close to being energy independent, a characteristic that makes the ion chamber particularly useful for measurements at low energies where the response of other instruments can change rapidly with energy. This energy independence is due to a variety of factors, e.g., the ion chamber is typically constructed of air equivalent walls, it operates in the current mode, and the W value (the average energy absorbed per ion pair produced) is independent of the energy of the charged particles. For what it is worth, Rutherford introduced the concept of the W value in a paper describing his attempts to improve upon Dorn's use of calorimetry to measure the energy output of x-ray tubes (Rutherford and McLung 1900).

Configuration of ion chambers

Since ion chambers don't employ gas multiplication, high electric field strengths are not required. This provides great latitude for the dimensions and spacing of the electrodes, and, as a result, there are probably more different configurations of ion chambers than any other instrument. Nevertheless, as early as 1912, Walter Makower and Geiger could describe two basic forms of ion chambers. The first consists of a cylindrical chamber (usually a few hundred cm³ in volume) with a rod running along the central axis that serves as the anode. In the second basic configuration, the electrodes consist of two parallel conductive plates a few centimeters apart.

Free air ionization chamber

The free-air, or standard, ion chamber is an instrument that everyone has read about but few have actually seen (Fig. 4). As best as this author can determine, the basic design was developed by Gino Failla (1929) in response to the establishment of the Roentgen as the first formal measure of x-ray intensity. The free air ion chamber comes as close as possible to allowing a direct measurement of the quantity exposure. It does so by measuring the charge of the ions collected in a specific volume of air due to the interactions of x rays (or gamma rays)-this volume of air is defined by the dimensions of the parallel plate electrodes and the x-ray beam. There is something of the chicken and egg issue here since the definition of the roentgen was constrained by the desire that the quantity be measurable (one can only wish that the same consideration had been given to the measurability of other quantities). The roentgen was defined in air for two reasons. First, when normalized to mass, air is Health Physics

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Fig. 4. Herb Parker's free-air ionization chamber (1935–1940). The top cover has been removed so that the guard wires on the back wall of the chamber can be seen.

more or less equivalent to human tissue with respect to the absorption of x-ray energy. Second, defining the roentgen in air permits the design of a wall-less detector (i.e., a free air chamber). This is important because a detector wall can complicate measurements by attenuating the photons and serving as a source of secondary radiation. Although the free-air ion chamber has walls, the latter have openings through which an x-ray beam passes unimpeded. In this sense it can be considered "wall-less."

A major concern in the design of the free air ion chamber was to avoid the distortions in the electric field that could occur at the edges of the instrument's sensitive volume. For this reason, Lauriston Taylor's idea (1981) to run guard wires along the front and back walls of the chamber has to be considered a major advance. By reducing the distortions in the electric field, the guard wires allowed the distance between the chamber walls and the sensitive volume to be decreased, something that resulted in a significant reduction in the size and weight of these systems.

Liquid ionization chambers

Following J.J. Thomson's demonstration (1896a, 1896b) that liquids could be ionized by an exposure to radiation, George Jaffe initiated a series of intensive investigations into this phenomenon, work that culminated in his classic paper "The Theory of Columnar Ionization" (1913). That the current in a liquid ionization chamber could be proportional to the intensity of the radiation had possible applications for the measurement of dose in tissue equivalent materials (Taylor 1937). Unfortunately, a number of factors held back the development of a practical detection system, e.g., the long charge collection times made liquid ionization chambers sluggish and the need for very high operating voltages necessitated extreme caution to avoid electrical shock (Adamczewski 1961).

The turnaround that would eventually allow liquid ionization detectors to be described as a "standard piece of instrumentation in elementary particle physics laboratories" (Brassard 1979) can be traced back to the late 1940's when Hutchinson (1948) and Davidson and Larsh (1950) independently observed that radiation could induce pulses of conductivity in liquid argon. In other words, liquid ion chambers could operate in the pulse mode! Two decades later the potential of the liquid ionization chamber would be realized, and to a large extent this was due to the efforts of Steven Derenzo and coworkers in Berkeley (Muller et al. 1971). What they did was develop a liquid argon detector with superior spatial resolution to that of a conventional multiwire proportional counter. While there are differences, liquids behave much the same as high density gases, and this high density provides superior energy absorption and spatial resolution. A key to the success of the liquid argon detector was the use of anode wires only a few micrometers in diameter. The resulting high electric field strength in the vicinity of the anodes permitted the formation of avalanches that were necessary for maintaining a simple readout of the multiwire system. Even though such devices operate as proportional counters, it is often the case that they are erroneously treated as ionization chambers.

ANALYZING THE IONIZATION CHAMBER SIGNAL

Although it is not common, ionization chambers can be operated in the pulse mode. The typical scenario where this would be done involves an alpha-emitting sample located inside the chamber, e.g., the analysis of radon in breath samples of radium dial workers. Not only are the alpha particle pulses large enough to be counted, they even permit pulse height analysis. Indeed, the Frisch grid ion chamber (Frisch 1944), an alpha particle spectrometer, has a resolution that compares favorably to that of semiconductor detectors.

In most situations, however, the small size of the individual pulses necessitates that the ion chamber operate in an integrating mode. There are two ways in which this can be done. The first is to operate the system as a passive condenser wherein the rate at which a stored charge decreases is related to the radiation intensity. In the early days, this decrease in charge was usually measured with an electroscope. The second and more familiar approach is to measure the current from the chamber while maintaining a constant potential between the electrodes. This was often done, at least during the first half of the twentieth century, using a quadrant electrometer.

Electroscopes

The sensing element of an electroscope consists of one or two light moveable leaves (e.g., gold, aluminum) or fibers (e.g., quartz, carbon) that are given a static charge. If a single leaf or fiber is used, the charge causes it to be repelled by a fixed rod that is also charged. If two leaves or fibers are used, the charge causes them to repel each other. During the measurement, the ions produced in the air of the ion chamber are collected by the electroscope's sensing element. This reduces the latter's charge and causes it to move back to its original resting position. The movement of the leaves or fibers might be observed with or without a microscope. In the former case, a scale is incorporated into the microscope optics. In the latter case, a scale might be positioned behind or in front of the leaf. The time required for the leaf or leaves to move a certain number of divisions across the scale gives a relative measure of the intensity of the radiation.

The invention of the electroscope can be, and is, attributable to a number of individuals. Sometimes it is credited to the sixteenth century physician/scientist William Gilbert, even though Gilbert's "electroscope," a magnetic needle on a pivot, was more akin to a compass. Others attribute the invention to John Canton whose electroscope consisted of two pith balls suspended at the end of linen thread. Nevertheless, it is probably fair to say that Tiberius Cavallo and Abraham Bennet were most responsible for developing what we normally imagine an electroscope to be. In 1770, Cavallo suspended Canton's pith balls inside a glass jar and attached metal strips to the inside of the jar to protect the device from the accumulation of charge on the glass. In 1787, Bennet replaced the pith balls and thread with strips of gold leaf attached to a brass rod that extended from the top of the jar.

This basic design remained unchanged through most of the 1800's until the discoveries of x rays and radioactivity spurred the development of instruments that would be capable of more reliable and quantitative measurements. Certainly, the tilted electroscope of CTR Wilson and the Wulf bifilar electroscope have to be included among the more significant of the new designs, as does the electroscope of Charles Cheneveau and Albert Laborde. The key feature of the latter instrument, designed according to the recommendations of Pierre Curie, was that it employed interchangeable ionization chambers (Cheneveau and Laborde 1909). A very similar electroscope developed by Samuel Lind is shown in Fig. 5. Lind had worked at the Curies' laboratory in 1910, and it is probable that while he was there he became familiar with the Cheneveau and Laborde electroscope. In his memoirs (1972), Lind described how he never managed to master the use of the piezoelectric electrometer invented by Pierre Curie, the laboratory's instrument of choice for quantifying radioactive samples. Perhaps he felt more comfortable using the electroscope, and this might have prompted him to develop his own version when he returned to the U.S.

Prior to the development of the first "modern" survey meters in the late 1930's and early 1940's, you used whatever was available when looking for lost sources, e.g., a hand-held fluorescent screen, 16 mm film unrolled down a sewer line. Although electroscopes are not well suited to this type of work, they were used. In



Fig. 5. Lind electroscope (ca. 1920–1930) with two interchangeable chambers. The electroscope is mounted on a flow-through ion chamber used for radon measurements. The chamber to the right with the open door would be used for measuring solid samples.

fact, Robert Taft, author of that cult classic "Radium Lost and Found," published a description of a "simple radium detector" that consisted of an electroscope housed inside a wooden box, an instrument that had an uncanny resemblance to a survey meter (1934).

A problem with early electroscopes was that their relatively large electrical capacitance resulted in a poor sensitivity. This changed with the development of what became known as the Lauritsen quartz fiber electroscope. Invented in 1937 by Charles and Thomas Lauritsen at the California Institute of Technology, its low capacity provided great sensitivity and portability. A commercial version of the Lauritsen electroscope produced by a former technician at CalTech, Fred Henson, proved so reliable that it continued to be used in some laboratories well into the 1970's.

Electroscopes are still used to measure radiation, and the design of the direct reading pocket dosimeter, which is actually an electroscope, has hardly changed in the 70+ years since it was developed by Charles Lauritsen (Fig. 6). The sensitive element is a horseshoe-shaped fiber that can be seen to move across a scale when viewed through a lens at one end of the dosimeter. Although light usually enters through a window on the opposite end of the dosimeter from the lens, a dosimeter developed by Frank Shonka for use at Argonne National Laboratory allows the light to enter through an annular window on the side of the barrel.

Electrometers

The standard role of an electrometer in radiation measurements has been to measure the extremely small currents produced by an ion chamber-if the currents were sufficiently large, a galvanometer could substitute for the electrometer. In the early 1900's the Dolezalek quadrant electrometer, invented at more or less the same time that Roentgen discovered x rays, was the instrument of choice. It employed a butterfly-shaped vane suspended inside four brass quadrants. As the collected charge from the ion chamber changed the electrical balance between the quadrants and the vane, the latter rotated. A beam of light was reflected off a mirror attached to the vane's suspension wire, and as the vane rotated, the reflected beam moved across a scale, often positioned one meter away. The time it took the reflected beam of light to move across a specific number of divisions on the scale



Fig. 6. Pocket dosimeter built by Charlie Lauritsen and given to Robley Evans (1932). The unitless scale reads 0-50. Almost certainly the world's oldest pocket dosimeter.

served as a measure of the ion chamber's current. Lord Kelvin, inventor of the quadrant electrometer, is said to have conceived the idea of using the mirror while watching a beam of light being reflected off a monocle dangling from a ribbon around his neck.

