

EDITORS

J. KISTEMAKER · J. BIGELEISEN · A. O. C. NIER

PROCEEDINGS  
OF THE  
INTERNATIONAL  
SYMPOSIUM  
ON ISOTOPE  
SEPARATION

# PROCEEDINGS OF THE INTERNATIONAL SYMPOSIUM ON ISOTOPE SEPARATION

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*Edited by*

J. KISTEMAKER

*Professor of Physics, F.O.M., Amsterdam*

J. BIGELEISEN

*Senior Chemist, Brookhaven National Laboratory*

A. O. C. NIER

*Professor of Physics, University of Minnesota*



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# PROCEEDINGS OF THE INTERNATIONAL SYMPOSIUM ON ISOTOPE SEPARATION

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## INTRODUCTION

It gives rise to great satisfaction that the many interesting papers read at the first Symposium on Isotope Separation, Amsterdam 1957, are now to be published.

We wish again to express our gratitude to the participants of the Symposium for their contributions and to the editors for accomplishing the task of preparing this publication.

More than on anyone else it was on Professor J. Kistemaker that the main burden of the organization of the congress fell, and thus it is also to him that the credit must go for its success of which this volume is a testimony.

**J. A. A. KETELAAR**

*President of the*

*International Symposium on Isotope Separation,*

*Amsterdam 1957*

## GENERAL INTRODUCTION

BY

HAROLD C. UREY

Isotopes were discovered as a result of the study of the natural radioactive elements. One of the first research papers that I read as a student was that of T. W. Richards on the atomic weight of leads from uranium and thorium minerals, showing that the atomic weights of the leads determined by chemical means were different from that of ordinary lead. Sir J. J. Thompson then demonstrated that neon consisted of a mixture of atoms of different atomic weights. He was the first to use electric and magnetic fields to separate the isotopes and this oldest method of separation has been discussed at this meeting in its most recent and elegant forms. It has led to our very precise mass spectrometers and spectrographs through the work of Aston, Dempster, Nier, Bainbridge, Mattauch and Inghram and others. This method of separation supplies us with small but useful amounts of isotopes of many elements.

Early in the history of this subject Harkins and Hevesy changed the ratios of the isotopes of mercury by molecular distillation and this has been applied to the lithium isotopes. Also diffusion through porous materials was used by Aston and Mulliken at about the same time to partially separate the isotopes of neon and chlorine. This latter method has become the most important one for the separation of the uranium isotopes and unfortunately it cannot be discussed as yet at scientific conferences of this kind.

Since the 1920's many methods for isotope separation have been proposed and developed. The differences in thermodynamic properties of isotopic substances, i.e. differences in vapor pressures and the equilibrium constants of exchange reactions have led to the large industrial scale separations of the hydrogen and boron isotopes and to the separation of the oxygen and nitrogen isotopes on large laboratory scales, as reported at this conference. These methods when applied on a large scale produce separated isotopes at the lowest cost of any processes so far used. However,

they can be applied only in very special cases and each separation of the isotopes of any element is a special problem.

The ion exchanger method and the electrolytic method depend for their effectiveness on a combination of kinetic and thermodynamic differences in properties. This is surely true of the electrolytic method, since a fractionation factor for the hydrogen isotopes is expected on thermodynamic grounds and at the same time the observed factor is larger than that predicted on this basis. The ion exchanger method is similar to the exchange reaction methods, but probably includes some kinetic effect.

Thermal diffusion has continued to be a most interesting and valuable method of separation since its discovery and development some twenty years ago. It supplied some of the enriched  $U^{235}$  for the Manhattan District program and that work has been presented in this symposium.

Two methods of separation have been presented in this symposium which are relatively new. They are the electromigration and gas centrifuge methods. The former has produced nearly pure lithium and chlorine isotopes and it appears to be capable of application to other elements. The gas centrifuge method is one which requires large and very accurately built apparatus. The theory is well understood but reduction to practice is difficult. Much has been accomplished in this problem as is reported in this symposium.

The electromagnetic methods produce large separation factors, but very small throughput of material, while other methods, notably the diffusion and chemical exchange methods, supply small separation factors but very large throughput. The latter mostly have won the field of large production, while the former have been more versatile in the field of small production. Whether this general pattern will change in the future as other methods are perfected is very difficult to predict. In many cases we can be quite sure that this will not happen, but in others a decision depends on very careful weighing of costs, reliability of operation and similar factors. That any new methods of decisively advantageous character will be discovered is doubtful in the opinion of the writer. Any certain prediction in this respect is dangerous, but this appears to be a reasonable estimate at the present time, because differences in the properties of the isotopes other than those of hydrogen are so small.

