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RADIOISOTOPE TECHNIQUES IN CLINICAL RESEARCH AND DIAGNOSIS

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FOREWORD

AT LAST here is a book which gives all necessary information in a detailed, simple, scientific and accurate way on the diagnostic use of isotopes in medicine.

Written by scientists expert in this field for many years, this book gives a comprehensive view of all that is necessary for a clinician who wants to use isotopes. The authors complement each other in their knowledge magnificently. One, who has developed quite a number of clinical isotope techniques himself has undoubtedly become one of the few experts in this field; the other, who has made a wide application of radioisotopes in a hospital, has the clinician's approach. The result is therefore an extremely useful book, which to my mind should be in every hospital's library and also in the possession of a great number of clinicians.

That there was a need for such a book in the past, is an understatement, and I am happy to say that the authors have succeeded excellently in presenting the essential facts concerning radioisotope uses which the clinical worker needs.

March, 1958 HENRY SELIGMAN

圖書室

GENERAL INTRODUCTION

> "*Claude Bernard me disait un jour 'Nous saurons la physiologie lorsque nous pourrons suivre pas à pas une molécule de carbone ou d'azote, faire son histoire, raconter son voyage dans le corps d'un chien, depuis son entrée jusqu'a sa sortie'*".
>
> (Taine, H. A. (1891). "Histoire de la France", Vol. 7, p. 28.)

In the same year as the remark quoted above was published, Manya Sklodovska enrolled as a student at the Sorbonne. As Marie Curie she was later to be awarded the Nobel Prize for her work on natural radioactivity; and subsequently her daughter Irene together with Frederic Joliot-Curie were to share a similar award for their discovery of artificial radioactive isotopes. A further Nobel Prize was awarded in 1943 to Georg von Hevesy for his pioneer work which clearly established the value of these new materials as tracers in biological research.

Until 1946 these substances were available to only a few workers, but in recent years radioactive isotopes have become generally available as a result of the large-scale expansion of work on nuclear energy. They are finding increasing application in modern industrial practice, but since it was in the fields of biology and medicine that their value was first realized, it is in these fields that the greatest progress has been made to date. It is now widely accepted that radioisotope techniques provide a unique and powerful research tool for the medical scientist, an ever-increasing range of potentially valuable diagnostic procedures for clinicians in general, and a number of useful technical improvements in the treatment of patients for the radiotherapist. Not unnaturally, a large proportion of the early effort has been devoted to exploring the possible therapeutic applications of radioactive isotopes, and several text-books have already appeared on this subject. A good deal is now known about the possibilities, and rather more about the limitations, of radioisotope therapy; but the formidable body of literature now extant on the use of isotopes in research and diagnosis serves only to emphasize the fact that this field is, by comparison, still largely unexplored.

So far as the basic medical sciences are concerned the value of isotope techniques is well established, and they are now taken for granted in any biochemical or physiological research laboratory. In effect, they have taken their place alongside other laboratory methods such as spectrophotometry and chromatography as yet

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another research tool. As with any other technique, isotopes are particularly applicable to certain kinds of problem, and in other kinds they have definite limitations. Like any other method of measurement, isotope techniques can play a vital part in a scientific investigation; and they can equally well be used to accumulate masses of meaningless data. In fact, the use of isotopes cannot eliminate the necessity for a properly planned experimental approach to a problem, but it can often make the design and execution of suitable experiments a very much simpler task.

Parallel with their use in purely scientific research, isotope methods have been employed increasingly widely, though as yet by no means generally, in a wide variety of clinical research studies. They have been applied to the study of physiological processes in health and disease; and they have been used for assessing objectively the results of various forms of treatment as well as for elucidating day-to-day practical problems arising in the treatment of patients. The situation now is that there is not one single branch of medicine or surgery where isotopes have not found some application.

As a consequence of the use of radioisotopes in clinical research, various potentially valuable diagnostic procedures have been developed. The value of some of these tests is now widely recognized, and they are in routine clinical use at least in those hospitals which have the necessary facilities. In general, however, the practical value and the limitations of most of these diagnostic procedures are yet to be established. However valuable a diagnostic test may appear to be when it is tested on an experimental basis, the fact remains that its ultimate acceptance must depend on extensive practical trials by the clinicians concerned, and it is they who will finally decide, on the basis of their experience, whether or not a given procedure is of any value in the diagnosis and treatment of disease. A prerequisite to the acquisition of the necessary clinical experience with these new methods is that they should be more generally available than is the case at present. The technical and financial difficulties attending the introduction of such methods are often overestimated; and it is hoped that the present volume will at least enable the potential user of radioisotopes in clinical medicine to see these difficulties in their proper perspective.