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The Curies preferred to use a piezoelectrometer, a somewhat complicated version of a quadrant electrometer developed by Pierre Curie. Operating in what is known as the "null mode," they prevented a deflection of the electrometer vane by counterbalancing the collected charge from the ion chamber with the charge created by stressing a piezoelectric crystal. Since this involved the addition of weights to a pan suspended from the crystal, the measurements were expressed in units of weight. Timing the positioning of the weights on the pan was tricky, a skill that more than one collaborating scientist (e.g., Sam Lind) never mastered.

The Dolezalek electrometer might have been beautiful to look at, but it was difficult to set up and susceptible to static electricity and vibrations. Ultimately, it would be replaced by easier to use and less expensive devices such as the Wulf string electrometer (Wulf 1914) and the Lindemann torsion electrometer (1924). With these types of instruments, the movement of a fine fiber across a scale was monitored using a microscope.

Although they represented an improvement over the quadrant electrometer, the Wulf and Lindemann electrometers were still mechanical devices and subject to the vagaries of operation that one might expect (Fig. 7). What was needed was a purely electrical method for measuring current, and it had been known since the 1920's that vacuum tubes offered one possible means to do so. By running the ion chamber current through a high resistance to the grid of a vacuum tube, the current of the tube's plate circuit would vary in a fashion that could be measured by a microammeter. The vacuum tubes developed for this purpose (known as electrometer tubes) used low electrode voltages and high vacuums, and they possessed low leakage grid currents. Unfortunately, it was not easy to design a circuit that could compensate for the voltage drifts and spontaneous fluctuations in the emissions of the tubes' filaments. As a result, it took several decades for circuits using electrometer tubes to completely replace mechanical electrometers. The longevity of the latter instruments is indicated by the fact that throughout the 1950's and probably into the 1960's, beta and gamma emitting sources leaving the U.S. National Bureau of Standards were measured with ion chambers coupled to Lindemann electrometers (Mann and Seliger 1958).

That the typical electrometer tube circuit operated in a DC mode meant that the instrument's response was



Fig. 7. Four pi ionization chamber built and used at the National Bureau of Standards (ca. 1953). The collected charge from the chamber is determined with a Lindemann-type electrometer that has been removed from the unit and placed on the lower left corner of the base for visibility.

susceptible to gradual variations in the operating characteristics of the electrical components. Vibrating reed electrometers solved this problem by converting the detector output into an AC signal that could then be amplified. Perhaps the best known of the vibrating reed electrometers were the Cary Model 30 and its successors produced by Applied Physics Corporation of Pasadena California.

The development of portable ionization chambers (i.e., survey instruments) posed an additional challenge because the standard electrometer tube of the 1930's, the FP-54, was too large and had too high a power requirement to be used in anything but laboratory instrumentation. As radiologist Dale Trout recalled (1980), the

breakthrough came when he learned that someone at Northwestern University was manufacturing extremely small electrometer tubes for use in hearing aids. Trout managed to get his hands on some of these and took them to John Victoreen in Cleveland where they collaborated on the production of what was the probably first commercial survey instrument, the Victoreen Model 241 (Fig. 8).

PROPORTIONAL COUNTERS

Thomson and Rutherford's development of the ionization chamber was crucial to the quantitative measurements of x-rays, but the study of radioactive decay called out for a method to count individual particles. The solution came with the development of the proportional counter. In 1901, John Townsend, an old school chum of Ernest Rutherford's, observed that a significant increase in an ion chamber's current could be produced at reduced gas pressures if the high voltage was increased well beyond that at which the saturation current had been reached. Townsend's explanation was that the increased velocity of the electrons traveling to the collecting electrode permitted them to ionize the air molecules. It was this additional ionization that produced pulses large enough to be counted.

Rutherford and Hans Geiger (1908) applied Townsend's observation in the construction of the first counting tube: a hollow brass cylinder (25 cm long and 1.77 cm in diameter) that was sealed at each end with an ebonite stopper and evacuated to a pressure of 20–50



Fig. 8. Ion chamber manufactured by Victoreen (ca. 1945). This model seems to have been the first commercially available survey meter. The cylindrical chamber is located behind the latched door on the front end of the case.

mm of mercury. A thin wire (ca. 0.45 mm in diameter) running along the central axis of the cylinder served as the collecting electrode. The entrance window was a thin mica sheet covering the end of a glass tube (1.5 mm inner diameter) that penetrated one of the stoppers. When an alpha particle entered the detector, the resulting pulse was sufficiently large to produce a visible "kick" of the needle of a quadrant electrometer. Initially the system's response was so sluggish that it couldn't keep up with more than five counts per minute, but later on Rutherford and Geiger discovered that by using photography to record the movement of the fiber of a string electrometer, the system could handle over 1,000 counts per minute (1912).

The first major advance in the design of proportional counters can be attributed to Hans Geiger and Otto Klemperer (1928), and in recognition these detectors were sometimes referred to as Geiger-Klemperer counters (Korff 1950). Ironically, this advance involved the modification of a Geiger ("point") counter rather than an improvement on Rutherford and Geiger's original proportional counter. What Geiger and Klemperer did was place a small sphere at the end of a point counter's central electrode and reverse the tube's polarity so that the central electrode served as the anode (these changes would also be incorporated into the design of Geiger Muller detectors). At low voltages their detector only responded to alphas, but above a "critical voltage" it responded to both alpha and beta particles. This is the true essence of the proportional counter: the ability to operate in the pulse mode and distinguish the signals from different types of radiation (e.g., alpha and beta particles). Another notable characteristic of the proportional counter is that the avalanches associated with a given pulse occur over a limited region of the anode. As a result, the detector's dead time is 10 to 100 times shorter than that of a GM detector (Korff 1950).

No discussion of proportional counters would be complete without acknowledging the contributions of John Simpson at the University of Chicago. Among other things, he built the first gas flow proportional counter capable of resolving alpha pulses in the presence of high beta count rates, he was the first to operate proportional counters using an argon-methane gas mixture, and he developed the first workable air proportional counter (Simpson 1945, 1947, 1948).

One of the most important methods for the primary standardization of radioactive samples has been the use of a four pi counter in which a thin source is sandwiched between two identical windowless gas flow detectors (Mann and Seliger 1958). Although John Simpson (1944) was the first to suggest this approach, the first true four pi detector is considered to be that described by Otto Haxel and Fritz Houtermans in 1948. While these systems usually operated in the proportional mode, they were also capable of functioning as GMs (Fig. 9).

In modern times the most celebrated application of proportional counters has been the construction of imaging systems that can pinpoint the location of radiation interactions over large detector areas. The multiwire proportional counters that made this possible were invented by George Charpak et al. (1968), work for which he received the Nobel Prize. A multiwire proportional counter consists of a series of wire anodes sandwiched between large two cathodes plates which might or might not be segmented. By analyzing the intensity of the signals from the various anodes and/or cathodes segments, the precise location of ionizing events can be determined. These detectors proved particularly useful for imaging the particles produced in high energy accelerators.



Fig. 9. Very early four pi gas flow proportional counter from Hanford (ca. 1950).

GEIGER MUELLER COUNTERS

In 1913, Hans Geiger described what became known as a "point" or "Geiger" counter. The outer wall, the anode, was a brass tube with a mica-covered entrance widow at one end. A short rod running partway along the tube's central axis served as the cathode. The end of this rod, located just underneath the mica window, was ground to a fine point. Fifteen years later, Geiger and Walter Muller (1928a,b) described what can be considered the modern day Geiger Muller (GM) detector: a cylindrical tube with the outer wall serving as the cathode and a fine wire stretched along the axis of the tube serving as the anode.

A problem with the early Geiger Mueller tubes was the fact that the following process could cause a single interaction to trigger a repetitive series of pulses. When the positive ions produced in a given pulse reached the cathode wall, some of them would "knock" electrons off the wall. These electrons would then drift towards the anode and trigger a spurious pulse. In one of the most important developments in the development of GM tubes, the Dutch physicist Adolf Trost discovered that such multiple pulsing could be prevented by adding a small quantity of ethanol to the fill gas (1937). This additive, known as a "quench gas," worked as follows. During the formation of a pulse, each positive ion drifting toward the cathode wall is almost certain to collide with one of the organic quench gas molecules. As a result of the collision, an electron is transferred from the quench gas molecule to the positive ion. The quench gas molecule, which now has a positive charge, drifts towards the cathode, collides with it, and breaks apart. Sufficient kinetic energy is dissipated in the breaking apart of the quench that no electrons are knocked off the cathode. As such, multiple pulsing will not occur. Ethanol was the most common of the early additives, but today isobutane is the quench gas of choice. In some circles, Trost's discovery was attributed to the fact that his glass blower, the best maker of GM tubes in Europe, was an alcoholic. When the latter's methods of manufacturing tubes was investigated, the benefits of ethanol became apparent (Fenton 2000).

Since the quench gas molecules are destroyed during the operation of the detector, an organically quenched tube has a limited lifespan—even if the fragments of the organic quench gas molecule can serve as quenching agents themselves. This limitation on self-quenching GM tubes was overcome by S. H. Liebson and Herbert Friedman (1948) who introduced the use of inorganic halogen quench gases. Since the fragments of broken halogen molecules recombine, unlike broken organic molecules, a halogen quenched tube has an infinite lifespan, at least in theory. In addition, it is less affected by excessive voltages than an organically quenched tube. Another reason why modern GM tubes almost always employ a halogen quench is that their operating voltages will be lower. This is due to the fact that the degree of ionization is greater in the halogen quenched tube collisions between excited fill gas atoms (e.g., neon) and halogen quench gas molecules ionize the latter.

The outsides of organically quenched tubes were usually painted black and/or the insides were coated with aquadag because these tubes tended to be light sensitive. Light sensitivity might have been a problem when it came to measurements of radiation, but it could be an advantage in other applications: at least since 1932 (Locher), GM detectors have been employed as counters for visible and ultraviolet light. In a nice instance of something old becoming new again, the Thomas A. Edison Research Laboratory built a prototype civil defense meter in the early 1960's using a GM tube that had been designed for detecting UV radiation. This inexpensive GM tube, they noted, served as an excellent detector of gamma rays (Figular 1964).

