Applications of Radioactive Isotopes to Measurements

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New applications of radioactive isotopes and radioactive detection procedures to measurement and control processes are described nearly every day. The research and control techniques, both depending on rapid, accurate measurement that is made possible by the application of radioactivity, are among the greatest advantages that have been conferred on the scientific and technical community of the atomic age. The limitations of measurements are the interferences they impose on a system under investigation. The final and inescapable limitation is expressed by the indeterminacy principle. The great advantage of the use of isotopes and radioactivity in measurement is the very small interference with normal processes—that is, the minimal disturbances of the system—that they introduce.

Evidence of the high sensitivity obtainable is the great dilution that can be tolerated in the employment of a radioactive isotope as a tracer. Dilutions of 10^6 are not uncommonly encountered. Quantities of a radioactive material as small as 10^{-4} microcuries can readily be measured. The number of moles represented by an activity of 10^{-4} microcuries depends on the disintegration constant of the particular isotope. For an isotope with a short half-life, such as sodium-24, the number of moles represented by 10^{-4} microcuries is 0.5×10^{-12} micromoles.

In addition to this, the gamma radiation from most radioactive isotopes is weakly absorbed by matter; therefore, this type of radiation can be measured or detected through intervening barriers such as body tissue, tanks, pipes, or reaction vessels. Many types of measurements that would otherwise require the introduction of disturbing probes, transducers, or sampling cannulae can be carried out with greater ease and much less uncertainty.

The interactions with matter of the characteristic radiations frequently provide facile means for determining important characteristics of the intervening matter or absorber itself. Thus, the differential absorption of gamma rays provides a means of radiography; the absorption of beta radiation provides a means of measuring density or thickness; and measurements of neutron recoil or secondary radiation provide a means of determining chemical or physical composition. All such measurements can be made with relative ease and simplicity.

Detection and Measurement of Radiations

The four basic means of detecting and measuring the characteristic radiations are (i) photographic emulsions, (ii) ionization chambers, (iii) Geiger counters, and (iv) scintillation counters.

The alpha, beta, and gamma radiations from radioactive materials all expose conventional photographic films and papers. The use of this means of detection to obtain a pattern of the distribution of a radioactive material or a picture of the differential absorption of the radiation is quite obvious. This is analogous to x-ray radiography with the additional possibility of autoradiography when the system is made to contain its own radiation source. Quantitative use of film is of relatively limited application, but use of it is made in monitoring devices for personnel protection and for measurements of radiation in relatively inaccessible places where electronic detection equipment would be cumbersome.

The radiations from radioactive materials produce ionization in gases. This is the basis for the ionization chamber. Some method for measuring the conductivity of a gas-filled chamber is provided. Since conductivity depends upon ionization, an electric measurement of the number of ions formed within the enclosed gas space is thereby obtained. The product of the number of ions and the energy required for the formation of each ion gives a measure of the energy absorbed from the beam by the ionization chamber. If the absorption characteristics of the chamber are known, the energy of the beam under observation can be determined. For any particular isotope, this energy will be a function of the number of radioactive disintegrations that take place in the source. Hence the concentration of the radiating source or tracer element can be determined. Primarily, the ionization chamber measures some fraction of the total energy of the radiation emitted by a radioactive source, but if the characteristics of the chamber, the geometric efficiency, and the nature of the source are likewise determined, the absolute strength of the source, or its concentration, may be readily calculated. In most applications, however, it is only necessary that these conditions be reproducible or be standardized. Relative measurements of concentration or radiation intensity suffice to provide the desired information for most tracer studies.

The most widely used detector is the Geiger-Müller counter. The Geiger counter tube may be looked upon as an ionization chamber with concentric electrodes at such a potential that the electric field will produce an avalanche of secondary ions whenever a primary ionization occurs. It operates as a gas filled diode below the potential of continuous discharge. It is a triggering device in which a voltage pulse is produced by a discharge initiated by an ionizing particle. Quenching of the discharge is controlled by the external circuit and by the pressure and nature of the gas used to fill the tube. It acts, therefore, as a particle detector or counter. The number of output voltage pulses in a specified time is a measure of the number of primary ionizing encounters that have taken place within the tube in that period of time. Over a quite wide range of output pulses or counts, the Geiger counter yields a number of pulses directly proportional to the number of disintegrations that occur in the radioactive source being measured. It provides a good means for determining relative concentrations provided that standardized conditions of geometry and efficiency are observed.