In preparing this book, we have had in mind the needs of the clinical worker who is primarily concerned with the management of patients, and who is interested in the possibility of using radioisotope methods either on a routine basis or for research work designed to achieve a better understanding of the clinical conditions which he sees. It is recognized that the majority of workers in this category do not have unlimited funds for equipment, an army of highly trained technicians, palatial laboratories and a great deal of spare time. Consequently, the first part of the book is mainly concerned with the fundamental principles of radioisotope methods and instrumentation, presented in such a way that a given individual may design techniques and choose equipment which best meet his own requirements, having regard to his own particular circumstances. At the same time, we have taken the view that it is no more necessary for a clinician to take a degree course in nuclear physics or electronic engineering before doing a thyroid test with radioiodine than it is for him to qualify as an optician before examining a tissue section with the aid of a microscope.

A simplified approach to the problem of assessing radiation dosage and the consequent risk to the patient has been adopted in view of the numerous factors other than purely physical ones which may influence the final conclusion. It is also shown that the problems of radiation protection so far as occupationally exposed workers are concerned can be overcome by fairly simple methods and the exercise of a certain amount of common sense.

It was not considered sufficient to simply describe a series of methods and techniques for obtaining data without at the same time discussing their interpretation, with particular reference to the possible sources of error which may be met with in practice. Consequently, most of the second part of the book is devoted to various clinical applications. In order to keep the emphasis on basic principles, we have preferred to discuss a limited number of representative problems in some detail rather than to present a superficial survey of the extensive and rapidly growing body of literature on the subject in general. The topics chosen for discussion are those which appear likely to be, or already are, of practical clinical importance; and hence are necessarily those which have already been extensively studied by several independent groups of workers. A further restriction has been imposed by the fact that we have not felt it possible to attempt to evaluate the often conflicting reports on any one subject unless one or other of us has had some practical experience of the work in question.

A number of the simpler methods have been described in some detail, either because they serve as useful examples, or because they are already widely established. In some cases the published techniques have been found impracticable in the absence of the resources of a major research institute, and it has been necessary to develop simpler procedures for everyday use. This accounts for the inclusion of a number of hitherto unpublished methods. Nevertheless, it is expected that any intending user will consult the appropriate original references; and the bibliography at the end of each chapter, though by no means complete, should serve as a useful starting-point.

The brief chapter on isotope therapy has been written on the assumption that this is a specialized field of medical practice, which in any case is adequately covered elsewhere; but at the same time it is of some interest for the non-specialist on the subject to have some idea of the possibilities and limitations of this form of treatment. Moreover, the decision to undertake radioisotope therapy and the planning of the treatment will often be based on the results of tracer diagnostic tests.

Inasmuch as the progress of this particular "Peaceful Application of Atomic Energy" depends on continued and closer collaboration between clinicians and workers in the physical sciences, it is hoped that this book will also be of some value to those hospital physicists who wish to gain a broader view of the problems on which they may at present be engaged in a somewhat specialized capacity.

Although the present work epitomizes the practical experience gained by the writers over the past decade, it must be emphasized that this experience has been acquired in the course of collaborative research and discussions with many other clinical workers, to whom we are overwhelmingly indebted. Lack of space precludes individual acknowledgement, but, more important, it would be unfair to imply that the many expressions of opinion in this book which may well be confounded by future work are the responsibility of anyone but the authors.

We are also indebted to numerous colleagues who have critically reviewed the manuscripts of some of the individual chapters; and in particular to Dr. R. Höfer and Mr. J. D. Pearson who have, in addition, kindly permitted us to use the results of hitherto unpublished work in which they have participated. We are also grateful to Professor K. Fellinger, Director of the 2nd Medical University Clinic, Vienna, who has given much encouragement and help.

Expert advice and assistance in connexion with the preparation of a number of the illustrations have been provided by Mr. C. E. Engel and Miss S. Treadgold of the Departments of Photography and Medical Illustration of Guy's Hospital; and, finally, we wish to acknowledge the skill and the patience of Miss Noreen Ellis and Miss Brigitte Pichler who have borne the burden of the typing.