The late 1940's saw a surge of interest in the use of light sensitive GMs as an alternative to photomultiplier tubes (PMTs) for converting the light emissions from scintillators into electrical pulses. GM detectors had a couple of key advantages over PMTs (Mandeville et al. 1950). First of all, the associated electronic circuits were simpler. Second, a usable signal from a PMT required the emission of many electrons from the photocathode, whereas a single photoelectron could initiate a GM pulse. Nevertheless, GM tubes had large dead times and they didn't respond well to the long wavelengths associated with most scintillators. Since these problems remained intractable at a time when steady progress was being made in the development of PMTs, GM tubes only saw limited real world application in scintillation detectors.

It might be worth mentioning that GM detectors can operate in the current as well as the pulse mode. The current from a GM is proportional to the logarithm of the exposure rate over two or three decades, and during the 1950's and 1960's a number of survey meters were constructed that operated in this fashion (Van Duuren et al. 1959).

Before the 1940's, when commercial GM tubes became commonly available, workers usually built their own (Fig. 10). This was no easy task since the construction of a GM tube required the performance of what appeared to be numerous arcane rituals. A modern day health physicist trying to return to his/her roots might consider building a GM tube from scratch. For guidance, let me recommend *Procedures in Experimental Physics*

Fig. 10. Hand-built GM tube (1940's). The wall of the tube is glass and a cylindrical copper tube serves as the cathode. Note that the ends of the tube have been coated with wax to reduce leakage currents.

(Strong 1938) and "Practical Aspects of Radioactivity Measurements" in the June 1951 issue of Nucleonics.

COUNTING THE COUNTS

The first method for recording a count was to observe the scintillations in a spinthariscope or the deflections of an electrometer fiber and write the number down on a piece of paper. Aside from the potential tedium, this approach was susceptible to error, especially at high count rates, and various attempts were made to automate the process. For example, the scintillations could be marked on a moving paper strip, or the deflections of the electrometer fiber could be photographed with a 16 mm movie camera.

Without doubt, the modern era of counting began in 1932 with the introduction by Wynn-Williams of the first binary (scale of two) counter. In the 1920's, vacuum tube circuits had been used to drive mechanical registers, but the latter were so slow that these circuits were pretty much limited to cosmic ray investigations where the counts came few and far between. What the Wynn-Williams circuit provided was a way to reduce high count rates to one that the mechanical register could handle. A typical binary scaler would display the count on five to eight lights and, if required, a mechanical register. As an example (see Fig. 11), there might be six lights numbered 1, 2, 4, 8, 16, and 32—such a scaler would be known as a "scale of 64" unit. With no counts registered, all the lights would be off. The first count would turn on light number 1. When the second count was registered, light 1



Fig. 11. Display for a scale of 64 binary scaler (late 1940's).

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turned off and light 2 came on. With the third count, light 1 would come back on while light 2 stayed on. You've got the idea—the total count was the sum of the numbers of the lights that were on at a given time. At the count of 63, all the lights would be on. With count 64, all the lights went off and the mechanical register turned to indicate a "1." Since calculating the count when the mechanical register had turned over several times required multiplication as well as addition, the good folks who put together the first edition of the *Radiological Health Handbook* included multiplication tables for scales of 64 and 256.

Unfortunately, the mechanical registers of the 1930's weren't designed to work with vacuum tube circuits and they would occasionally "act up." Plagued by this problem, Robley Evans became preoccupied with finding an alternative method for registering counts. The answer came to him one evening in 1936 while playing table tennis in the basement of his home. Perhaps the random clicks of the ball striking the table stimulated his thought processes, perhaps not, but before the night was over Evans had designed the circuitry for the first practical count rate meter. The very next day, he rushed back to his laboratory at the Massachusetts Institute of Technology and built a working unit of what he would call a "speedometer" (Gingrich et al. 1936; Evans 1978[†]).

Although Evan's rate meter couldn't always serve as a suitable substitute for a binary scaler, it was the only real alternative until 1946. That was the year that Victor Regener's scale of ten (decade) counting circuit appeared on the scene (Regener 1946). The decade scaler employed several vertical columns of lights with each column containing ten lights numbered 0 through 9 (bottom to top). The count was indicated by noting which numbers were lit up in each column. No addition, no multiplication. What a concept! Within a couple of years, several commercial versions were being produced. That it took until 1960 or so for decade scalers to supplant binary scalers was primarily due to the fact that the early decade scalers were not as stable as the binary scalers or as capable of operating at high count rates (Newell 1952).

One novel approach worth mentioning was for the instrument to indicate the count by the resting position of a needle that had moved across the meter scale during a timed count. This was the method used in Precision Radiation Instruments' "Royal Scintillator," probably the top-of-the-line portable gamma scintillator of the 1950's.

In my mind nothing can touch the visual appeal of the "glow transfer" (also known as Dekatron) tubes that

[†] Origin of standards for internal emitters. Unpublished. Symposium of the Bluegrass and Hoosier Chapters of the Health Physics Society; September 8–10, 1978.

started to be used in scalers in the early 1950's (Fig. 12). The front panel of such a scaler would have the ends of several (e.g., four or five) of these tubes exposed in a row, and the numbers 0 through 9 would be indicated on the panel around the end of each tube like the numbers on a clock. During the count, a glowing dot would move in a circle around the end of each tube. When the measurement was finished, the count was indicated by the numbers adjacent to the resting positions of the glowing dots. That these tubes were fun to watch, as well as being extremely reliable, might explain why they continued to be used well into the 1980's.

Nixie tubes and several variations thereof came along in the 1960's (Fig. 13). Depending on the type, the ends or the sides of several tubes would be exposed in a row across the face of the scaler. At the end of the count, each tube displayed a single number, 0 through 9. As was the case with glow transfer tubes, the total count was made up of the numbers displayed from left to right. The number displayed by a single tube was formed by a glowing filament bent into the shape of that number. Since each tube needed ten separate filaments, there was some overlap of the different numbers.

Sadly, the vacuum tube as a device for displaying counts has lost its once warm glow. Modern generations of instruments display data via the LED, the LCD, the CRT, or even the PDA.

BARIUM PLATINOCYANIDE: THE FIRST RADIATION DETECTOR

Perhaps the most authoritative description of the first radiation detector is found in an interview of Wilhelm Roentgen conducted by H.J.W Dam for the April 1896 issue of *McClures Magazine*. In it, Roentgen stated that on the eighth of November 1895 "I was working with a Crookes tube covered by a shield of black cardboard. A piece of barium platinocyanide paper lay on the bench there, I had been passing a current through the tube, and I noticed a peculiar black line across the paper



Fig. 12. Two glow transfer tubes (ca. 1960).



Fig. 13. Nixie tube (1960's). Which numbers can you make out?

... The effect was one of which could only be produced in ordinary parlance, by the passage of light." This was how barium platinocyanide provided the means by which Roentgen discovered x rays.

In another interview, this one with the British radiologist James Davidson, Roentgen explained why he happened to have a screen of barium platinocyanide on hand (Patton 1993): "I thought it a suitable substance to use to detect any invisible light the tube might give off." By "invisible light," Roentgen was either referring to ultra violet light, or more likely, the cathode rays he was investigating. As might be imagined, the discovery of x rays created a huge demand for barium platinocyanide, and the resulting price increase was described in an editorial of the March 1896 issue of the Electrical World as the "Roentgen raise."

Part of this demand was for barium platinocyanide to coat the screen of what Thomas Edison would later dub the fluoroscope, a device invented independently by Enrico Salvioni and William Magie in February of 1896. In March of that year, after his staff had investigated more than a thousand substances, Edison reported that calcium tungstate was an even better phosphor than barium platinocyanide, and he selected it for his version of the hand-held fluoroscope (Grigg 1965). The beauty of the fluoroscope was that it was portable and that it permitted an immediate visualization of the x-ray image. Unlike the use of photographic plates or film, no messy and time consuming processing was required (Fig. 14). These advantages were fully appreciated by the New York dentist who developed the Indian Head X-ray Reflector seen in Fig. 15, a 1950's era intra-oral, not to mention illegal, fluoroscope.



Fig. 14. Hand-held fluoroscope manufactured by the Patterson Screen Company (ca. 1920's).



Fig. 15. Indian Head X-ray Reflector (1950's).

SCINTILLATION COUNTERS

Alpha scintillation detectors: ZnS

The scintillation method for counting the alpha particles emitted in radioactive decay was discovered in 1903 by William Crookes, and independently by Julius Elster and Hans Geitel. Crookes' discovery occurred while he was observing the phosphorescent glow of a zinc sulphide (ZnS) screen exposed to ²²⁶Ra. In what turned out to be a stroke of luck, he accidentally spilt some of the radium onto the screen. Hoping to find the tiny bits of lost material, Crookes put the ZnS under a microscope, and what he saw astonished him. Instead of the expected uniform glow, Crookes saw individual flashes of light. This happy event led him to construct the first radiation counter, a device he called the spinthariscope. It consisted of a short brass tube with a zinc sulphide screen at one end and a magnifying lens at the other. A ²²⁶Ra source positioned a few mm above the ZnS

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screen served as the alpha source. When the screen was observed through the lens after dark adapting the eyes, the flashes of light produced by the alpha particles striking the ZnS gave the appearance of a "turbulent sea."

Initially, the spinthariscope was more of a curiosity than a scientific instrument (Fig. 16). It took Erich Regener (1908) to describe a methodology by which this "toy" could be transformed into a reliable tool for quantifying the flux of alpha particles. Regener's technique, employed by Geiger and his undergraduate assistant Ernest Marsden (1909) in their investigations of alpha particle scattering, paid enormous dividends: the results of these studies allowed Ernest Rutherford to unveil the nature of the atomic nucleus (Rutherford 1911). Nevertheless, working with a spinthariscope could be a tedious business as the following quote from James Chadwick testifies: "the normal procedure was for an observer to count for one minute (sometimes less), being relieved by another observer and each observer might have up to 20 periods of one minute each during an experiment. The total duration of an experiment was limited by the decay of the active deposit source as well as by the fatigue of the observers" (Oliphant 1972).

Almost from the beginning, it was recognized that a metal impurity was required in the ZnS for it scintillate. At first copper and manganese were used (Rutherford et al. 1932), but today silver is the activator of choice. That it is even possible to detect the scintillations with the human eye is due to the fact that ZnS emits a very large percentage of the incident alpha particle energy as light,



Fig. 16. Crookes spinthariscope from Robert Millikan's laboratory (ca. 1910–1920). This version of the spinthariscope would have been used for demonstration purposes, not quantitative measurements.

and the fact that the dark-adapted eye is quite sensitive to the wavelength of the light emitted by ZnS.