A more recent development for the detection of radioactivity is the scintillation counter. It depends on the production of fluorescence in a crystal by the absorption of ionizing radiation. When electrons drop into vacancies in the shells left by the production of ion pairs, the energy liberated appears as visible light. It was by the visible counting of these tiny flashes of light in zinc sulfide phosphors that much of the original work in nuclear physics was done. By this method, Rutherford obtained the data that demonstrated the nuclear structure of the atom. The method fell into disuse until the development of photomultiplier tubes provided a sensitive electronic means for

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counting the little flashes of light or scintillations. The most widely used phosphor today is a crystal of sodium iodide activated with thalium.

Two great advantages are offered by the scintillation counter. Its efficiency for the detection of gamma radiation is much higher than that of the Geiger counter. A Geiger counter will detect about 1 percent of the gamma ray photons that enter its sensitive volume; a scintillation counter will detect 10 to 15 percent of the gamma radiation it absorbs. The second advantage is that the scintillation counter produces a pulse of an amplitude that is proportional to the energy of the absorbed photon. The crystal must be thick enough to absorb all the gamma radiation that falls on it. It acts, therefore, not only as a detector of photons but also as a device for the measurement of the photon energy. It provides the particle-counting and -detecting advantages of the Geiger tube with the energy-measuring advantages of the ionization chamber.

Radioactive Isotopes as Tracers

The most versatile uses of isotopes are as tracers in the investigation of chemical and physical processes. The concept of the tracer technique has derived from the early investigations of George Hevesy, who used the naturally occurring radioactive isotopes of lead and bismuth as early as 1920 (*1*). He made use of radium D and thorium B in studies of both chemical exchange reactions in solution and the self-diffusion of metals. Later, by use of radium D and E as tracers, he studied the distribution of lead and bismuth, respectively, in animals. The discovery of artificial radioactivity by Joliot and Curie in 1934 opened the way to a great expansion of the use of isotopes as tracers. The development of the cyclotron made it possible to produce relatively large numbers of isotopes of many of the elements. The nuclear energy program of World War II, culminating in the bomb and the nuclear reactor, made possible the production, in relatively large quantities, of radioactive isotopes of nearly all the elements. The needs of the nuclear energy program also stimulated the rapid development of measuring equipment for the detection and measurement of radioactivity; the continuing needs of the U.S. Atomic Energy Commission in particular have led to the commercial production of numerous adequate instruments for use in the general field of radioactivity.

The basic property of isotopes that makes them useful as tracers is the chemical identity of the isotopes of the same element. The characteristic physical property that serves to identify a particular isotope is independent of the chemical transformations which the element may undergo. The chemical properties depend upon the number and arrangements of the extranuclear, planetary electrons. The number of these electrons is determined by the number of protons and is equal to the atomic number Z of the element. The neutrons contained in the nucleus contribute mass but no electric charge; the number of protons and neutrons together gives the mass number A of a particular nuclear species. Species that contain the same number of protons and hence the same number of extranuclear electrons but differ in the number of neutrons are called isotopes. There is thus a mass difference between the various isotopes of any element. The statement that chemical properties of isotopes of a given element are identical is not, therefore, completely true. The isotopes of a given element all go through the same reactions and form the same compounds. There are differences, however, in reaction rates, which depend upon differences in isotopic mass. Ordinarily these differences are not great enough to be significant, but they may assume importance in some systems, particularly systems that are not in equilibrium.

One of the simplest uses of an isotopic tracer is for the quantitative analysis of a mixture of related substances; the method has been called "isotope dilution." Isotope dilution analysis eliminates the necessity of quantitative isolation.

