Instrumentation for Radioactivity[1]

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WITH THE ADVENT of the nuclear chain-reacting pile as a result of wartime research in the field of atomic explosives, an extremely important tool became available for widespread use shortly after the close of the war. The immediate large-scale availability of radioactive isotopes, the by-products of bomb manufacture, has paved the way for the introduction of new methods and tools into industry, methods that may well enable us to deal simply with problems hitherto insoluble, tools that high costs have previously confined to the research laboratory.

The usefulness of radioactivity stems from its ability to make its presence known through the disintegration of unstable nuclei, with the emission of energy in the form of electromagnetic radiation or high-speed particles. Regardless of the particular application of a radioactive isotope, suitable means must be employed to establish the amount of activity present, or to determine what changes have occurred in the flux of radiation at a given surface in space as a result of other factors—for example, changes in geometry or changes in interposed absorbing material. The purpose of this paper is to review briefly the instruments required for the detection of the presence of radioisotopes. A number of excellent articles (*1*) that cover this subject, or sections of it, more thoroughly can be found in the literature. Several texts (*7*) also have chapters devoted to various phases of instrumentation.

The most common forms of radiation-detecting devices are based upon the ionization produced in gases by quanta and high-speed particles. Of these detectors, the simplest is the ionization chamber. This is an electrical device that measures the number of ion pairs produced by a particle or quantum in passing between two electrodes. Although any shapes may be used, most commonly the electrodes are constructed in cylindrical or plane parallel geometry, and an appropriate fill-gas is enclosed in the space between them. If such a chamber is exposed to a constant intensity of radiation, and the ionization current is measured as a function of the voltage applied between the electrodes, the current is found to increase with voltage quasi-exponentially to a constant value, a result of the phenomenon of recombination. When the fraction of ions lost by recombination goes to zero, a saturation condition is reached at which no further increase in current is obtained with moderate increase in voltage. Ionization chambers are most frequently operated in the saturation region, because here fluctuations in voltage are not important.

Ionization chambers may be used to record the passage of a single particle through them; in fact, they are able to distinguish between different types of radiation as a result of the different ionizing abilities of the various nuclear emissions. In this case the shape of the voltage pulse that appears on the collector electrode is extremely important. More frequently, chambers are used to measure the average ionization current resulting from a steady flux of radiation. Here statistical variations in the average current are observed, and these variations depend for their magnitude on the average radiation flux and the time constant of the chamber circuit.

If the ionization produced by alpha particles is to be measured in an ionization chamber, the "window" through which the alphas are introduced must be very thin, certainly less than 5 mg/cm^2. Thin sheet mica or stretched nylon or Zapon films about 1/10,000 inch thick, made conducting by a negligibly thin coat of colloidal graphite, make satisfactory alpha chamber windows. Such chambers operate at atmospheric pressure; hence the window will not be subject to a pressure differential. The problem in designing a chamber for use with beta particles is not one of window thickness (unless very low-energy particles are being measured), but is rather one of obtaining a maximum energy absorption in the gas. Ionization currents can be increased by the use of dense gases at high pressure, but even then only a fraction of the energy of a fast beta particle is expended in gas ionization in a chamber of average size. Still different considerations dictate the design of a high-efficiency gamma ray ionization chamber. Here interest lies in converting the photons into high-speed electrons by means of the photoelectric effect, Compton effect, and pair production. These electrons will then produce ion pairs in the fill-gas. To have an efficient gamma-

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to-electron conversion, the window should be of a material having a large gamma-ray absorption coefficient, such as tantalum, tungsten, or lead, and the thickness of the window should be approximately equal to the range of the most energetic secondary electron.[3] For still higher efficiency, the chamber may contain added plates similar to the window.

Almost any gas may be used as a fill-gas in an ionization chamber; in general, the densest ionization conveniently obtainable is desired in the gas. This can be achieved by the use of argon or krypton, at high pressures if necessary. For air at atmospheric pressure, a field of several hundred volts per centimeter between the electrodes is needed to establish a saturation condition. Smaller field strengths are required for saturation in very pure gases, such as hydrogen, nitrogen, and the inert gases in which negative ions are not formed by electron attachment. In purified argon (not ordinary tank argon) at pressures of 7 atmospheres a field of only 70 volts/cm is necessary (*6*), although the field strength required for saturation increases rapidly with increasing gas pressure.