The first electronic scintillation counter was built by Samuel Curran and W. Baker during World War II as part of the Manhattan Project activities at Berkeley, California. It used an RCA Type 1P21 photomultiplier tube to register the scintillations produced by alpha particles striking a silver activated ZnS screen. When declassified, their 1944 report was published as the Manhattan District document, MDDC 1296 (Curran and Baker 1948). Since this work took so long to appear in the open literature, the credit for the first electronic alpha scintillation detector is sometimes attributed to Marietta Blau and B. Dreyfus (1945) or John Coltman and Hugh Marshall (1947). Blau and Dreyfus used the PMT current as a measure of the intensity of the alpha radiation, while Coltman and Marshall registered the individual pulses. For what it's worth, Ruby Sherr (1947) was the first to employ a PMT in an alpha survey meter.

Photomultiplier tubes and their first applications in radiation measurements

Before there were photomultiplier tubes, there were electron multiplier tubes (Bay 1938). Since the latter didn't possess a photocathode, they weren't used to convert scintillations into electronic pulses. Instead, they were used to perform direct counts of particulate radiation. In a typical application, a beam of electrons or beta particles would be directed onto the first dynode of the tube. The radiation might, or might not, have to pass through some sort of window (Allen 1947, 1948). Multiplier tubes had one significant advantage over the Geiger Mueller counter: a short resolving time.

The first photomultiplier tube to see the light of day was described by Larson and Salinger in 1940 as "a combination of a vacuum type photoelectric cell and an electronic multiplier." While they proposed several potential applications for their invention (e.g., movie projection systems), none of their suggestions involved radiation measurements. It would seem that the first use of a photomultiplier tube in the radiological sciences was in an exposure meter described by Russell Morgan (1942) at a meeting of the Chicago Roentgen Society in April of 1941. More specifically, the meter employed an RCA Type 929 photomultiplier tube coated with crystals of calcium tungstate. The current, as measured by a microammeter, served as a measure of the intensity of an x-ray beam.

The impact of the photomultiplier tube cannot be overstated. Without it, any number of analytical methods as we now know them would have been impossible, e.g., gamma scintillation spectroscopy, liquid scintillation counting, thermoluminescent dosimetry. The invention of the PMT also spurred the development of a whole host of new scintillating materials—by the end of the 1940's, the growing list of scintillators included naphthalene, anthracene, phenanthrene, stilbene, NaI, KI, CaF, fluoranthene, fluorine, $CaWO_4$ (scheelite), and $CdWO_4$, to name a few (Moon 1948; Jordan and Bell 1949).

Gamma scintillation: Napthalene and NaI

The story of organic scintillators begins in warravaged Berlin in late 1945. Hartmut Kallmann had returned there after several years' absence only to find that his old university laboratory had been stripped of its equipment by the Russian army. Although discouraged, Kallmann was determined to continue his research using whatever materials and equipment he could find. Lead paint scraped from the laboratory walls would serve as his radioactive source, and for a detector he decided to couple photographic film to some type of scintillator. To fabricate the latter, he rounded up one of the few things readily available in the bombed out city: moth balls. Melted down, they could be recast as blocks of naphthalene. The system worked, but when Kallmann showed it to an official from the American Military Government, the visitor found it ludicrous. Kallmann suggested that instead of laughing, the American should help by supplying him with the only type of currency worth anything in Berlin's thriving black market: cigarettes. Much to Kallmann's astonishment and delight, 10,000 cigarettes showed up on his desk the very next day. In this way, Hartmut Kallmann assembled the world's first organic scintillator: a naphthalene crystal made from mothballs that was coupled to a PMT purchased with Lucky Strike cigarettes (Deutsch 1948; Hine 1977). Other organic scintillators were quick to make their appearance, e.g., the following year P.R. Bell (1948) and Koski and Thomas (1949) showed that even more efficient detectors could be produced using anthracene and stilbene crystals, respectively.

Nevertheless, it was an inorganic scintillator that proved to have the brightest future. In 1948, Robert Hofstadter reported extremely intense scintillations from sodium iodide crystals to which he had added "a pinch of thallium." Given this material's tremendous potential for gamma spectroscopy, Hofstadter wasn't about to relinquish its exploitation to others. Within two years, he had not only published gamma spectra for a wide variety of radionuclides, he had even described the presence in these spectra of what we now refer to as escape, annihilation and backscatter peaks (Hofstadter and McIntrye 1950).

Liquid scintillation counters

The first to produce detectors by dissolving an organic scintillator in a liquid were Hartmut Kallmann

and Milton Furst at New York University (1950), and George Reynolds and his colleagues at Princeton (Reynolds 1950; Reynold et al. 1950). Kallmann and Furst's detector consisted of anthracene dissolved in toluene. Reynolds and his coworkers employed various combinations of scintillators and solvents, but they obtained their best results with terphenyl dissolved in benzene or xylene. Both groups used coincidence counting to reduce the noise produced by the thermionic emissions from their PMT's photocathodes, a technique Morton and Robinson had employed earlier (1949) with solid scintillation counters.

Originally, liquid scintillators, just like solid scintillators, were designed for external counting applications. It was Maurice Raben from Tufts College Medical School and Nicholaas Bloembergen from Harvard University (1951) who proposed the idea of dissolving the sample in the scintillator: "a simple and geometrically ideal counting system might be obtained by dissolving the material to be counted directly in liquid. This method would facilitate particularly the counting of soluble compounds labeled with a weak beta emitter, such as C-14."

The commercialization of liquid scintillation counting had its origins in the early 1950's at the University of Chicago where James Arnold had constructed his own liquid scintillation counter as a tool for carbon dating. In 1952, Lyle Packard, a colleague of Arnold, left the university to run his own company, and he was immediately asked to build a liquid scintillation counter by George Leroy of the Argonne Cancer Research hospital. Packard couldn't ask for a better customer—Leroy had access to nearly unlimited Atomic Energy Commission funding. Since the counter was to be used for double labeling experiments with ³H and ¹⁴C, Packard named it the Tri-Carb and assigned it the model number 314 (Rheinberger 1999).

The major market for LSC units was the medical bioresearch community where it was not unusual to perform runs with dozens or hundreds of samples. With the original counting systems this was an onerous task. First, each sample container was optically coupled to the PMTs by immersing it in oil. Next, the doors to the shield and freezer were closed—the early counters were housed inside commercial freezers to reduce thermionic noise. Finally, the high voltage was turned on and the count initiated. One can only imagine the joy that accompanied the introduction in 1957 of Packard's automatic 100 sample changer.

Plastic scintillators

The earliest readily available reference to plastic scintillators is in the August 1950 issue of the Physical Review (Schorr and Torney). Amazingly, the Tracerlab company managed to introduce a plastic scintillator into their product line that very year: a polystyrene matrix (still used today) incorporating a terphenyl scintillator. At first, plastic scintillators were viewed as an alternative to external liquid scintillation counters for gamma ray work. The detection efficiencies of plastic and liquid scintillators were not as high as that of sodium iodide, but these detectors had shorter decay times and could be fabricated in much larger sizes. Although it was easier to produce very large detectors with liquid scintillators than with plastic scintillators, the latter had several advantages. They could be produced in a wider range of shapes, and they didn't require a container that would attenuate low energy photons and particulate radiation.

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Since plastic scintillators have a low atomic number, gamma rays interact almost exclusively via Compton scattering. This means that their spectra lack full energy photopeaks, something that rules out plastic scintillators as a serious tool for gamma spectroscopy. Even so, it was recognized early on that the Compton pulse height distribution could provide some degree of information about energy (Brownell et al. 1958).

SEMICONDUCTOR DETECTORS

The ancestors of semiconductor detectors were known as "crystal detectors," solid detectors whose conductivity varied with the intensity of the radiation (Jaffe 1932; Van Heerden 1945). For the most part, however, these were crude devices of questionable utility. The first "modern" semiconductor detector is generally considered to have been built at Bell Laboratories by Kenneth McKay (1951). It was a reverse biased p-n junction detector used to detect alpha particles. At the time, McKay's report did not seem to be a particularly auspicious event. Among other things, the detector was extremely small, and when it came to alpha detection there were far better alternatives, e.g., proportional counters and ZnS scintillators.

By 1960, improvements in their quality and size, as well as the development of practical pulse height analysis systems, revealed the true promise of semiconductors. This promise would be particularly evident at the Seventh Scintillation Counter Symposium held in 1960 in Washington D.C. (Nucleonics 1960), but it was only after being pressured that the organizers allowed the presentation of any papers dealing with semiconductors. After all, this was a scintillation counter symposium. Ironically, the controversy might have had a salutary effect: according to one participant (McKenzie 1979), a competition developed between the different laboratories to report the best resolution. It was here that Stephen Friedland et al. (1960) of Hughes Aircraft reported the A history of radiation detection instrumentation • P. W. FRAME

production of diffused junction silicon detectors for alpha particle spectroscopy, and James McKenzie from Chalk River described detectors with thick enough depletion layers to be suitable for the pulse height analyses of electrons with energies up to several hundred keV.

Nevertheless, it was an instrument described in 1963 by Alister Tavendale and George Ewan of Chalk River Laboratories that truly revolutionized the way we do things: the first lithium-drifted germanium detector capable of truly high resolution gamma spectroscopy, a detector with a resolution more than an order of magnitude better than that possible with NaI detectors. Barely a year later, it would be said that Ge(Li) detectors had "replaced NaI(Tl) scintillation crystals for almost every serious investigation in gamma ray spectroscopy" (Shirley 1965). Who among us has not fantasized about the thrill of experiencing their first high-resolution spectrum after a decade of working with NaIs?

The fly in the ointment during the 1960's was the difficulty in obtaining high quality germanium and silicon. For reasons that had yet to be determined, there was only a 50-50 chance that a germanium or silicon ingot would be good enough to fabricate into a detector (Davis 1967). Eventually, with the financial assistance of the Atomic Energy Commission, methods were developed that yielded reliable predictions about the quality of the material, and by the end of the decade the problem had been solved. Even as this was being accomplished, the nuclear industry was looking towards the next generation of detectors, and efforts began at Oak Ridge National Laboratory to produce germanium with impurity levels below 10^{10} cm⁻³, pure enough that lithium drifting would no longer be required. The payoff came a decade or so later when the first high purity germanium (HPGe) detectors became commercially available.