To determine the concentration of a single component of a mixture by this method, an accurately weighed amount of compound to be measured, marked with an isotopic label, is added to the mixture. After adequate assurance that complete mixing has taken place has been obtained, a sample of the compound is isolated and purified by appropriate means. The dilution of the tracer isotope is measured and the amount of compound originally present in the mixture Q, is calculated from the following equation.

$$\frac{Q+q}{q}=\frac{a_1}{a_2},$$

or

$$Q=q\left(\frac{a_1}{a_2}-1\right).$$

The quantity of the component originally present in the mixture is Q; the labeled sample added is q; the specific activity of the added tracer is a_1; the specific activity of the labeled component after complete mixing is a_2. Specific activity is taken to mean radioactivity per unit volume or unit mass.

Synthesis of Tagged Components

The necessary first step of synthesizing the tagged component may be difficult. A modification of procedure made by Keston, Udenfriend and Cannan (*2*) points the way to simplification of the synthetic procedure and adapts the process to the analyses of very small amounts of materials. In their procedure, a mixture of amino acids was treated with *p*-iodophenyl sulphonyl chloride labeled with iodine-131. Quantitative conversion of all the amino acids to the "pipsyl" derivatives was obtained. Excess of unlabeled pipsyl derivative of the desired amino acid was added to the mixture in known amounts, then isolated and purified. The ratio of its specific activity to that of the original pipsyl chloride used provided data for the determination of the relative amounts of amino acids in the protein hydrolysate from as little as 1.3 milligrams of enzyme protein. An exact set of equations that describe applications of isotopic dilution has been published by Gest and his coworkers (3).

Many times it is desirable to study not only the constituents but also the kinetic properties of a system. Only tracer studies of systems involving first order reactions are considered here, even though a great extension of the general technique is obviously possible. When it is known that the reactions in a system are of the first order and irreversible; that no distinction is made between labeled and unlabeled substances of the same chemical or physical form; that the participation of molecules is purely random and that there is no distinction between old and newly formed reactants; that the system remains in a steady state except for the concentration of tagged molecules; and that the mixing time is short compared with the time in which changes in concentration of the tracer take place—then the application of tracer methods and the mathematical description are comparatively simple. In the system, there may be transfers of substances from one region to another; synthesis or degradation of material; transitions from one chemical form to another; and movement from one specified volume, tank, or compartment to a succeeding one. These changes may be generally described as changes of phase.

The addition to a system meeting the afore-mentioned specifications of a small amount of material homologous to that already present, but labeled with a radioactive isotope, will not disturb the system. The tagged component will undergo the same processes or changes of phase as the normal homolog. The tracer atoms or molecules themselves, however, will not be in equilibrium. The changes in concentration of the tagged material are described by the same equations that govern the changes in phase of the unlabeled material normally present. Since the tagged molecules are distinguishable to the observer, their changes in concentration can be measured to yield the parameters in the pertinent equations.

The concentration of tracer substance as a function of time in any phase is given by the equation

$$X = a_1e^{-k_1t} + a_2e^{-k_2t} + \ . \ . \ . + a_ne^{-k_nt}.$$

If X, the concentration of the labeled substance, is measured at suitable intervals of time t, the parameters a_j and k_j can be evaluated. In practice, the periodic measurements of X are plotted as the ordinate on semilogarithmic paper against a linear time base as the abscissa. The resulting curve is analyzed by subtracting out, as straight lines, the separate exponential components. By extrapolating each component back to zero time, the values of a_j are obtained. The value of k is easily determined from the half-time of each component by the relationship

$$k = \frac{\ln 2}{T_{\frac{1}{2}}}.$$

The half-time ($T_{\frac{1}{2}}$) is the period in which a_j is reduced by one-half and can generally be obtained by inspection.

$$k_j = 0.693/T_{\frac{1}{2}}.$$

The physical or chemical significance of a_j and k_j is often not obvious until other phases of the system have been investigated.

Three simple cases are presented in some detail: (i) output flow from a single phase in the steady state; (ii) constant input rate and exponential output of tracer substance; and (iii) transfer of tagged substance between phases.

Output flow from a single phase in the steady state. It is assumed that the total quantity of substance in the phase remains constant and that input and output in grams or cubic centimeters per second are equal and constant. After labeled substance has been added and completely mixed to make the specific activity in the phase at $t = 0$ equal to A_0 microcuries per gram, the specific activity in subsequent time decreases exponentially.

$$A_t = A_0e^{-kt}.$$

The parameter k in this application has the physical significance of the fractional amount of the total substance in the phase that is replaced in unit time. It can be determined from the plotted measurements as described in a previous paragraph.