The usefulness of separated isotopes for atomic energy applications are obvious, but in the case of the less spectacular nuclear species very many applications to research can be made, as has been true in the past. Separated isotopes will appear in problems of chemistry, physics and biology and many industrial problems and we will meet them again in papers presented at most scientific meetings and symposia. Even in such

a wide spectrum of scientific and technological disciplines, there are several common interests in the uses of isotopes. They lie primarily in the areas of measurement and purification. The results of a scientific investigation, which may utilize isotopes as a tool, are most effectively discussed in the domain of the specific scientific or technological discipline.

## PREFACE

The first international symposium on isotope separation convened in Amsterdam in April 1957, almost half a century after the discovery of isotopes. This long lapse may at first seem surprising, but is primarily an indication of the difficulty in finding coupling mechanisms whereby separative work can be fed into a mixture of isotopes. The short history of the subject of isotope separation, incorporated in Professor Urey's introduction, points out that up until the late nineteen thirties the science of isotope separation was studied by a few pioneers, hardly sufficient to support more than a round table conference. The discovery of nuclear fission, followed by its application to energy production, and the uses of separated isotopes in nuclear science, biology, chemistry, and other scientific disciplines have provided a major impetus to the development of the science and technology of isotope separation. In the past decade there have been several small conferences on isotope separation; such as the Gordon Research Conferences on the chemistry and physics of isotopes, the Harwell Conference on electromagnetic separation of isotopes in 1955, and the symposium on heavy water production in Rome in the spring of 1955 under the auspices of the European Atomic Energy Society. In addition several papers on isotope separation were presented at the 1955 International Conference on the Peaceful Uses of Atomic Energy held in Geneva. None of these conferences afforded a medium for a general discussion of the principles and the application of the many different methods of isotope separation. None of them provided the opportunity for the exchange of information on an international scale between scientists working in the field.

In the spring of 1956 the Netherlands Physical Society, in collaboration with the International Union of Pure and Applied Physics, actively undertook the arduous task of organizing a symposium on isotope separation. The response to the preliminary announcements indicated widespread interest in such a symposium and it became necessary to limit the attendance to about 200 specialists in order to provide an effective medium for discussions. Invaluable advice and collaboration came spontaneously from many quarters. Special mention must be made of the valuable suggestions rendered by Dr. Jules Guéron of Saclay, Dr. H. Kronberger of Risley, and Dr. C. P. Keim of Oak Ridge.

The participants of the symposium were unanimous in their expression of gratitude to the Netherlands Physical Society for organizing the symposium, to the City of Amsterdam for their hospitality, and to the Vrije Universiteit of Amsterdam for their role as hosts for the meeting. They also acknowledged their thanks to Professor J. A. A. Ketelaar for his efforts as president of the symposium.

Arrangements were made at the outset with North-Holland Publishing Co. to publish the papers presented at the symposium and the pertinent discussion. In the case of the long invited talks, it was deemed desirable to include, in the proceedings, material of a review nature. For the publication of the contributed papers on original research, criteria similar to those of the major scientific journals, were adopted by the editors. Some of the authors have preferred to publish their papers in full in the regular scientific journals. These authors have kindly consented to the inclusion of abstracts of their papers in the present volume.

The sessions of the symposium were organized according to method of separation rather than by particular isotope. This organization of the program proved very successful in containing the interest of the participants in the symposium and in stimulating discussion. Therefore, the same arrangement was adopted in the compilation of the present volume. In the case of isotope separation by distillation, the papers have been divided into two categories: (1) those dealing primarily with the theory and measurement of vapor pressure differences, and (2) those concerned mainly with the distillation process.

We are grateful to the members of the Laboratorium voor Massaspectrografie and the Department of Physics of the Vrije Universiteit for their aid in the compilation of the discussions at the symposium. The burden of the preparation of the manuscript was materially lightened by the aid given by our secretary, Miss J. J. M. van der Berg. Without her help this volume could not have been prepared.

Finally, we wish to thank North-Holland Publishing Co. for the care and speed with which they have expedited the publication of this volume.

*Amsterdam, Netherlands*

*Upton, L. I., New York*

*Minneapolis, Minnesota*

**March 1958**

**J. KISTEMAKER**

**J. BIGELEISEN**

**A. O. C. NIER**

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