The superb resolution of a germanium detector is due to the extremely large number of charge carriers associated with its pulses, and this large number of charge carriers results from germanium's narrow band gap (< 1 eV). Unfortunately, the latter characteristic creates a significant potential for thermally generated noise that can only be reduced by operating the detector at low temperatures. Room temperature detectors can be produced using semiconductor materials (e.g., CdTe, CZT) with larger band gaps than that of germanium, but this inevitably involves a sacrifice of resolution, and at least for the time being, a small size that limits efficiency at high energies.

RADIOPHOTOLUMINESCENT (RPL) DOSIMETERS

That an exposure to radiation can alter the luminescent spectrum of a material excited by ultraviolet light, a phenomenon known as radiophotoluminescence (RPL), was reported in 1912 by E. Goldstein. Nevertheless, it wasn't until 1950 that James Schulman at the U.S. Naval Research Laboratory in Washington would propose that RPL could be exploited in radiation dosimetry (Schulman 1950; Schulman et al. 1951).

Schulman's dosimeters consisted of blocks or rods of silver-activated aluminophosphate glass. During an exposure of the glass to radiation, some of the absorbed energy promotes electrons to the conduction band, and these electrons migrate to the silver activation sites. The resulting reduction of the silver atoms at these sites produces a new absorption band around 20 nm, and when the glass is subsequently exposed to light of this wavelength, the electrons at the activation sites are excited. As the electrons deexcite, the glass exhibits an orange luminescence, and the intensity of this luminescence is linearly related to dose. One important feature of radiophotoluminescence is that the color centers are not destroyed by this procedure.

The first RPL dosimeter to actually be deployed was the U.S. Navy's DT-60/PD unit. It consisted of a square block of RPL glass inside a circular black plastic holder that would be hung from the neck or pinned to the clothing (Schulman et al. 1953, 1967). The dosimeter's over-response at low energies was corrected for with a circular lead filter that had a small hole in the center. Without the hole, the filter would eliminate any response to the lowest photon energies. The DT-60 was not used for routine dosimetry since the lowest dose that could be measured was 10 R. Hence, its designation as a "casualty" dosimeter. Similar RPL dosimeters were used by the military in other countries, e.g., France, Japan, Austria (Becker 1968).

During the 1960's, improvements made to the glass by the Toshiba Corporation and the concurrent improvements in the design of the readers increased the sensitivity of RPL dosimeters to the point that they became suitable for routine health physics (Yokota and Nakajima 1967; Becker 1968). RPL dosimeters had numerous advantages: there was little to no variability from one batch to another, they were inexpensive and rugged, they could be reread with no loss of signal, and their accuracy was as good as, or better than, that of film (Becker 1966). Despite this, the use of film was firmly entrenched, and the potential of RPL dosimetry went largely unrealized (Fig. 17).

THERMOLUMINESCENT DOSIMETERS

crystalline materials that have been heated to temperatures below those that would result in incandescence. Health Physics

Fig. 17. Two DT60 radiophotoluminescent glass dosimeters used by the U.S. Navy (ca. 1960). The lower of the two has been opened so that the square block of RPL glass can be seen.

Stripped of its complications, the process works as follows. The absorption of radiation energy promotes electrons to the conduction band where they move to positively charged impurities or defects in the crystalline lattice. At these sites (electron traps), the electrons remain trapped between the valence and conduction bands. This process also creates vacancies (holes) at other impurities or defects. Upon heating, the trapped electrons are freed, and as they return to lower energy levels at least some of them emit light photons. The greater the absorbed dose, the greater the number of the electrons that become trapped at impurities, and the greater the number of light photons that are emitted when the material is heated.

"Glimmering light" was how the seventeenth century chemist Robert Boyle described the first recorded observation of thermoluminescence, an event he witnessed when he heated diamonds by holding them against his naked body in the dark (Horowitz 1984). More than two hundred years later, shortly before Roentgen's discovery of x rays, Wiedemann and Schmidt (1895) demonstrated that a variety of materials could be made thermoluminescent by the exposing them to cathode rays (electrons). Even then the importance of activators (impurities) to thermoluminescence was recognized. In fact, one of the thermoluminescent materials investigated by Wiedemann and Schmidt is still in widespread use: calcium fluoride activated with manganese (CaF₂:Mn). Nevertheless, it was a Russian, Ivan June 2005, Volume 88, Number 6

Borgman, who was the first to demonstrate that x rays and radioactive material induced thermoluminescence (1897).

Farington Daniels was the first to investigate and promote the use of thermoluminescent materials as radiation dosimeters (Daniels et al. 1953). Some of his initial work, conducted in the early 1950's for the military, involved the development of a dosimeter that would permit a rapid determination of the dose in a combat situation. The purpose of this was to estimate the potential for acute radiation sickness among troops operating in an area where a nuclear device had been recently detonated. One of Daniel's prototype dosimeters employed LiF crystals that were heated by igniting flammable pellets. The "reading" involved a simple visual estimate of the intensity of the luminescence. Unfortunately, a soldier's appreciation of the beauty of lithium fluoride's blue light might be tempered somewhat by the knowledge that the glow could indicate a dose of several gray. Perhaps the first use of a thermoluminescent material to determine an individual's dose involved a novel collaboration between Daniels and Marshal Brucer at the Oak Ridge Institute of Nuclear Studies. One of Brucer's cancer patients being treated with ¹³¹I was given a LiF crystal to swallow. A couple of days later, the crystal was recovered and read. For the record, the dose was 60 R.

Despite Daniel's efforts, the behavior of LiF was considered too complex for routine work. One reason for LiF's initial lack of acceptance was the inconsistency in its light output. The cause of the inconsistency would later be traced to the fact that Harshaw had "improved" the quality of its LiF by producing purer crystals unfortunately, the thermoluminescent properties depended on the impurities. While there were alternatives to LiF, Al_2O_3 and $CaSO_4$:Mn for example, they were not without their own problems. The former lacked sensitivity and the latter's traps were too shallow (Schulman 1967).

Thermoluminescent dosimeters certainly had a lot going for them: they were reusable, and the process for reading TLDs lent itself to automation. Nevertheless, it wasn't until the early 1960's that thermoluminescent dosimetry would be viewed as a viable alternative to film, and much of the credit goes to John Cameron for demonstrating the importance of magnesium activation to the TL properties of LiF. Another reason for the increasing acceptance of thermoluminescence dosimetry was the fact that commercial TLD dosimeters and readers were becoming available. As is often the case, supply creates demand.

OPTICALLY STIMULATED LUMINESCENCE (OSL) DOSIMETERS

Optically stimulated luminescence is a very similar phenomenon to thermoluminescence. Indeed, the materials that exhibit optically stimulated luminescence are also thermoluminescent. The difference between the two processes is that the emission of light in OSL is accomplished by illuminating the material rather than by heating it.

It has been known since the 1950's that materials capable of optically stimulated luminescence have the potential to serve as radiation dosimeters (Schulman 1959). Antonov-Romanovsky et al. (1956), who employed infrared light to stimulate luminescence from strontium sulfide, were probably the first to recognize this potential. Nevertheless, for several decades thereafter the main application of OSL would be the dating of sediments.

Ironically, the modern era of OSL dosimetry came about because of an interest in aluminum oxide as a thermoluminescent dosimeter. That aluminum oxide might serve as a TLD was recognized in the 1950's, but a practical dosimeter could not be developed. All this changed in the late 1980's with the production at the Urals Polytechnical Institute in Russia of anion deficient Al₂O₃ grown in the presence of carbon. Mark Akselrod et al. (1990) at Oklahoma State University observed that the great sensitivity of this material, 40 to 60 times that of LiF:Mg,Ti, and its relatively flat energy response, revealed a potential for use as a thermoluminescent dosimeter. More significant, however, was the fact that others saw opportunity in aluminum oxide's major flaw: a susceptibility to light-induced fading. This fading suggested to Brian Markey et al. (1995) that Al₂O₃:C might serve as an ideal OSL dosimeter, and when this possibility was investigated, Al₂O₃:C proved to exhibit even greater sensitivity in the OSL mode than in the TL mode!

At present, aluminum oxide is the only material being used for OSL dosimetry (the Luxel dosimeter of Landauer). It is grown under conditions that produce defects in the crystalline structure due to missing oxygen atoms. Depending on the number of electrons that become trapped at a given defect, the latter is known as an F or F+ center. An F center has two trapped electrons (the site is electrically neutral) while an F+ center has one trapped electron (the site has a charge of +1). F centers serve as hole traps and the F+ centers serve as recombination centers. At present, the identity of the electron traps is unknown. Following the absorption of radiation energy, electrons move to the electron traps and holes move to the hole traps (F centers). When a hole moves to the hole trap, the latter is converted into a recombination center (F+ center) because it loses an electron. Later, when the Al_2O_3 :C is exposed to the stimulating source of light, the electrons that are freed from the electron traps move to the recombination centers (F+ centers). The latter are thereby converted into hole traps (F centers), and, when they are, each newly formed hole trap is left in an excited state. When it deexcites, a photon is emitted.

A major consideration in the design of the OSL reader is to ensure that the light detection system (photomultiplier tube) responds to the light emitted by the OSL material, but not to the stimulating light source. One way in which this is accomplished is to employ an argon laser emitting light in 250 nanosecond pulses as the stimulating light source. Since the system only measures the light emitted by the aluminum oxide during the intervals between the pulses, it cannot mistakenly respond to light from the laser. A different approach is to use light-emitting diodes (LEDs) in a continuous emission mode as the stimulating light source. This has the advantage that it avoids any issues associated with laser safety. By limiting the measurements of the luminescence from the aluminum oxide to wavelengths that don't overlap with the emission spectrum of the LEDs, the latter will not interfere with the measurements.

DIRECT ION STORAGE (DIS) DOSIMETERS

A direct ion storage (DIS) dosimeter is a small volume ion chamber coupled to a non-volatile memory cell (EEPROM), an idea that was patented in 1996 by Jukka Kahilainen of RADOS Technology in Finland. The system is currently in production as both a passive dosimeter and an electronic dosimeter with a real-time readout (Wernli and Kahilainen 2001).

The sensitive element of the dosimeter is an airfilled ion chamber, the dimensions of which are limited to a few mm. The small size of the chamber means that it only requires a potential of 30 volts or less for the electric field to be strong enough to prevent a recombination of the ion pairs formed in the air of the chamber. Prior to use, electrons are injected into the EEPROM's floating gate. This creates the potential between the aluminum wall of the chamber and the gate.

