PEACEFUL USES OF ATOMIC ENERGY

Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy

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Volume 20
Isotopes in Research

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PREFACE

More than 2100 papers were submitted by the nations, the specialized agencies, and the International Atomic Energy Agency, which participated in the Second United Nations International Conference on the Peaceful Uses of Atomic Energy. The number of papers was thus about twice that involved in the First Conference. Provision was therefore made to hold five concurrent technical sessions in comparison with the three that were held in 1955. Even so, the percentage of orally presented papers was less in 1958 than in 1955.

In arranging the programme, the Conference Secretariat aimed at achieving a balance, allowing adequate time for presentation of as many papers as possible and, nevertheless, leaving time for discussion of the data presented. Three afternoons were left free of programme activities so that informal meetings and discussions among smaller groups could be arranged. No records of these informal meetings were made.

A scientific editorial team assembled by the United Nations checked and edited all of the material included in these volumes. This team consisted of: Mr. John H. Martens, Miss L. Ourom, Dr. Walter M. Barss, Dr. Lewis G. Bassett, Mr. K. R. E. Smith, Martha Gerrard, Mr. F. Hudswell, Betty Guttman, Dr. John H. Pomeroy, Mr. W. B. Woollen, Dr. K. S. Singwi, Mr. T. E. F. Carr, Dr. A. C. Kolb, Dr. A. H. S. Matterson, Mr. S. Peter Welgos, Dr. I. D. Rojanski, Dr. David Finkelstein, Dr. Cavid Erginsoy (Dr. Erginsoy's services were furnished through the courtesy of the International Atomic Energy Agency), Dr. Vera J. Peterson, Dr. Paul S. Henshaw, Dr. Hywell G. Jones, Dr. Alvin Glassner and Mr. J. W. Greenwood.

The speedy publication of such a vast bulk of literature obviously presents considerable problems. The efforts of the editors have therefore been primarily directed towards scientific accuracy. Editing for style has of necessity been kept to a minimum, and this should be noted particularly in connection with the English translations of certain papers from French, Russian and Spanish.

The Governments of the Union of Soviet Socialist Republics and of Czechoslovakia provided English translations of the papers submitted by them. Similarly, the Government of Canada provided French-language versions of the Canadian papers selected for the French edition. Such assistance from Governments has helped greatly to speed publication.

The task of printing this very large collection of scientific information has been shared by printers in Canada, France, Switzerland, the United Kingdom and the United States of America.

The complete Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy are published in a 33-volume English-language edition as follows:

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Production and Use of Radioisotopes in Research

By Henry Seligman*

When discussing the progress of the production of isotopes and their use in research one must realise that an international conference on this very subject arranged by UNESCO took place just one year ago and, therefore, it is not surprising that during this relatively short interval not much new research work with radioisotopes has been developed which will be reported in this session. However, a number of interesting papers have been presented, and one can try no more than to highlight the recent developments or the trend of future developments in each of the various fields comprised in the session.

Almost half the papers presented deal with the production of radioactive material in some form or other either in reactors and linear accelerators or by the enrichment of specific activity by chemical means and, quite a number of papers, with organic synthesis of radioactive compounds. The rest of the papers are divided for convenience arbitrarily under the following headings: diffusion, reaction mechanism, adsorption phenomena, development of applications, special general subjects and stable isotopes.

PRODUCTION AND ENRICHMENT

The production of radioactive isotopes in reactors has followed the expected line. There were three obvious problems: first to use higher neutron fluxes in order to get higher specific activities, second to make big megacurie sources, and third to make new labelled compounds. Therefore all the papers presented in this field by the major isotope-producing countries are similar in the sense that they are mainly dealing with the technology of achieving these aims, except perhaps one paper which discusses in great detail the theoretical aspect of the influence of the disturbance of the neutron flux by inserting neutron absorbing material.

Hand in hand with this development goes, of course, the handling of the big irradiation sources, the details of which are given in a number of interesting and useful papers. Also, more attention is being given to the design of big cobalt-60 sources, and whilst only one or two papers deal specifically with this subject there has been work going on in many countries for quite some time to find the most economic and reasonable physical form of cobalt which would be applicable to the many fields in which radiation is playing its part in research and development.

One paper touches the question of costing isotopes produced in testing reactors, but it is difficult at this time to put a price tag on any produced radioisotope, which is acceptable to everybody, since there is no agreement on which part of the cost of a reactor should be recovered. The price of isotopes, however, is important, especially if we remember that big isotope radiation sources may have to play their part in the many massive radiation applications described in another session. During the past year, a number of conflicting publications have appeared on this subject, and predictions by enthusiastic reactor men and optimistic scientists producing the machines are wide apart. This is especially so for predictions on the future, when the suggested production cost of cobalt would, in many cases, have to be competitive with machines. However, the progress in the construction of linear accelerators is considerable, and one paper gives some details on this, namely how the efficiency can be increased and how technical progress has been made by improving the technology for the irradiation of liquids. The main problem of the inferior penetration by the radiation from such machines when compared with cobalt-60 has been partially overcome by the inversion of the beam to give double bombardment. According to this paper the efficiency is increased by 2.4.