A proportional counter is an ionization chamber in which the number of ion pairs is increased by collision in a region of high electric field. Such counters are most useful when the initial ionization is lower than that which can be measured by using an ionization chamber, or when it is desired to detect one type of radiation in the presence of another. If an ionizing particle passes through the gas of a cylindrical proportional counter with a positive center wire, the electrons will be forced toward the wire by the low field (proportional to $1/r$) until they reach the immediate vicinity of the center wire. In the large field near the wire, the initial electrons will produce other electrons by collisions with the gas atoms or molecules, and avalanches will occur. For each initial electron, A electrons will reach the wire, where A is called the gas amplification factor. It should be noted that the voltage pulse on the center wire arises from the motion of the positive ions formed in the avalanche as they move away from the wire, producing a pulse with a very short rise time because electrons are produced too close to the wire (within a few mean free paths) to give rise to any appreciable induced voltages. The gas multiplication factor A will usually have a value of the order of 10^3; hence an alpha particle that produces about 10^5 ion pairs and a beta particle giving rise to, say, 100 ion pairs, will produce pulses corresponding to 10^8 and 10^5 ions, respectively. A discriminator in the associated electronic circuit can easily be adjusted to detect alpha particles in the presence of high intensities of beta and gamma radiation. To detect betas in the presence of gammas is usually more difficult, but if enough difference exists in their energies, it can be done in this way. In the proportional region, A—and hence the pulse size—will vary rapidly with electrode voltage, and it is necessary to use well-stabilized power supplies or many miniature batteries. If possible, an argon-carbon dioxide or a hydrogen-methane mixture should be used for a fill-gas. Both require less voltage than most other gases and both give stable operation, with good time resolution.

The theory of the operation of Geiger counters is very complex (*9*). A Geiger counter is a proportional counter in which the applied voltage is high enough for the gas multiplication of the initial ions to produce a discharge that spreads along the entire length of the wire. Gas multiplication ceases when the combination of the space charge that is due to the positive ion sheath and the motion of the sheath away from the anode lowers the field below the multiplication threshold. The size of the pulse produced by a counter of this design is independent of the nature of the ionizing event; it is usually great enough to allow recording of the pulse without further amplification. Usually there is a delay averaging a few tenths of a microsecond between the ionizing event and the start of the rise of the Geiger pulse. The pulse ordinarily reaches usable size in a fraction of a microsecond. Pulse length is several microseconds, being determined by the time constant of the amplifier input circuit. The counter is then dead for a period of a few hundred microseconds and then gradually recovers in a period of comparable length. Ionizing events occurring during the dead time cannot be recorded; those occurring during the recovery time are recorded, but as pulses of reduced size.

The common Geiger counter uses a thin wire as an anode and a coaxial metal cylinder as a cathode. The whole arrangement is enclosed in, or forms part of, an airtight chamber that may be evacuated and filled with suitable gases, usually at reduced pressure. The characteristic feature of counters used today is the inclusion of a polyatomic gas as a part of the filling. Numerous filling mixtures have been tried, a common one being 9 parts argon to 1 part ethyl alcohol at a total pressure of 10 cm of mercury. Other polyatomic vapors, such as methane, butane, acetone, xylene, and amyl acetate, may also be used. Counters containing such gases are commonly called "self-quenching"; they might be more accurately termed "non-self-reiniti-ating." The role of the polyatomic molecules (*8*) is a dual one: to absorb ultraviolet quanta and thus eliminate the possible occurrence of the photoelectric effect

[3] Optimum conversion thickness is given somewhat more accurately by $t = ln\frac{\mu_e}{\mu_\gamma} \Big/ \mu_e - \mu_\gamma$ where μ_γ is the gamma absorption coefficient, and μ_e is the absorption coefficient of the electrons.

at the cathode, and to prevent secondary electron ejection by positive ions at the cathode. In an argon-ethyl alcohol Geiger counter, the positive ion cloud will contain both kinds of ions. Since the ionization potential of argon is 15.7 ev and that of ethyl alcohol 11.3 ev, in a collision between an argon ion and an alcohol molecule it is energetically possible for the ion to obtain an electron from the organic molecule. It is not possible, however, for the opposite process to occur—an alcohol ion cannot become a neutral molecule while producing an argon ion. Since each ion may make as many as 10^5 collisions in crossing the counter, the ion cloud reaching the cathode will consist entirely of alcohol ions. The ions that approach very close to the cathode surface will pull electrons from the metal and become neutral molecules. Many of these neutral molecules will be in excited states and, were they argon, could emit photons or liberate a secondary electron by an inelastic collision with the counter wall, either of which would reinitiate the discharge. Polyatomic molecules, however, have a lifetime against dissociation that is much shorter than the time required for either of these processes; consequently there is no mechanism by which the discharge can be reinitiated. It is also necessary to consider the excess energy available when an argon ion becomes neutralized in a collision with an alcohol molecule. This excess, 4.4 ev, is radiated as an ultraviolet quantum and might conceivably reach the cathode to produce a photoelectron and a second avalanche. Fortunately, most polyatomic molecules have broad absorption bands in the ultraviolet; hence they absorb the quanta and dissociate.