Direct ion storage dosimeters are read by measuring a current between the source and drain of the EEP-ROM—the magnitude of this current reflects the charge on the floating gate. Since the ions produced inside the chamber by an exposure to radiation reduce the charge on the floating gate, the change in the current between the source and drain is related to the radiation exposure. Among the key advantages of DIS dosimeters are their flat energy response, non-destructive readout, and small size.

ELECTRETS

While many of us are inclined to think of the electret as a radiation detector of recent vintage, it is actually another member of the detector baby boom generation of the 1940's and 1950's. The idea of measuring the intensity of radiation via a decrease in the electric charge on an insulating material (e.g., fluorothene), an idea beautiful in its simplicity, was patented by Harry Marvin of the General Electric Company in 1954. In principle, the device is a condenser ion chamber-the ions generated by an exposure to radiation in a volume of encapsulated air are attracted to the electret thereby reducing the stored charge. The difference in the charges before and after deployment serves as the measure of the radiation intensity. One of Marvin's proposed applications was to employ an electret in a dosimeter fashioned like a wrist watch as a means to determine radiation exposures to the hands. Today, of course, electrets are most widely used for measuring indoor radon and gamma ray exposures.

CLOUD CHAMBERS, BUBBLE CHAMBERS AND SUPERHEATED DROP DETECTORS

Cloud chambers

It was a fond recollection of sunlight shining on tiny water droplets in the clouds swirling around the summit of Scotland's Ben Nevis that spurred the attempts in 1895 of Charles Wilson to investigate cloud formation (Keller 1983). The simple apparatus that Wilson, a young graduate student in J.J. Thompson's Cambridge laboratory, constructed to duplicate Nature's magic consisted of an air-filled cylinder, the bottom of which was formed by the top of a piston. Rapidly drawing down the piston increased the chamber volume, and the resulting sudden drop in the temperature of the saturated air led to the formation of liquid droplets. At the time, it was well known that airborne dust particles would serve as the condensation nuclei around which droplets would form, but much to Wilson's amazement droplets formed even in the absence of dust. This led him to propose that electrical charges must be spontaneously forming in the air and that these charges were acting as condensation nuclei.

At the same time that Wilson was mimicking cloud formation, J.J. Thompson was investigating the ionization of gases by Roentgen's newly discovered x rays. Therefore, it was only natural that Wilson would investigate the effect of x rays on "cloudy condensation." As June 2005, Volume 88, Number 6

Wilson probably expected, the x rays increased the number of water droplets by producing the condensation nuclei around which these droplets could form (Wilson 1896). Although the first major application for the cloud chamber was to measure the charge on the ions that served as condensation nuclei, the chamber's biggest role would turn out to be the visualization of the tracks of charged particles traveling through the gas.

In subsequent decades, and the 1930's in particular, the cloud chamber proved to be an invaluable tool for investigating cosmic rays. Indeed, it could be said that "Wilson's expansion chamber is to the atomic physicist what the telescope is to the astronomer" (Lewis 1979). In use, a typical experimental setup consisted of the cloud chamber, a camera, and a GM tube. The latter was used to detect the passage of a cosmic ray, trigger the expansion of the chamber, and activate the camera to record the tracks. Perhaps no one profited more from the cloud chamber than Carl Anderson at the California Institute of Technology. With it, he discovered a number of exotic particles including the positron (1933), work for which he was awarded the Nobel Prize (Fig. 18).

Bubble chambers

Ironically, it was a student of Anderson's, Donald Glaser (1952), who was responsible for taking the air out of the cloud chamber. Glaser had been searching for a way to overcome an inherent limitation of cloud chambers: the low density of the gas. Among other things, the low density made it impractical to build a chamber large enough to visualize the entire tracks of low LET radiation. Glaser's idea, to replace the cloud chamber's gas



Fig. 18. Cloud chamber used by Carl Anderson at the California Institute of Technology (ca. 1935). The piston on this chamber is made of glass so that it can be illuminated from below.

with a superheated liquid, was a stroke of genius said to have come to him while consuming beer in a tavern. In his acceptance speech for the Nobel Prize, Glaser described early experiments in which he opened up heated bottles of beer, with and without radiation sources nearby, to observe any effects the radiation might have on the degree of foaming.

A superheated liquid is a liquid that has been heated to a temperature above its boiling point. It remains in a liquid state until something induces a phase transition to a gas, e.g., the presence of impurities, or increasing the temperature above the superheat limit.

Glaser's first bubble chamber employed a solution of ethyl ether heated to its boiling point. When the moment had come to visualize charged particle tracks, the chamber volume was suddenly increased. The drop in pressure left the liquid at a temperature above its boiling point so that any charged particle (e.g., an electron) traversing the liquid would produce small localized regions of high temperatures (thermal spikes). The resulting evaporation of the liquid at these points produced a trail of bubbles. Later, Luis Alvarez latched onto Glaser's idea and pioneered the construction of huge chambers filled with hundreds of gallons of liquefied hydrogen. These monster chambers, for a time at least, would become the mainstay for imaging the alphabet soup of particles produced in accelerators.

A major problem with both cloud chambers and bubble chambers is the fact that the particle tracks must be recorded on photographs—analyzing photographs is not always a reliable or reproducible method for obtaining data. Ultimately, the bubble chamber was replaced by multiwire proportional counters and semiconductor imaging systems. Since the output from these latter devices is purely electronic, there are far more options available for the storage, display, manipulation, and analysis of the signals. In addition, proportional counters and semiconductor detectors are faster than cloud and bubble chambers because they have no moving parts.

Superheated drop/bubble dosimeters

In the early 1970's, a concerted effort got underway to develop an alternative to NTA film (Kodak Nuclear Film Type A) for neutron dosimetry. The most novel device to arise from this search was developed by Robert Apfel at Yale University (1979), the superheated drop detector. It consists of microscopic droplets of a superheated liquid (e.g., Freon) dispersed throughout an immiscible plastic matrix. Each of these droplets acts like a miniature bubble chamber except that some of the droplets are completely vaporized, not something you would want to happen to a bubble chamber. This is how the process works: if the volume of the microscopic vapor cavities produced in an individual droplet by the passage of a charged particle (e.g., a recoil proton) exceeds a critical size, the vaporization becomes self perpetuating and the entire droplet evaporates to produce a visible bubble. Apfel's initial approach was to estimate the neutron dose by measuring the total volume of the gas. Later, Harry Ing and Chaim Birnboim (1984) at Chalk River developed a similar device, known as a bubble detector, but instead of measuring the total volume of the gas, Ing and Birnboim determined the dose by counting the number of bubbles. This latter approach is particularly versatile because the bubbles can be counted in a number of ways, e.g., with a simple visual inspection, or a computerized scanning analysis. It is even possible to employ a transducer to "hear" each bubble as it is formed, just as you might listen to the "clicks" of a Geiger counter.

FINAL THOUGHTS

This "history" is certainly one that is rich with detail, but in truth it was far too brief to do the subject justice. The choice of instruments was arbitrary, and the discussions all too often took the form of "the first [fill in the blank] was developed by [fill in the blank]." Perhaps the silver lining is that the reader has been presented with innumerable opportunities to search out missing pieces of information and experience the joy that comes when that search is successful. Even if the sought-after information is never found, other jewels of knowledge will be. That's part of what makes the history of radiation protection measurements so fascinating.

Review Articles and Recommended Sources of General Information

- Attix FH. Luminescence dosimetry. In: Proceedings International Conference Luminescence Dosimetry, June 21–23, 1965; 1967.
- Beringer R. Charged particle counters in the premodern period. Nucl Instr Meth 162:15–19; 1979.
- Botter-Jensen L. Luminescence techniques and methods. Radiat Meas 27:749–768; 1997.
- Bromley DA. Evolution and use of nuclear detectors and systems. Nucl Instr Meth 162:1–8; 1979.
- Brooks FD. Development of organic scintillators. Nucl Instr Meth 162:477–505; 1979.
- Brown SC. Theory and operation of Geiger-Muller counters—I the discharge mechanisms. Nucleonics (June):10–22; 1948a.
- Brown SC. Theory and operation of Geiger-Muller counters—II counters for specific purposes. Nucleon-ics (August):50–64; 1948b.
- Cameron JR, Suntharalingam N, Kenney GN. Thermoluminescent dosimetry. Madison, WI: University of Wisconsin Press; 1968.
- Curtiss LF. Measurements of radioactivity. NBS Circular 476:October 15; 1949.

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- Curtiss LF. The Geiger Muller counter. NBS Circular 490:January 23; 1950.
- Ewan GT. The solid ionization chamber. Nucl Instr Meth 162:75–92; 1979.
- Fulbright HW. Ionization chambers. Nucl Instr Meth 162:21–28; 1979.
- Heath RL, Hofstadter R, Hughes EB. Inorganic scintillators—A review of techniques and applications. Nucl Instr Meth 162:431–476; 1979.
- Ing H, Noulty RA, McLean TD. Bubble detectors—A maturing technology. Radiat Meas 27:1–11; 1997.
- Kathren RL. Instrumentation for monitoring and field use. In: Stannard JN, ed. Radioactivity and health a history. Pacific Northwest Laboratory; DOE/RL/ 01830-T59; 1988.
- Kathren RL. Health physics instruments yesterday: some notes on the evolution of health physics instruments. In: Proceedings of the 22nd Midyear Topical Meeting on Instrumentation. McLean, VA: Health Physics Society; 1988: 1–9.
- Kathren RL, Ziemer PL. Health physics: A backward glance. New York: Pergamon Press; 1980.
- Korff SA. Proportional counters—I. Nucleonics June: 5–36; 1950a.
- Korff SA. Proportional counters—II. Nucleonics November:46–52; 1950b.
- Korff SA. Proportional counters—III. Nucleonics January:38–43; 1951.
- Krebs AT. Early history of the scintillation counter. Science 122:17–18; 1955.
- Larsen NA. Dosimetry based on thermally and optically stimulated luminescence. Roskilde, Denmark: Riso National Laboratory; Riso-R-1090(EN); January 1999. Dissertation.
- Lipetz LE. The x-ray and radium phosphenes. Int J Ophthal 39:577–598; 1955.
- Mann WB, Seliger HH. Preparation, maintenance, and application of standards of radioactivity. NBS Circular 594; 1958.
- Pringle RW. The scintillation counter. Nature 166(July 1):11–14; 1950.
- Quimby EH. The history of dosimetry in roentgen therapy. Am J Roentg Rad Ther 54:688–703; 1945.
- Romer A. The restless atom. Garden City: Anchor Books; 1960.
- Shutt RP. Cloud chamber: expansion and diffusion. Nucl Instr Meth 162:379–388; 1979.
- Various authors. Practical aspects of radioactivity instruments. Supplement to June issue of Nucleonics: S-1–S-32; 1951.
- Wyckoff HO, Attix FH. Design of free-air ionization chambers. National Bureau of Standards Handbook 64; 1957.