Complementing the production of radioactive materials in reactors is the production in cyclotrons and accelerating machines. Considerable literature exists on the production of isotopes in cyclotrons but not much on production in linear accelerators. A number of firms are now producing such machines which are especially suited to making neutron-deficient isotopes. The neutron flux in machines with energies between 2 and 20 Mev produced by bombardment of beryllium varies between 10^4 and 10^8 neutrons/cm^2/sec. However, one must not forget, especially if one deals with long-lived material like sodium-22 for instance, that one has to take into account the competitive production in a cyclotron where this may be done by a stray beam.

The production of high specific activities may be achieved either by increased neutron fluxes or by chemical separation methods. A number of papers

* Deputy Director General, Department of Research and Isotopes, International Atomic Energy Agency.

report on the theoretical aspects of the production of high specific activity isotopes and discuss the flux depressions which occur when different neutron-absorbing materials of varying shapes are inserted in different reactors.

As more and more powerful reactors are available it is obvious that during the last years much more attention has been given to the recovery of big amounts of separated fission products. All major countries have done some work in this field and, in fact, some of this work is presented in detail in this session. There is no doubt that the trend in this field is towards bigger and bigger production units in order to make fission products cheaper, and within the next year or so the long-lived caesium-137 should be priced in the same order of magnitude as its competitor cobalt-60, taking into account the different K-factors.

Quite a number of refinements of fission product pilot plants have been reported since the last Geneva conference and even since last year's Paris meeting. There is also no doubt that next to caesium-137 and strontium-90, krypton-85 will play an ever increasing part in the years to come.

A number of operational details are given for the production of colloidal solutions and inorganic compounds. Further research has been made on the production of a suspension of chromic phosphate containing P^{32} and another report gives details of activated alumina as an adsorbent for carrier-free isotopes.

For a long time research has been going on to produce carrier-free radioactive material by the Szilard-Chalmers process and, in fact, one or two production methods are making use of such a reaction. It is obviously of interest to find new applications for this gamma recoil method and some work has been published on this, which, however, shows only too well what has been known for some time, that the field looks more and more disappointing as research progresses. One paper deals with the theoretical studies on the influence of exchange reactions on the yield of the recoil reaction, and in another one the chelates of cobalt have been studied. It has been found that the enrichment factor is very much influenced amongst other parameters by the neutron irradiation, the temperature, and the time which elapses between irradiation and chemical separation. Both papers point out the difficulties encountered in trying to apply the Szilard-Chalmers method and, while it may of course be possible that ideal reactions may be found which lend themselves to radioisotope production, the difficulties should not be underestimated.

LABELLED ORGANIC COMPOUNDS

The important production of organic compounds labelled with radioisotopes can be achieved in three different general ways: The compounds can be produced by orthodox chemical synthesis, by biosynthesis using labelled gases and by chemical exchange reactions where the energy comes either through nuclear recoil or from an outside source.

A number of papers report on the method of orthodox synthesis of organic compounds; one gives details on some carbon-14 and sulphur-35 labelling. Other papers deal with the formation of phenyl ketone, and some illustrate methods of production of labelled amino acids and other biologically important compounds. A number of these compounds, however, have been commercially available for some time.

It is not surprising that special interest is devoted to the production of labelled compounds by the method of chemical exchange under the influence of radiation. This method which has been developed during the last few years has prospects of being used for the labelling of complicated compounds which are difficult or costly to make by chemical or biosynthetic methods. But a great deal of study is necessary in order to find conditions suitable to achieving certain reactions not producing a great number of mixed labelled compounds, the usual end result with this method. The three main recoil reactions are the $Li^6(n,\alpha)H^3$, the $He^3(n,p)H^3$ and the $N^{14}(n,p)C^{14}$, the recoil energy varying from many thousands to 40 kev. Only a small fraction, usually between 0.1 and 10%, of the recoil atoms substitute an inactive carbon atom or form an addition to the molecule, as is pointed out in one interesting survey paper.

The labelling may also be induced by any kind of radiation like that coming from either an accelerating machine or a radioactive isotope. The radiation usually initiates an ionisation of the organic compound which is a prerequisite for an exchange reaction. One paper reports on a method in which radioactive isotopes in a gaseous state are added in order to produce C^{14}-labelled organic compounds.