If Q is the total amount of substance and r is the rate of input or outflow, then

$$k = \frac{r}{Q}.$$

The reciprocal,

$$\frac{1}{k} = \frac{Q}{r},$$

equals the time required for a replacement of the substance by an amount Q. This frequently is called the turnover time. The value of Q may be determined by isotope dilution at $t = 0$. From Q and k the value of r can be determined.

Constant input rate and exponential output of tracer substance. When the input rate of a tagged substance into a single phase is constant at a value of q microcuries per unit time, but the output rate is proportional to the concentration, the specific activity A_t is given by the expression

$$A_t = \frac{q}{kQ}\ (1 - e^{-kt}).$$

As in the previous case,

$$k = \frac{r}{Q},$$

where Q is the total quantity of substance in the phase and r is the output rate.

Transfer of tagged substance between phases. A rate problem that is of frequent occurrence is that of the transfer of a tracer from one phase to one or more others. Again a steady state is assumed, with a constant amount Q of the unlabeled material in each phase, and constant input and output r.

It is considered that material from initial phase 1 flows directly into phases 2, 3, . . . , n. Rates and total amounts for each phase are given by the expressions

$$k_1 = \frac{r}{Q}\,;\ k_2 = \frac{r_2}{Q_2}\,;\ k_n = \frac{r_n}{Q_n}.$$

At $t = 0$, the specific activity of phase 1 is raised to A_0 microcuries per gram. Its subsequent activity is

$$A_1 = A_0e^{-k_1t}.$$

The specific activity A_2 of phase 2 is zero at $t = 0$, but after a time t,

$$A_2 = A_0\,\frac{Q_1}{Q_2}\cdot\frac{k_1}{k_2 - k_1}\,(e^{-k_1t} - e^{-k_2t}).$$

The activity in phase 2 increases to a maximum in the time

$$t_{\text{max.}} = \frac{1}{k_1 - k_2}\log\frac{k_1}{k_2}.$$

Thereafter, the activity decreases at a rate equal to the smaller value of k.

As k_1 approaches k_2, the equation for A_2 becomes indeterminate and a limiting form must be used:

$$A_2 = A_0\,\frac{Q_1}{Q_2}\,kte^{-kt}.$$

These three simple cases are merely indications of the great power of tracer methods in measurement procedures. More detailed discussions are to be found in the works of Siri (*4*), Sacks (*5*), and Solomon (*6*), together with references to more advanced discussions and applications.

Special Measuring Techniques

Many special measuring techniques are made possible by the interactions with matter of the radiations emitted from a radioactive source. Typical of these is the beta gage.

Measurements with the beta gage are made in either of two ways: (i) by the absorption of the radiation as it passes through a material or (ii) by the back scatter of the radiation from the material. The gage is generally useful for measuring thickness of moving materials in fabrication or concentrations of solutions in chemical processing. A quantitative analysis of the performance of the beta gage has been made by Zumwalt (*7*). A summary of industrial experience has been compiled by Foster (*8*). Improvement in quality control has been consistently obtained.

The gamma radiation provides an especially valuable tool for measurement because it is weakly absorbed. It makes possible the use of external detectors to measure the distribution of a tracer or changes in tracer concentration in closed or inaccessible systems. An illustration of this, now in widespread use, is the technique of measuring the functioning of the thyroid gland by observing, with an external counter, the concentration of an orally administered dose of radioactive iodine.

A further example of a technique in which advantage is taken of the external detection of radiation is a method for measuring the velocity profile of a fluid that was devised by Richardson and coworkers (*9*). The lower part of a vertical tube is filled with a tracer solution, the upper part with tracer-free solution. The flow is brought quickly to constant velocity. An external detector downstream from the interface measures a continuous decrease in the activity as the tracer is displaced. Since flow is more rapid in the center, the detector sees a continuous decrease in tracer layer thickness.