N. VEALL
H. VETTER

London and Vienna, 1958

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CHAPTER 1

RADIOACTIVE ISOTOPES

NATURALLY OCCURRING ISOTOPES

ATOMS are generally believed to consist of a central nucleus, surrounded by a number of electrons. The chemical identity of an atom is determined by the number of orbital electrons which it possesses, since ordinary chemical reactions between different elements only involve interactions between their outermost electron shells. Electrons carry a single unit of negative electrical charge and have only small mass, about $\frac{1}{2000}$ of that of a hydrogen atom. The greater part of the mass of an atom is thus associated with the nucleus, which can be assumed to be made up of two types of particle, namely protons and neutrons.

The proton has a mass of approximately one unit on the scale of mass used in atomic and nuclear physics, and carries a positive electrical charge equal to that of the electron. The atom is normally electrically neutral, the number of protons in the nucleus and the number of orbital electrons being equal. This number is the so-called atomic number which characterizes each individual element, and for the naturally occurring elements ranges from 1 in the case of hydrogen to 92 for uranium.

The neutron has a mass similar to that of the proton, but carries no electric charge. Consequently, the presence of an additional neutron in an atomic nucleus will not alter the chemical identity of that atom, but it will result in an atomic weight which is approximately 1 mass unit greater. Most naturally occurring elements exist in a mixture of two or more forms, variations in the number of neutrons in their nuclei being reflected by differences in their atomic weight. The individual components which go to make up the mixture are known as isotopes of the element concerned. Table I shows how the hydrogen, carbon, nitrogen and oxygen occurring in nature all consist of a mixture of isotopes.

By using rather complicated physical methods it is possible to modify the ratios in which these isotopes are normally found (normal abundance ratios), and individual isotopes may be obtained as enriched preparations or even in the almost pure separated form. The "heavy hydrogen" isotope (deuterium), the isotope of carbon with mass 13 (written ^{13}C) and the isotopes ^{15}N and ^{18}O have found important applications in biochemical tracer work. The latter isotopes are particularly useful since there are at present no suitable radioactive isotopes of oxygen and nitrogen available for this purpose.

The usefulness of stable isotopes as tracers is limited by the inability to carry out measurements on samples where the administered isotope has been highly diluted by the naturally occurring mixture, and in any case comparatively expensive equipment and specialized techniques are necessary for their measurement. Stable isotope techniques are beyond the scope of the present volume; some recent text-books dealing with this subject are given in the bibliography.

TABLE I

STABLE ISOTOPES OF HYDROGEN, CARBON, NITROGEN AND OXYGEN

Isotope	*Number of protons in nucleus*	*Number of neutrons in nucleus*	*Atomic weight A.*	*Natural abundance per cent*
^{1}H	1	0	1	99·984
^{2}H	1	1	2	0·016
^{12}C	6	6	12	98·9
^{13}C	6	7	13	1·1
^{14}N	7	7	14	99·62
^{15}N	7	8	15	0·38
^{16}O	8	8	16	99·76
^{17}O	8	9	17	0·04
^{18}O	8	10	18	0·20

In some publications the word "nuclide" is used as a general term and the term "isotope" used in a more restricted sense, for example, "^{15}N and ^{18}O are nuclides, they are isotopes of nitrogen and oxygen respectively".

RADIOACTIVE ISOTOPES

Although many elements have a number of naturally occurring isotopes, the number of stable arrangements of neutrons and protons is rather limited and any other combination of these particles results in an unstable nucleus. Such a nucleus will tend to assume a more stable configuration, gaining or losing a unit of electrical charge and

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disposing of excess energy by the emission of radiation. A nucleus which behaves in this way is described as being radioactive.

A few naturally occurring isotopes, notably of the heaviest elements, but including also the isotope of potassium ^{40}K, are weakly radioactive. Any less stable isotopes have long ago disintegrated completely. The strongly radioactive substances such as radium which occur naturally, only do so because they are continually being formed as products of the radioactive decay of the weakly radioactive elements uranium and thorium.