Since some of the quench gas is dissociated in each discharge, self-quenching counters have a finite life. A normal counter contains on the order of 10^{20} polyatomic molecules, and about 10^{10} of these are "used up" by dissociation at each discharge. The maximum counter life will then be about 10^{10} counts, and counting will probably become erratic after about 10^8. Counters in which the quench gas is not used up have recently been constructed (*10*) by employing very small amounts (0.1 percent) of chlorine, bromine, or iodine. These gases apparently have one excellent property: after they dissociate, they tend to recombine, thus repairing the quench gas for reuse. There is also a drawback: the halogens are all strongly electronegative gases and therefore tend to form negative ions that can reinitiate the discharge, producing spurious counts. Halogen counters, however, have been operated successfully.

Neutrons may be detected in gas counters of all three types with a fair degree of ease. Fast neutrons may be counted by adding some hydrogen to the fill-gas, for they produce recoil protons that initiate ionization. Slow neutrons may be detected by adding a boron-containing gas such as boron trifluoride to the regular fill-gas, or by applying a thin coat of boron to the inner surface of the cylinder. Neutrons react with the B^{10} isotope to produce an alpha particle and Li^7, both of which ionize intensely. Since the cross section for the neutron-B^{10} reaction follows a $1/v$ law, where v is the neutron velocity, slow neutrons are counted much more efficiently than fast ones. It should also be noted that radioactive gases, such as $C^{14}O_2$, can be counted in any type of gas counter by using the active gas as a part or all of the fill-gas.

For counting weak beta samples, such as C^{14} or S^{35}, a windowless flow counter is desirable. This is essentially a shielded Geiger tube into which solid samples are inserted directly, and through which a constant gas flow is maintained to prevent air contamination.

There are many other instruments available for the detection of particles and quanta. These include electroscopes, electrometers, cloud chambers, electron multipliers, scintillation counters, crystal counters, and photographic films. Of these, the last four named are the most frequently employed. Photographic films are widely used in radiographic work of many varieties. They can be manufactured with emulsions that are sensitive to various ranges of radiation intensity and, to some degree, selectively sensitive to different types of radiation. They can be utilized for purposes ranging from the determination of casting flaws and the radiation exposure of personnel to the investigation of the tracks of single particles.

A crystal counter may be made by plating or painting electrodes on the sides of an appropriate crystal and applying a potential difference across the crystal. A pulse is produced when an ionizing particle raises electrons to the crystal's conduction band. Crystals have the advantages over Geiger counters of fast-rising pulses, no delays in pulse formation, a dead time of about 10^{-8} second, small size, and high efficiency for counting energetic quanta. Whether crystals of a given material will count seems to depend upon purity and crystal perfection. Some that have been reported (*13*) suitable for counting are silver chloride, zinc sulfide, diamond, and thallium bromide-thallium iodide. Silver chloride is an easy crystal to obtain commercially, but its counting properties, unfortunately, depend markedly on the way in which it is prepared and handled. Diamond is probably as good a counting crystal as any, and fortunately there is no correlation between counting ability and price.

In another new and very popular counting technique, radiation is allowed to fall on a crystalline phosphor that stops part, at least, of the radiation and passes on a portion of the energy thus obtained in the form of a scintillation of light quanta. This

flash of light, which is of very short duration—about 10 microseconds' decay constant at the very most—is then reflected onto the photosensitive cathode of an electron multiplier tube or photomultiplier. The photomultiplier then transforms the light pulse into an electrical pulse of sufficient amplitude to actuate a scaling unit. The advantages of scintillation counters over Geiger counters are the same as those of crystal counters. Zinc sulfide is generally used as a phosphor for alpha counting, whereas naphthalene, anthracene, and trans-stilbene are good examples of organic phosphors that may be used for beta and gamma detection (*12*). Efficiencies of 20 percent have been reported (*2*) in counting gamma rays of about 1 mev, using naphthalene, in contrast to the usual 0.1–1 percent efficiency of ordinary Geiger counters. Recently the scintillation properties of certain liquids have been under investigation. The most promising solution discovered by the Princeton group (*11*) consists of 0.5 g of terphenyl in 100 ml of *m*-xylene. This solution counted almost as well as a fairly good naphthalene crystal. Recently phenylcyclohexane has been found to be a slightly better solvent (*5*). Applications of liquid scintillation counters with solutions of activities should be forthcoming soon.

The photomultiplier tube most frequently used in scintillation counting is the RCA type 5819. In it one or more photoelectrons from the cathode are multiplied to a readily measurable number by the ejection of more than one (usually 5–8) secondary electrons per incident primary at each of 10 successive anodes. Photomultipliers are sometimes used by themselves for radiation detection, although in most cases efficiency is not high. They also have one very undesirable characteristic—a residual noise or dark current is always present in the output of the tube as a result of leakage across insulators and of thermal emission from the low work function photocathode. This is usually not important in the case of alpha detection and may be eliminated in working with betas and gammas by cooling the tube to liquid air temperature. For scintillation counter work, an even better method is to use 2 photomultipliers in coincidence (*3*). In this arrangement, backgrounds as low as 5 counts per hour are not uncommon.