If x is the distance from the initial interface to the detector downstream, the velocity of flow v is given by the expression

$$v = \frac{t}{x}.$$

The relationship between velocity and tracer layer thickness is

$$v/v_0 = 4[y/d - (y/d)^2],$$

where v is velocity at the inner edge of the y layer, v_0 is the velocity at the center of the stream (maximum velocity), and d is the diameter of the pipe. The counting rates at the detector are given by the expression

$$C/C_0 = 4[y/d - (y/d)^2],$$

where C_0 is the rate with the tube full of tracer and C is the rate at time t after flow has started.

Conclusion

The applications of radioactive isotopes reviewed here are of the simplest type, but they indicate the powerful contributions that the development of nuclear physics and engineering have brought to the whole science and art of measurement. These advantages reduce to two major improvements. The first is the increased ability to make observations without grossly disturbing the properties of the system that is being investigated. The second improvement is relief from the necessity of isolating the system to the extent that conventional procedures would otherwise require. It is interesting that the atomic age, which offers tremendous extension of man's muscle, should also open new windows for his observation of himself and his world.

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Mechanical Translation

New Challenge to Communication

Jacob Ornstein

One of the thorniest problems in present-day communication is the translation of important writings, especially in the fields of science and technology, from one language to another. The machine age has done little to alter the translation process, which today, as it was centuries ago, must be laboriously performed at considerable expense by human beings. Moreover, growing concern over the dangerous lag between the appearance of important works in other languages and their translation into English and other tongues has within the past decade focused serious attention on the problem of mechanical translation. Until recently, however, mechanical translation has remained one of those human dreams the realization of which was relegated to some point in the unpredictable future.

It was with the development of the electronic computer that real hope came to be felt concerning the possibility of mechanical translation. The man who first envisioned the possibility of translation by electronic means was apparently Warren Weaver, director of the Natural Sciences Division of the Rockefeller Foundation. First of all in 1945, and more concretely in a memorandum dated 15 July 1949, Weaver raised the question of the feasibility of designing a computer-like machine capable of translating from one language to another. This memorandum was circulated among a number of linguists and scientists; their reactions ranged from high optimism to complete skepticism. Weaver's concepts, nevertheless, aroused considerable interest and stimulated preliminary research in the field.

The problem of mechanical translation is being approached by a growing number of scholars, although much of the research is in the realm of speculation and theory. Among mathematicians, one may mention such figures as Yehoshua Bar-Hillel, formerly of Massachusetts Institute of Technology and now of Hebrew University, whose articles on mechanical translation were accorded considerable space in the 5 April 1954 issue of *Time,* the weekly news magazine. Important research is being carried on at Massachusetts Institute of Technology by Victor H. Yngve of the Research Laboratory of Electronics and William N. Locke, professor of modern languages. Anthony Oettinger of Harvard University's Computation Laboratory has also been conducting experiments. Erwin Reifler of the University of Washington has concerned himself with the elaboration of a mechanical dictionary. An active group, headed by William Bull and Victor Oswald, is working on problems of mechanical translation at the University of California at Los Angeles.

The first successful experiment in performing mechanical translation was the result of a joint project undertaken by Georgetown University's Institute of Languages and Linguistics, Washington, D.C., and the International Business Machines Corporation. Considerable publicity followed their demonstration at I.B.M. headquarters in New York on 7 January 1954 of the translation of more than 60 sentences from Russian into English. An invited group of government officials, linguists, and scientists watched a typist who knew no Russian type the sentences, which had been transliterated into the Roman alphabet, on an electric card punch and feed them into the "electronic translator," which produced accurate English translations. The sentences were from the workaday fields of science, technology, communications, and international affairs. The following are a few examples of the transliterated Russian sentences and the English equivalents:

Myezhdunarodnoye ponyimanyiye yavlyayetsya vazhnim faktorom v ryeshyenyiyi polyityichyeskyix voprosov.
International understanding constitutes an important factor in decision of political questions.

Dorogi stroyatsya yiz byetona.
Roads are constructed from concrete.

Komandyir poluchayet svyedyenyiya po tyelyegrafu.
A commander gets information over a telegraph.

Vyelyichyina ugla opryedyelyayetsya otnoshyenyiyem dlyiny dugi k radyiusu.
Magnitude of angle is determined by the relation of length of arc to radius.

Obrabotka povyshayet kachyestvo nyeftyi.
Processing improves the quality of crude oil.

It is revealing to consider in some detail the background of the Georgetown-

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