RADIOACTIVE DECAY

The greater the degree of instability of an atomic nucleus, the greater the probability that during any given interval of time it will disintegrate. Beyond this statement it is not possible to make any prediction about the exact moment of disintegration of a particular atom. In practice, however, one is dealing at any one time with samples of material containing large numbers of atoms, and though the individual disintegrations occur at random, the statistical behaviour of such a large population can be described mathematically with a high degree of accuracy.

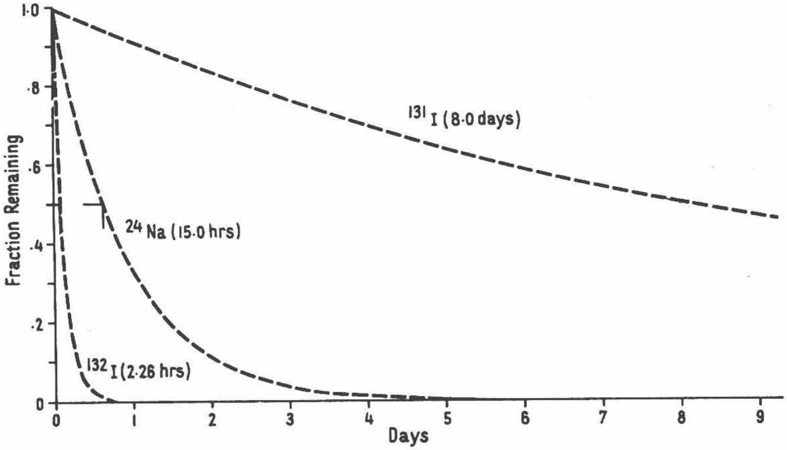

FIG. 1 —Decay curves for ^{132}I, ^{24}Na and ^{131}I (Linear scale.)

The fundamental law of radioactive decay states that the number of disintegrations occurring in one second in a sample of radioactive material at any time is directly proportional to the number of radioactive atoms actually present in the sample at that time. This is the assumption made when two samples of the same radioactive isotope are compared by measuring the radiations which they emit.

The number of radioactive atoms (N_t) remaining at time t is given by the exponential expression

$$N_t = N_0 e^{-\lambda t} \tag{1}$$

where λ is the radioactive decay constant, which is a characteristic of the particular isotope concerned. It represents the fraction of the atoms disintegrating in unit time. Fig. 1 shows the decay curves for two radioactive isotopes of iodine ^{132}I and ^{131}I, and for the isotope of sodium ^{24}Na, whose decay constants are 0·307, 0·0036 and 0·046 hrs^{-1}, respectively. Other examples of exponential processes are discussed on page 176.

RADIOACTIVE HALF LIFE

Examination of Fig. 1 will show that in the case of radioactive sodium, ^{24}Na, the activity remaining has decreased to half its initial value after 15 hours. After another 15 hours there is further decrease by a factor 2 to one-quarter of the original activity. Instead of using the decay constant λ, it is possible, and much more convenient, to define the rate of decay of ^{24}Na in terms of a half life of 15·0 hours. The half lives of ^{132}I and ^{131}I are 2·26 hours and 8·0 days respectively.

Mathematically, the half life ($T_{\frac{1}{2}}$) is related to the decay constant λ by the expression

$$\lambda = \frac{0 \cdot 693}{T} \tag{2}$$

so that equation (1) can be written

$$N_t = N_0 e^{-\frac{0 \cdot 693}{T_{\frac{1}{2}}} t} \tag{3}$$

In practice, the half life is usually used in preference to the decay constant, since apart from being more informative, it is easily memorized.

Equation (1) can be written

$$\log_e N_t = \log_e N_0 + \lambda_t \tag{4}$$

Consequently, if the logarithm of the observed disintegration rate is plotted against time, or if N_t is plotted on logarithmic graph paper, a straight line is obtained whose slope depends on the value of the constant λ. The decay curves in Fig. 1 are shown plotted on a logarithmic scale on page 116.

THE UNIT OF RADIOACTIVITY (THE CURIE)

The amount of radioactivity present in a sample of material at any given time is specified in terms of a unit named after the pioneer workers Marie and Pierre Curie. The curie is defined as that amount of radioactive material in which the number of disintegrations per second is $3 \cdot 700 \times 10^{10}$. The corresponding figures for the millicurie and the microcurie are thus $3 \cdot 7 \times 10^7$ and $3 \cdot 7 \times 10^4$, respectively. This unit was originally based on the number of disintegrations occurring per second in 1 g. of radium, but has recently been redefined in its present terms so that it may be equally applicable to other isotopes.