REFERENCES

- Adamczewski I. Ionization chambers with liquid and their practical application. In: Selected topics in radiation dosimetry. Vienna; IAEA; 191–201; 1961.
- Akselrod MS, Kortov VS, Kravetsky DJ, Gotlib VI. Highly

sensitive thermoluminescent anion-defective Al2O3:C single crystal detector. Radiat Protect Dosim 32:15–29; 1990.

- Allen JS. An improved electron multiplier particle counter. Rev Sci Instr 18:739–749; 1947.
- Allen JS. Particle detection with multiplier tubes. Nucleonics (July):34–39; 1948.
- Anderson CD. The positive electron. Phys Rev 43:491–494; 1933.
- Andrews HL. Laboratory measuring instruments. In: Stannard JN, ed. Radioactivity and health: a history. Richland, WA: Pacific Northwest Laboratory; DOE/RL/01830-T59; 1988.
- Antonov-Romanovsky VV, Keirim-Markus IF, Poroshina MS, Trapeznikova ZA. Dosimetry of ionizing radiation with the aid of infra-red sensitive phosphors. USAEC Report AECtr-2435 pt 1:239–250; 1956.
- Apfel RE. The superheated drop detector. Nuc Instr Meth 162:603–608; 1979.
- Attix FH, Roesch WC. Radiation dosimetry. Vol II. 2nd ed. New York: Academic Press; 1966.
- Bay Z. Electron-multiplier as an electron counting device. Nature 141:284; 1938.
- Becker K. Photographic, glass, or thermoluminescence dosimetry. Health Phys 12:955–964; 1966.
- Becker K. Recent progress in radiophotoluminescence dosimetry. Health Phys 14:17–32; 1968.
- Becquerel H. Sur quelques propietes nouvelles des radiations invisibles emises par divers corps phosphorescents. C R Acad Sci (Paris): 559–564; 1896.
- Becquerel H. Recherche sur une propriete nouvelle de la matiere: activite radiante et spontanee ou radioactivite de la matiere. Paris: Didot; 1903.
- Bell PR. The use of anthracene as a scintillation counter. Phys Rev 73:1405–1406; 1948.
- Benoist L. Definition experimentale des diverse sortes de rayons X par le radiochrometre. C R Acad Sci (Paris) 134:225–227; 1902.
- Blau M, Dreyfus B. The multiplier photo-tube in radioactive measurements. Rev Sci Inst 16:245–248; 1945.
- Bordier L. Radiometric methods. Arch Rontg Ray 11:4–13; 1906.
- Borgman II. La thermoluminescence provoquee par les rayons de M. Roentgen et les rayons de M. Becquerel. Comptes Rendus 124:895–896; 1897.
- Brandes GSB. Preuss Akad Wiss 1:547; 1896.
- Brassard C. Liquid ionization detectors. Nucl Instrum Methods 162:29–47; 1979.
- Brecher R, Brecher E. The rays—a history of radiology in the United States and Canada. Baltimore: Williams and Wilkins Co; 1969.
- Brownell GL, Ellet WH, LeVine HD. Measuring gamma ray spectra with large plastic scintillators. Nucleonics (December):68–70; 1958.
- Charpak G, Bouclier R, Bressani T, Favier J, Zupancic C. The use of multiwire proportional counters to select and localize charged particles. Nucl Instrum Meth 62:202–226; 1968.
- Cheneveau C, Laborde A. Appareils pour la mesure de la radioactivite, d'apres la methode electroscopique. J de Phys 4e serie, tome VIII: 161–174; 1909.
- Coltman JW, Marshall FH. Photomultiplier radiation detector. Nucleonics (November):58–64; 1947.
- Crookes W. The emanations of radium. Proc R Soc Lond 71:405-408; 1903.
- Curie P, Laborde A. Sur la radioactivite des gaz qui se degagent de l'eau des sources thermals. Comp Rend Acad D Sc Par 138:1150; 1904.

- Curran SC, Baker WR. A photoelectric alpha-particle detector. U.S. Atomic Energy Commission Report MDDC 1296; 1944.
- Curran SC, Baker WR. Photoelectric alpha-particle detector. Rev Sci Instr 19:116; 1948.
- Daniels F, Boyd CA, Saunders DF. Thermoluminescence as a research tool. Science 117:343–349; 1953.
- Davidson N, Larsh AE. Conductivity pulses induced in insulating liquids by ionizing radiations. Phys Rev 77:706–711; 1950.
- Davis HL. The day the germanium turned bad. Nucleonics (February):23–24; 1967.
- Deutsch M. Naphthalene counters for beta and gamma rays. Nucleonics (March):58–59; 1948.
- Dorn E. Heating effects of Roentgen rays. Ann Phys Chem 63:160–176; 1897.
- Failla G. Criteria for the design of a standard ionization chamber. Am J Roent Rad Ther 21:47–63; 1929.
- Fenton KB. Geiger Muller counters before and after 1946. In: ANARE Research Notes 102. 50 years of cosmic ray research in Tasmania. Twelfth National Congress of the Australian Institute of Physics; 2000: 61–80.
- Figular LG. Development of a low cost gamma radiation survey meter. Orange, NJ: Thomas A. Edison Research Laboratory; Final Report; Project No. 2134B; 1964.
- Fournier P, Fournier JA. Niepce de Saint-Victor (1805–1870), M. E. Chevreul (1786–1889) et la decouverte de la radioactivite. New J Chem 14:785–790; 1990.
- Friedland SS, Mayer JW, Wiggins JS. Tiny semiconductor is fast linear detector. Nucleonics (February):54–59; 1960.
- Frisch O. British atomic energy report. BR-49; 1944.
- Fuchs AW. Evolution of Roentgen film. Am J Roent Rad Ther Nucl Med 75:30–48; 1956.
- Geiger H. A method of counting alpha and beta rays. Deutsch Phys Ges 15:534–539; 1913.
- Geiger H, Klemperer O. Mode of action of the Geiger point counter. Zeits Physik 49:753–760; 1928.
- Geiger H, Marsden E. On a diffuse reflection of the alpha particles. Proc R Soc 82:495–500; 1909.
- Geiger H, Muller W. Das elektronenzahlrohr i wirkungsweise und herstellung eines zahlrohrs. Phys Zeit 29:839–841; 1928a (in German).
- Geiger H, Muller W. Elektronenzahlrohr zur messung schwachster aktivitaten. Naturwissenschaften 16:617–618; 1928b (in German).
- Gingrich NS, Evans RD, Edgerton HE. A direct-reading counting rate meter for random pulses. Rev Sci Instr 7:450–456; 1936.
- Glaser D. Some effects of ionizing radiation on the formation of bubbles in liquids. Phys Rev 87:665; 1952.
- Glasser O. Technical development of radiology 1906–1956. Am J Roent Nucl Med 75:7–13; 1956.
- Glasser O. Dr. W. C. Rontgen. Springfield: Charles C Thomas; 1958.
- Grigg ERN. The trail of the invisible light. Springfield: Charles C Thomas; 1965.
- Hardy WB, Wilcock EG. On the oxidizing action of the rays from radium bromide as shown by the decomposition of iodoform. Proc R Soc 72:200–204; 1903.
- Haxel O, Houtermans FG. Gleichzeitige emission von zwei elektronen beim radioaktiven zerfall des rubidium 87. Zeit Physik 124:705–13; 1948 (in German).
- Hine GJ. The inception of photoelectric scintillation detection commemorated after three decades. J Nucl Med 18:867–871; 1977.

- Hofstadter R. Alkali halide scintillation counters. Phys Rev 74:100–101; 1948.
- Hofstadter R, McIntrye JA. Gamma-ray spectroscopy with crystals of NaI (Tl). Nucleonics (September):32–37; 1950.
- Holzknecht G. Ueber das chromoradiometer. Fortschr A d Geb D Rontgenstrahlen. 6:102; 1902–1903 (in German).
- Horowitz YS. Thermoluminescence and thermoluminescent dosimetry. Vol I. Boca Raton, FL; CRC Press Inc; 1984.
- Hudson JC. Roentgen-ray dosimetry. In: Glaser O, ed. The science of radiology. Springfield: Charles C Thomas; 1932: 120–138.
- Hutchinson GW. Ionization in liquid and solid argon. Nature 162:610-611; 1948.
- Ing H, Birnboim HC. A bubble-damage polymer detector for neutrons. Nucl Tracks Radiat Meas 8:285–288; 1984.
- Jaffe G. Zur theorie der ionisation in kolonnen. Ann d Phys 42:303–344; 1913.
- Jaffe G. Effect of alpha rays on the passage of electricity through crystals. Phys Z 33:393–399; 1932.
- Jordan WH, Bell PR. Scintillation counters. Nucleonics (October):30–41; 1949.
- Kahilainen J. The direct ion storage dosemeter. Radiat Protect Dosim 66:459–462; 1996.
- Kallmann H, Furst M. Fluorescence of solutions bombarded with high energy radiation (energy transport in liquids). Phys Rev 79:857–870; 1950.
- Keller A. The infancy of atomic physics: Hercules in his cradle. Oxford: Clarendon Press; 1983.
- Kienbock R. Uber dosimeter und das quantimetrische verfahren. Fort Rontgenstr 9:276–295; 1905 (in German).
- Korff SA. Proportional counters—I. Nucleonics (June):5–36; 1950.
- Koski WS, Thomas CO. Scintillations produced by alpha particles in a series of structurally related organic crystals. Phys Rev 76:308–309; 1949.
- Land EH. Radiation detection device. U.S. Patent No. 2, 687:478; 1954.
- Larson SS, Salinger H. Photo-cell multiplier tubes. Rev Sci Instrum 11:226–229; 1940.
- Lauritsen CC, Lauritsen T. A simple quartz fiber electrometer. Rev Sci Instrum 8:438–439; 1937.
- Lewis WB. Early detectors and counters. Nucl Instrum Methods 162:9–14; 1979.
- Liebson SH, Friedman H. Self-quenching halogen-filled counters. Rev Sci Instrum 19:303–306; 1948.
- Lind SC. The memoirs of Samuel Colville Lind. J Tenn Acad Sci 47:3–40; 1972.
- Lindemann FA, Lindemann AF, Kerley TC. A new form of electrometer. Phil Mag 47:577–583; 1924.
- Locher GL. Photoelectric quantum counters for visible and ultraviolet light. Part I. Phys Rev 42:525–546; 1932.
- Lucretius. On the nature of things. Bennett CE translation. New York: Walter J. Black; 1946.
- Makower W, Geiger H. Practical radioactivity measurements. New York: Longman, Green and Co; 1912.
- Mandeville CE, Scherb MV. Photosensitive Geiger counters: their applications. Nucleonics November: 34–38; 1950.
- Mann WH. A radiation balance for the microcalorimetric comparison of four national radium standards. J Res Natl Bur Std 52:277; 1954.
- Mann WB, Seliger HH. Preparation, maintenance, and application of standards of radioactivity. NBS Circular 594; June 11; 1958.
- Markey BG, Colyott LE, McKeever SWS. Time-resolved, optically stimulated luminescence from alpha-Al2O3:C. Radiat Meas 24:457–463; 1995.