Whilst this method does not perhaps give the specific activities which have been achieved by nuclear recoil labelling with tritium it nevertheless must be regarded as one of the methods which will be very useful indeed in the future where relatively small amounts of complicated organic compounds have to be produced.

The main problem which occurs through this type of labelling is the separation of the many differently labelled species of organic compounds produced simultaneously from the original molecule. Therefore, in most cases the main effort is really on the purification process and is not so much a production problem. However, if a simple purification system can be found, this method of labelling will be by far the most convenient for complicated organic compounds.

Another paper is concerned with the recoil labelling of carbon-14 in benzene systems, and the studies reveal that the product yield and composition were influenced by the concentration and the type of nitrogen used for the recoil reaction. From these studies conclusions are drawn on the process which might be an intermediate in this kind of labelling

which in this case may be a carbene. Without going into further details of any such processes it should be pointed out that, whilst we do not yet understand the process of C^{14} labelling by this method, there is hope that the next few years will enlighten us considerably in this matter through research of the type just mentioned.

One paper gives the details of the use in biology of tritium-labelled compounds produced by self-irradiation. The studies were on the subcellular localisation and protein binding of radioactive digitoxine. The authors proved that tritium-labelled digitoxine made by self-irradiation behaved exactly in the same way as the biosynthetically labelled C^{14} digitoxine in these biochemical tracer studies.

The whole field of nuclear recoil and radiation labelling is still in an early stage. Within the next few years we expect to have much more knowledge of this method of synthesis, and we may be able to produce an array of new organic compounds although with smaller specific activities than are normally obtained through straight chemical synthesis.

DIFFUSION AND SELF-DIFFUSION

For many years isotopes have been by far the best means for the study of diffusion and self-diffusion. Recently such research has received impetus from the general tendency to use high temperature reactions in all fields of science, including nuclear technology. Not only are measurements of atomic mobility useful in order to find out something about the state of the crystal lattices at high temperatures, but also such measurements are necessary to assess the energy required to create lattice defects.

Usually there are many experimental difficulties in measuring self-diffusion in solids, but during the last few years experimentalists have mastered most of these problems, and a number of papers are available which deal with such work. Some of these papers cover self-diffusion studies in oxide systems, which are of the greatest importance for high temperature techniques and the elucidation of solid state reactions including powder reactions. Other papers deal with diffusion phenomena like recrystallisation, grain growth, creep and many other problems. One paper stresses the importance of pressure as a variable in studies of self-diffusion in liquids and crystallised solids and points out the very big influence which the application of pressure has on the self-diffusion rate in certain solids and liquids. This effect has been measured in lead and the results have been expressed in terms of one variable only, namely temperature.

Uranium self-diffusion was studied by using U^{233} as a tracer and by measuring the intensity of α radiation. In this way the uranium self-diffusion coefficient could be determined. Also, autoradiography was used to determine the value for the heat of activation of specimens which had been subjected to diffusion annealing. Valuable work has been done so far in this field; it seems to be concentrated, however, in certain countries.

EXCHANGE REACTIONS AND REACTION MECHANISMS

Isotopes are the ideal tool with which to study exchange reactions and reaction mechanisms. There are virtually hundreds of papers which have been published in the past on this subject. There are a dozen or so papers which deal with this subject at this Conference. An extension of earlier work in a new and important field is the paper on the temperature-independent isotope effect in chemical exchange equilibria involving linear molecules. This rather involved subject is presented in a paper dealing with a general method of treating exchange reactions involving linear molecules. The knowledge of the temperature-independent factor enables one to calculate the temperature dependence of the equilibrium constant of an isotopic exchange reaction from measurement at a single temperature.

Other papers deal with the use of isotopes in the study of organic reaction mechanisms. One paper is concerned with the mechanism of certain free radical reactions, especially those which are important in the polymerisation of monomers. Also a double labelling method with C^{14} and tritium has been used to study transfer problems, and it has been found that there is evidence for a large isotope effect in the abstraction of hydrogen atoms from molecules such as triphenylmethane.

The field of cationic exchange between metals and electrolytic solutions, the use of isotopes and radiation in the study of heterogeneous catalysis, and similar studies in other papers show the increased interest which chemists have in the use of isotopes to solve specific exchange problems.

Fluorine-18 (half-life 2 hours) is being used more and more to discover details of exchange reactions in fluorides. A number of exchange reactions and catalytic oxidation studies have been made with this isotope.

ADSORPTION

Radioisotopes have always played a great part in the investigation of adsorption phenomena. Some of the papers on this subject are concerned with more practical aspects, like the adsorption of certain ions on glass; in this case special studies have been made on the adsorption properties of glass powder.