The number of radioactive atoms present in a 1-mc. sample is equal to $3 \cdot 7 \times 10^7$ divided by the decay constant λ, which, in the case of ^{131}I for example, is about 10^{-6} sec^{-1}. 1 mc. of ^{131}I thus contains $3 \cdot 7 \times 10^{13}$ ^{131}I atoms. One gramme molecular weight of any substance corresponds to $6 \cdot 02 \times 10^{23}$ molecules (Avagadro's Number). Consequently, 1 mc. of ^{131}I is equivalent to

$$\frac{131 \times 3 \cdot 7 \times 10^{13}}{6 \cdot 02 \times 10^{23}} = 0 \cdot 008 \ \mu\text{g. of iodine}$$

Most isotopes used clinically have half lives measured in hours or days, and if it is appreciated that it is possible to detect quantities of radioactivity of the order of one-millionth part of a millicurie it is apparent that it is the amount of the stable isotope present and acting as a diluent which is the practical factor limiting the actual amount of the element which can be detected.

SPECIFIC ACTIVITY

This is a term which, in the absence of a recognized definition, is usually used rather loosely. It is sometimes defined as the ratio of the number of active atoms to the number of corresponding inactive atoms present in a sample, but in actual practice it is used with various meanings, involving the use of units which need to be specified. Thus in the case of individual elements, the specific activity is usually given as millicuries per milligram of element, rarely if ever as millicuries per milliequivalent. On the other hand, the activity of organic compounds, particularly those labelled with ^{14}C, is usually quoted in millicuries per millimol. The term "specific activity" has sometimes been used to report the number of millicuries per millilitre of a solution, but the term "concentration" seems more appropriate for this purpose. In subsequent sections covering various clinical applications the expression is used whenever it is necessary to deal with the amount of radioactive material associated with a given amount of the corresponding inactive material, the precise sense in which it is used depending on the context.

The specific activity available is one of the factors which will govern the choice of an isotope for any particular purpose. It is necessary to have a sufficient amount of the radioisotope present to permit its detection and measurement; but if in order to attain this, the material has to be given in excessive amounts, the results of the investigation may not reflect normal physiological processes.

RADIATIONS FROM RADIOACTIVE ISOTOPES

A radioactive nucleus tends to adopt a more stable configuration by either gaining or losing a unit of positive electrical charge, so that, in effect, one of the neutrons is transformed into a proton or vice versa. In the majority of cases this process is characterized by the emission of a charged particle having a mass equal to that of the electron; this so-called β-particle or β-radiation may be either negatively or positively charged. In the latter case the particle is known as a positron.

β^--*Radiation*

Isotopes having an excess of neutrons usually decay by the emission of a negative β-particle, which is in fact a fast-moving electron. The β-particles from a particular isotope have varying energies, up to a maximum ($E_{\max}$) which is a characteristic of that isotope. The energy of these and other nuclear radiations are usually measured in million electron volts (MeV), one electron volt being the energy acquired by a particle having a charge equal to that of the electron when it falls through a potential gradient of one volt ($1 \text{ eV} = 3{\cdot}8 \times 10^{-20}$ g. calories $= 1{\cdot}6 \times 10^{-12}$ ergs). As an indication of the orders of magnitude involved it may be noted here that most chemical reactions involve energies in the range 1–10 eV.

Radioactive phosphorus ^{32}P, sulphur ^{35}S, carbon ^{14}C and hydrogen ^{3}H are typical β^--emitters. In each case the loss of a unit of negative charge means that the nucleus has effectively gained a proton in place of a neutron, and the resulting nucleus is that of the next highest element in the periodic system.

β-rays are comparatively easily absorbed in matter, their penetrating power depending mainly on their energy and on the density of the absorber. The β-rays of ^{32}P, whose maximum energy is about 1·7 MeV, have a maximum range in soft tissue of about 7 mm. If the number of β-rays passing through an absorber is plotted against the absorber thickness, which is usually expressed in mg./cm^2, the resulting curve is approximately logarithmic. The half absorption thickness for ^{32}P is then found to be about 100 mg./cm^2, corresponding to about 1 mm. of tissue.