- Marvin HB. Method and apparatus for measuring ionizing radiations. U.S. Patent No. 2,695,363; November 23, 1954.
- McKay KG. Electron-hole production in germanium by alpha particles. Phys Rev 84:829–832; 1951.
- McKenzie JM. Development of the semiconductor radiation detector. Nucl Instrum Methods 162:49–73; 1979.
- Moon R. Inorganic crystals for the detection of high energy particles and quanta. Phys Rev 78:1210; 1948.
- Morgan RH. An exposure meter for roentgenography. Am J Roentgenol Radium Ther 47:777–784; 1942.
- Morton GA, Robinson KW. A coincidence scintillation counter. Nucleonics (February):25–29; 1949.
- Muller RA, Derenzo SE, Smadja G, Smith DB, Smits RG, Zaklad H, Alvarez LW. Liquid-filled proportional counter. Phys Rev 27:532–535; 1971.
- National Bureau of Standards, Radiological monitoring methods and instruments. Handbook 51; 1952.
- Newell RR. Binary vs. decade scalers. Nucleonics (February):82–83; 1952.
- Niepce de Saint-Victor. Deuxieme memoire sur une nouvelle action de la lumiere. C R Acad Sci 46:448–452; 1858.
- Nucleonics. Scintillators and semiconductors. Nucleonics reports the scintillation counter symposium. Nucleonics May 18:85–100; 1960.
- Oliphant M. Rutherford, reflections of the Cambridge days. New York: Elsevier Pub Co; 1972.
- Pardue LA, Goldstein N, Wollan EO. Photographic film as a pocket radiation dosimeter. CH-1553; April 8, 1944.
- Patton DD. Roentgen and the "new light"—Roentgen's moment of discovery. Part 3: the genealogy of Roentgen's barium platinocyanide screen. Invest Radiol 28:954–961; 1993.
- Posey JC. Calorimetry for measuring radioactivity. Isotop Rad Tech 1:92–95; 1963.
- Raben MS, Bloembergen N. Determination of radioactivity by solution in a liquid scintillator. Science 114:363–364; 1951.
- Regener E. Uber zahlung der a-teilchen durch die szintillation und die grobe des elektrischen elementarquantums. Verh D D Phys Ges 10:78–83; 1908 (in German).
- Regener VH. Decade counting circuits. Rev Sci Instr 17:185– 189; 1946.
- Reynolds GT, Harrison FB, Salvini G. Liquid scintillation counters. Phys Rev 78:488; 1950.
- Reynolds GT. Liquid scintillation counters. Nucleonics May: 68–69; 1950.
- Rheinberger H. Putting isotopes to work: liquid scintillation counters 1950–1970. Preprint. Max-Planck-Institut fur Wissenschaftsgeschichte; 121; 1999.
- Roentgen W. Further observations on the properties of the x-rays. In: Otto Glasser O, ed. Math u Naturw Mitt a d Sitzgsber Preuss Akad Wiss, Physik-Math; Kl 392; 1897. [Reprinted, Roentgen W, Springfield: Charles C Thomas; 1958.]
- Rutherford E. Uranium radiation and the electrical conduction produced by it. Phil Mag ser 5(xlvii):109–163; 1899.
- Rutherford E. The scattering of the alpha and beta rays and the structure of the atom. Proc Manchester Lit Phil Soc, IV 55:18–20; 1911.
- Rutherford E, Chadwick J, Ellis CD. Radiations from radioactive substances. Cambridge: University Press; 1932.
- Rutherford E, Geiger H. An electrical method of counting the number of alpha particles from radio-active substances. Proc R Soc A 81:141–161; 1908.
- Rutherford E, Geiger H. Photographic registration of alpha particles. Phil Mag ser 6(xxiv):618–623; 1912.

Rutherford E, McLung RK. Energy of Roentgen and Becquerel rays, and the energy required to produce an ion in gases. Phil Trans Roy Soc ser A 196:25–59; 1900.

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- Sabouraud R, Noire H. Traitements des teignes tondantes par les rayons X a l'Ecole Lailler. Presse Med 12:825–827; 1904.
- Schorr MG, Torney FL. Solid non-crystalline scintillation phosphors. Phys Rev 80:474; 1950.
- Schulman JH. Dosimetry of x rays and gamma rays. Washington, DC: Naval Research Laboratory; Report No. 3736; 1950.
- Schulman JH. Solid state dosimeters for radiation measurement. In: Marley WG, Morgan KZ, eds. Progress in nuclear energy. Series XII Health Physics Vol I. New York: Pergamon Press; 1959: 150–159.
- Schulman JH. Survey of luminescence dosimetry. In: Attix FH, ed. Luminescence dosimetry. Proc Int Conf Lumin Dosimetry, June 21–23 1965. 1967:3–33.
- Schulman JH, Ginther RJ, Klick CC. Dosimetry of x-rays and gamma rays by radiophotoluminescence. J Appl Phys 22:1479–1487; 1951.
- Schulman JH, Shurcliff W, Ginther RJ, Attix FH. Radiophotoluminescence dosimetry system of the U.S. Navy. Nucleonics (October):52–54; 1953.
- Schulman JH, Kirk RD, West EJ. Use of lithium borate for thermoluminescence dosimetry. In: Attix FH, ed. Luminescence dosimetry. Proc. Int Conf Lumin Dos June 21–23, 1965. 1967: 113–117.
- Sherr R. Scintillation counter for the detection of alphaparticles. Rev Sci Instr 18:767–770; 1947.
- Shirley DA. Applications of germanium gamma-ray detectors. Nucleonics (March):62–66; 1965.
- Simpson J. Four-pi solid angle G-M counters. Rev Sci Instr 15:119–122; 1944.
- Simpson J. Air proportional counters. United States Atomic Energy Commission; MDDC-1315; October 8, 1945.
- Simpson J. A precision alpha-proportional counter. Rev Sci Instr 18:884–893; 1947.
- Simpson J. Air proportional counters. Rev Sci Instr 19:733– 743; 1948.
- Steidley KD, Eastman RM, Stabile RJ. Observations of visual sensations produced by cerenkov radiation from highenergy electrons. Int J Radiat Oncol Biol Phys 17:685–690; 1989.
- Steidley KD, Zeik GS, Ouellette R. Another 60Co hot cell accident. Health Phys 36:437–441; 1979.
- Strong J. Procedures in experimental physics. New York: Prentice-Hall, Inc.; 1938.
- Taft RB. A simple radium detector. Am J Roentgenol Radiat Ther 31:689–690; 1934.
- Taplin GV. Chemical and colorimetric indicators. In: Hine GJ, Brownell GL, eds. Radiation dosimetry. New York: Academic Press Inc; 1956: 357–410.
- Taplin GV, Douglas CH, Sanchez B. Colorimetric method for dosimetry of 10–100 r. Nucleonics 9(August):73–77; 1951.
- Tavendale AJ, Ewan GT. A high resolution lithium-drift germanium gamma-ray spectrometer. Nuc Instr Meth 25:185–187; 1963.
- Taylor LS. The measurement of x-rays with liquid ionization chambers. Radiology 29:323–328; 1937.
- Taylor LS. X-ray measurements and protection 1913–1964. NBS Special Publication 625; 1981.
- Thomson JJ. On the discharge of electricity produced by the Roentgen rays, and the effects produced by these rays on dielectrics through which they pass. Proc R Soc Lond 59:274–276; 1896a.

Thomson JJ. The Roentgen rays. Nature 53:391–392; 1896b.

- Thomson JJ, Rutherford E. On the passage of electricity through gases exposed to roentgen rays. Phil Mag ser 5(xlii):392–407; 1896.
- Tobias CA, Todd P. Space radiation biology and related topics. New York: Academic Press; 1974.
- Tochilin E, Davis RH, Clifford J. A calibrated roentgen-ray film badge dosimeter. Am J Roentgenol Radiat Ther Nucl Med 64:475–488; 1950.
- Townsend JS. The conductivity produced in gases by the motion of negatively charged ions. Phil Mag 1:198–227; 1901.
- Tracerlab Inc. Tracerlog No. 30. November 1950.
- Trost A. Uber zahlrohre mit dampfzusatz. Zeit Physik 105:399–444; 1937.
- Trout D. Health physics before there were health physicists. In: Kathren RL, Ziemer PL, eds. Health physics: a backward glance. New York: Pergamon Press; 1980: 11–20.

- Van Duuren K, Jaspers AJM, Hermsen J. G-M counters. Nucleonics (June):86–94; 1959.
- Van Heerden PJ. The crystal counter. Amsterdam: North Holland Publishing Co; 1945.
- Wernli C, Kahilainen J. Direct ion storage dosimetry systems for photon, beta and neutron radiation with instant readout capabilities. Radiat Protect Dosimetry 96:255–259; 2001.
- Wiedemann E, Schmidt GC. Ueber luminescenz. Ann Phys Chem Neue Folge 54:604; 1895.
- Wilson CTR. The effect of Roentgen's rays on cloudy condensation. Proc Roy Soc (London) 59:338–339; 1896.
- Wulf T. Phys Zeits 15 15:250, 611; 1914.
- Wynn-Williams CE. A thyratron "scale of two" automatic counter. Proc R Soc (London) A 136:312–324; 1932.
- Yokota R, Nakajima S. Recent improvements in radiophotoluminescent dosimetry. In: Attix FH, ed. Luminescence dosimetry. Proc Int Conf Lumin Dos June 21–23, 1965; 1967: 284–299.