A similar research on the fixing of ions is the one on the measurements of various amounts of ions in solution which may become attached to the surface of apparatus. As the authors point out, three types of deposition can be distinguished: exchange of cations in solution with the same material, the deposition of more noble materials from solution onto less noble surfaces, and the deposition of less noble materials

from solution onto more noble surfaces. It is especially this last one which has not received much attention in the past and which was investigated in more detail. It is obvious that such an investigation must deal with many variables like purity of materials, the condition of the surface, concentration of cations, the presence of other substances in solution and many others. The radioactive cations studied were those which are of some technological interest. In this case dilute solutions of strontium-90, cerium-144 and cobalt-60 and their deposition on aluminium, lead, copper, zinc and stainless steel were measured. The results of this work show that factors other than potentials of the metals play an important role. This has been shown very well by the tendency of strontium-90 and cerium-144 toward deposition on metals which are more noble. Quite a number of other important fundamental conclusions were reached by the authors.

RESEARCH ON APPLICATIONS

The research with radioactive materials for eventual applications is of ever-increasing interest. Only the pure research activities in this field are reported here as the applications themselves fall into the next session. An important problem on which considerable work has been done already is the study of isotopes in hydrology. One paper deals with studies of groundwater flow. This, of course, is a specially difficult problem to cope with since so much depends on local circumstances because different ions behave differently, depending on the geology. Although tritium will always be one of the most important isotopes in this connection, this problem is still not quite solved and in fact may often require some special investigation according to the local circumstances. The same authors made also some very interesting theoretical approaches to the problem of water tracing.

An interesting research application is the detection of fingerprints by autoradiography. Although this has been published some time ago it still remains an important contribution. The main advantage of this new method is that it can detect fingerprints even on relatively rough surfaces, like paper or wool. The authors employed successfully the chemical reaction between formaldehyde labelled with carbon-14 and either the protein in the fingerprint or the adsorption of radioactive stearic acid on the fingerprint.

An important aspect of the use of isotopes in research is the fact that alpha-emitting materials are used more and more. The reason that these materials have not been used much before is because the only convenient alpha emitter is polonium-210 which is a rather toxic material. However, with proper safeguards and handling techniques this isotope can be used more frequently now. One very interesting paper deals with the use of polonium for the measurement of interfacial area. The authors developed this method by using polonium and a radiation-sensitive substance. As the alpha particles emitted have a very short range the reaction is restricted to the vicinity of the interface, and it can be assumed that the reaction rate is directly proportional to the interfacial area. The measurement of the interfacial area is often of great importance, especially in the analysis of mass transfer operations. The experimental system consisted of a solution of polonium nitrate which was in contact with the liquid fluorocarbon. Boron trifluoride tubes were used for the detection of neutrons created by the α,n reaction on fluorine. Of course, it must be pointed out that this method is only possible if one has two immiscible liquid phases. But the method is directly adaptable to remote operations and, perhaps more important, can be applied to opaque systems.

Another important paper which deals with the use of alpha particles is the one on the determination of mass distribution. The possibility of using alpha particles for this purpose has been discussed many years ago. In this case the authors set out to construct an alpha camera which can work under reduced pressure in order to change the length of the path of the alpha particles. The great advantage of using alpha rays is, of course, the almost straight path which the particles take and their impressive effect on photographic emulsions. The difference in mass per unit area between different parts of the object under investigation causes a change in the energy distribution of the penetrating alpha particles before striking the photographic emulsion. The blackening of the exposed emulsion will be proportional to the energy of the penetrating particles, and the variation in the density of the film will correspond to the variation in stopping power and mass per unit area. In addition it is necessary to expose a suitable object in order to have a calibrated reference system. The polonium-210 was in this case fixed by electrodeposition on platinium, the active area having a diameter of 2 mm. The materials chosen were biological ones having a wide mass distribution. Microtome sections of the selected substances which had thicknesses varying between 8–20 μ were used. The enlarged microradiographs of a section of human appendix which were obtained as a result of this method are very impressive indeed. The smallest mass differences determined under the experimental conditions were 3.3×10^{-13} and 6.7×10^{-14} g, depending on the absolute mass. It was estimated that the experimental errors should be within $\pm 18\%$. Considering the limit of the resolving power of the emulsion used, the authors believe that mass differences of 10^{-15} g can be detected by improving the experimental set up. This work has quite definitely established the value of this method in biology, and the same method is applicable to medicine, textile research and in many other fields. It will therefore be a very powerful and novel help in many research applications.

Interesting results were achieved by an investigation of steel corrosion in the presence of H_2S using radioactive methods. This paper shows once more how effective such research application can be.