At present Geiger counters and ionization chambers are the only radiation-sensitive devices that are available in quantity and suitably reliable for commercial application. Since the stability of Geiger counters over long periods of time is frequently questionable, and their life is inherently limited, ionization chambers are usually employed wherever the radiation level is sufficiently intense. Recently some scintillation counters have appeared on the market, and it is expected that they will be increasingly available in the immediate future. Their advantages over Geiger counters are so great that they are preferable for most applications, particularly those in which good geometry is desired.

All the radiation-detecting devices that have been described require some associated electronic circuit in order to present the information obtained in usable form. If an amplifier is necessary to increase signal size, it should reproduce as faithfully as possible the voltage that appears on the collector electrode, but at the level of 10 or more volts necessary to actuate a scaler or discriminator. Several excellent amplifiers have been designed, most of them being based on the Los Alamos model 501 or on the more recent circuit of Jordan and Bell (*4*). Most of these contain discriminators, that is, a bias adjustment at some stage which allows the further amplification of only those pulses that exceed a certain minimum size. In order to keep the input capacity to the amplifier as small as possible, it is customary to mount the first stage or two of the amplifier as close to the counter as possible. For this purpose, special amplifier tubes, called electrometer tubes, especially constructed to reduce grid current, are very useful. Such tubes operate with low electrode voltages so that electrons do not acquire sufficient energy to ionize the residual gas in the tube. In this way grid currents as low as 10^{-15} amp can be easily obtained—a necessity when the current to be measured is of the order of 10^{-14} amp.

Scaling circuits are required to reduce the counting rate of a Geiger counter—for example, from an ordinary value of, say, 3 or 4 thousand counts per second, to a value that a mechanical register can handle, about 50 counts per second. They are all based essentially upon the Eccles-Jordan flip-flop circuit or a multivibrator action, circuits producing one output pulse for two input pulses. Most scalers thus count in powers of 2; they can, however, by appropriate feedback networks, be made to count in powers of 10, thus reducing the effort of extrapolation for untrained personnel.

By adding an integrator circuit with a long time constant, a scaler may be converted easily into a counting rate meter. Such a circuit will show little response to a single pulse but will adjust itself to the average number of pulses received over one time constant. There will thus be a voltage that will vary with the average number of pulses received per unit time; this can be easily measured. It is equally effective and considerably simpler to feed the counter pulses directly to the integrator circuit. The only poor feature of counting rate meters is the fact that they cannot be made conveniently with accuracies of better than about ± 3 percent.

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The Reflecting Microscope[1]

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THE USE OF REFLECTING SYSTEMS of mirror-pairs in microscope objectives extends the range of achromatism of the microscope through the entire optical spectrum—the infrared, visible, ultraviolet, and vacuum ultraviolet regions. This constitutes the most significant advance since the microscope was designed for use in the ultraviolet region by Köhler (*38*) and combined with quantitative spectroscopic techniques by Caspersson (*16*). The reflecting microscope, which historically dates back about three centuries, has been developed in England, Russia, and the United States. Burch (*14*) in 1939 designed a reflecting objective of numerical aperture (N.A.) 0.65 with an aspheric mirror-pair of Schwarzschild (*52*) aplanats, spherically corrected and coma-free. In 1940 Gershgorin, Radchenko, and Brumberg (*13*) extended the system of Maksutov (*43*) and designed a reflecting objective with a spheric mirror-pair and N.A. 0.5. The combination of reflection and refraction was used in objectives designed by Linfoot (*41*) in 1938 and by Johnson (*32*) in 1941. Grey (*28, 29*) in 1949 described a series of microscope objectives of N.A. 0.4 to 1.0 in which Schwarzschild pairs of spheric mirrors are combined with refracting components. Objectives with spheric mirror-pairs have been designed by Seeds and Wilkins and by Kavanagh (*55, 37*). Drew (*22*) has described a solid reflecting objective.

Specifications for various designs of reflecting objectives are given in Table 1. The linear obscuring ratio of numerical aperture has a maximum permissible value of about 0.4 (*23*), beyond which a deterioration of the image may occur. Burch (*15*) has used aspheric mirror-pairs to reduce the fraction of the numerical aperture obstructed by the convex mirror to 0.2 or less and has added a normal-incidence immersion lens, the surface of which is spherical and concentric with the axial object point, to achieve N.A. 0.98. The transmission of all-mirror systems is limited only by the reflectivity of surfaces and extends through the infrared, visible, and ultraviolet regions. The transmission limits of combined reflection and refraction systems are determined by the elements in the refracting component, which include quartz and fluorite.

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