The limited range of penetration of β-rays greatly simplifies the problem of protection from external radiation hazards, but it also results in some practical difficulties when these radiations have to be detected and measured. Thus the low energy, or "soft" β-rays from tritium are completely stopped by an absorber weighing only 1·0 mg./cm^2.

γ-*Radiation*

In many cases the available energy in the radioactive nucleus is not accounted for completely by the process of β decay, and the excess energy is then emitted as a quantum of γ-radiation. This is electromagnetic radiation of short wavelength, and is therefore similar to the radiation produced by high-voltage x-ray apparatus. The effective mass of a γ-ray quantum is directly proportional to its energy. The relationship between mass and energy is given by Einstein's famous equation

$$\text{Energy (ergs)} = \text{mass (grammes)} \times c^2$$

where c is the velocity of light (3×10^{10} cm./sec.). Under some circumstances a γ-quantum can be transformed into an electron and a positron provided that the mass equivalent of its energy is greater than the combined mass of the two particles. This implies an energy of slightly more than 1 MeV.

γ-radiation is highly penetrating. The absorption of γ-ray quanta by matter depends on their quantum energy and on the nature of the absorber. The absorption curve in a given medium is an exponential one, but instead of dealing with absorbers weighing only a fraction of a gramme per square centimetre, it is necessary to think in terms of half absorption thicknesses expressed as centimetres of lead.

Many isotopes, including ^{131}I, ^{132}I, ^{24}Na and ^{59}Fe decay with the emission of a β -particle together with one or more γ-rays. In the case of isotopes such as ^{86}Rb and ^{42}K only a fraction of the β-particles are associated with a γ-ray. Thus ^{42}K emits β-particles with E_{max} 3·6 MeV, but about 15 per cent of the disintegrations result in the emission of lower energy β-particles (E_{max} 2·1 MeV) together with 1·5 MeV γ-rays. The fraction of the disintegrations occurring in this way is known as the branching ratio.

β^+-*Radiation* (*positron*)

Many neutron-deficient isotopes decay by the emission of a positron, thereby reducing the atomic number of the atom by one unit. The long-lived sodium isotope ^{22}Na decays in this manner, as do ^{58}Co and ^{74}As. All positron emitters can be regarded as sources of γ-radiation, since when a positron ultimately comes to rest after losing its kinetic energy in its passage through an absorbing medium, it immediately combines with an electron. The two particles are then transformed into annihilation radiation; this usually consists of two γ-ray quanta, each with an energy of $\sim 0{\cdot}5$ MeV, which are emitted in opposite directions. This is the reverse process to that of pair formation from a γ-quantum which is described above.

K-Capture

As an alternative to positron emission a neutron-deficient nucleus may effectively lose a positive charge by capturing an orbital electron from the innermost or *K* shell. The filling of the resulting gap by an electron from outside the atom results in the emission of the characteristic x-rays of the product atom. Both ^{51}Cr and ^{55}Fe decay in this manner, in the case of chromium 8 per cent of the nuclear transformations are accompanied by a γ-ray of energy $0{\cdot}32$ MeV. The majority of the positron-emitting isotopes may also decay by *K*-capture, the branching ratio varying with different isotopes.

α-Particles

Many of the heavier elements, notably radium, decay with the emission of α-particles. These are identical with helium nuclei in that they consist of a combination of two protons and two neutrons; the emission of an α-particle therefore results in a loss of four mass units and two units of positive electrical charge. α-Emitters are of little practical importance in clinical tracer work and need not be considered further here.

PRODUCTION OF RADIOACTIVE ISOTOPES

Nuclear reactors

Introduction.—Radioactive isotopes became generally available some years ago as a result of the development and operation of controlled nuclear reactors. In the nuclear reactor, or "pile", fuel rods of so-called fissile material, for example the isotope of uranium ^{235}U, are irradiated with neutrons. A ^{235}U nucleus on capturing a neutron becomes so unstable as to disintegrate forthwith giving two "fission products" of approximately equal size, together with two or three more neutrons which may be captured by other ^{235}U nuclei so that the process continues and a chain reaction is set up. At the same time a considerable amount of energy is produced. Any neutrons over and above those used up in maintaining the chain reaction are available for other purposes, such as the production of radioisotopes by neutron irradiation.

Fission products.—The fragments resulting from the fission process occurring in a reactor consist of the nuclei of elements occupying

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