

The
Transuranium
Elements



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Division IV—Plutonium Project Record
Volume 14 B

THE TRANSURANIUM ELEMENTS

Research Papers

Part I: Papers 1.1 to 6.39

Contributing Authors of Part I

B. M. Abraham	E. L. King
D. P. Ames	C. W. Koch
H. H. Anderson	T. P. Kohman
W. C. Beard, Jr.	D. E. Koshland, Jr.
N. A. Bonner	K. A. Kraus
B. B. Brody	J. C. Kroner
H. D. Brown	T. J. LaChapelle
H. S. Brown	C. K. McLane
M. Calvin	E. M. McMillan
M. Cefola	W. H. McVey
R. E. Connick	L. B. Magnusson
O. A. Cook	G. E. Moore
C. D. Coryell	T. B. Novey
B. B. Cunningham	E. F. Orlemann
J. R. Dam	R. L. Patton
N. R. Davidson	M. L. Perlman
J. S. Dixon	T. E. Phipps
N. Elliott	W. H. Reas
D. W. Engelkemeir	G. T. Seaborg
M. W. Evans	G. W. Sears
S. Fried	E. Segrè
C. S. Garner	R. L. Seifert
L. H. Gevantman	I. Sheft
J. W. Gofman	G. E. Sheline
F. Hagemann	O. C. Simpson
J. W. Hamaker	N. R. Sleight
R. E. Hein	C. Smith
J. C. Hindman	L. Spector
H. H. Hopkins, Jr.	A. Turkevich
J. J. Howland, Jr.	A. F. Voigt
E. K. Hyde	A. C. Wahl
A. H. Jaffey	L. B. Werner
A. Kant	E. F. Westrum, Jr.
I. Karle	M. J. Wolf
M. Kasha	F. J. Wolter
J. J. Katz	J. M. Wright
J. W. Kennedy	

A complete list of contributing authors for Parts I and II, with affiliations, is given at the end of Part II.

THE TRANSURANIUM ELEMENTS

Research Papers

Edited by

GLENN T. SEABORG

Department of Chemistry and Radiation Laboratory
University of California, Berkeley

JOSEPH J. KATZ

Chemistry Division, Argonne National Laboratory

and

WINSTON M. MANNING

Chemistry Division, Argonne National Laboratory

Part I
(Papers 1.1 to 6.39)

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THE TRANSURANIUM ELEMENTS

Research Papers

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FOREWORD

The United States program of development of atomic energy has been described by Major General L. R. Groves, who, as Commanding General of the War Department's Manhattan Project, directed the program from mid-1942 until December 31, 1946, as "a generation of scientific development compressed into three years." The tremendous scope of the Manhattan Project Technical Section of the National Nuclear Energy Series, which has been in preparation since 1944, is a tribute to the unprecedented accomplishments of science, industry, government, labor, and the Army and Navy working together as a team. These volumes can be a firm foundation for the United States atomic energy program which, in the words of the Atomic Energy Act of 1946, is ". . . directed toward improving the public welfare, increasing the standard of living, strengthening free competition in private enterprise, and promoting world peace."

David E. Lilienthal, Chairman
U. S. Atomic Energy Commission

ACKNOWLEDGMENT

The Manhattan Project Technical Section of the National Nuclear Energy Series embodies results of work done in the nation's wartime atomic energy program by numerous contractors, including Columbia University. The arrangements for publication of the series volumes were effected by Columbia University, under a contract with the United States Atomic Energy Commission. The Commission, for itself and for the other contractors who contributed to this series, wishes to record here its appreciation of this service of Columbia University in support of the national nuclear energy program.

PREFACE

This volume is one of a series which has been prepared as a record of the research work done under the Manhattan Project and the Atomic Energy Commission. The name Manhattan Project was assigned by the Corps of Engineers, War Department, to the far-flung scientific and engineering activities which had as their objective the utilization of atomic energy for military purposes. In the attainment of this objective, there were many developments in scientific and technical fields which are of general interest. The National Nuclear Energy Series (Manhattan Project Technical Section) is a record of these scientific and technical contributions, as well as of the developments in these fields which are being sponsored by the Atomic Energy Commission.

The declassified portion of the National Nuclear Energy Series, when completed, is expected to consist of some 60 volumes. These will be grouped into eight divisions, as follows:

- Division I — Electromagnetic Separation Project
- Division II — Gaseous Diffusion Project
- Division III — Special Separations Project
- Division IV — Plutonium Project
- Division V — Los Alamos Project
- Division VI — University of Rochester Project
- Division VII — Materials Procurement Project
- Division VIII — Manhattan Project

Soon after the close of the war the Manhattan Project was able to give its attention to the preparation of a complete record of the research work accomplished under Project contracts. Writing programs were authorized at all laboratories, with the object of obtaining complete coverage of Project results. Each major installation was requested to designate one or more representatives to make up a committee, which was first called the Manhattan Project Editorial Advisory Board, and later, after the sponsorship of the Series was assumed by the Atomic Energy Commission, the Project Editorial Advisory Board. This group made plans to coordinate the writing programs at all the installations, and acted as an advisory group in all matters affecting the Project-wide writing program. Its last meeting was held on Feb. 9, 1948, when it recommended the publisher for the Series.

The names of the Board members and of the installations which they represented are given below.

Atomic Energy Commission Public and Technical Information Service	Alberto F. Thompson
Technical Information Branch, Oak Ridge Extension	Brewer F. Boardman
Office of New York Operations	Charles Slessor, J. H. Hayner, W. M. Hearon *
Brookhaven National Laboratory	Richard W. Dodson
Carbide & Carbon Chemicals Corporation (K-25)	R. B. Korsmeyer, W. L. Harwell, D. E. Hull, Ezra Staple
Carbide & Carbon Chemicals Corporation (Y-12) †	Russell Baldock
Clinton Laboratories ‡	J. R. Coe
General Electric Company, Hanford	T. W. Hauff
General Electric Company, Knolls Atomic Power Laboratory	John P. Howe
Kellex Corporation	John F. Hogerton, Jerome Simson, M. Benedict
Los Alamos	R. R. Davis, Ralph Carlisle Smith
National Bureau of Standards	C. J. Rodden
Plutonium Project Argonne National Laboratory	R. S. Mulliken, H. D. Young
Iowa State College	F. H. Spedding
Medical Group	R. E. Zirkle
SAM Laboratories §	G. M. Murphy
Stone & Webster Engineering Corporation	B. W. Whitehurst
University of California	R. K. Wakerling, A. Guthrie
University of Rochester	D. R. Charles, M. J. Wantman

* Represented Madison Square Area of the Manhattan District.

† The Y-12 plant at Oak Ridge was operated by Tennessee Eastman Corporation until May 4, 1947, at which time operations were taken over by Carbide & Carbon Chemicals Corporation.

‡ Clinton Laboratories was the former name of the Oak Ridge National Laboratory.

§ SAM (Substitute Alloy Materials) was the code name for the laboratories operated by Columbia University in New York under the direction of Dr. H. C. Urey, where much of the experimental work on isotope separation was done. On Feb. 1, 1945, the administration of these laboratories became the responsibility of Carbide & Carbon Chemicals Corporation. Research in progress there was transferred to the K-25 plant at Oak Ridge in June, 1946, and the New York laboratories were then closed.

Many difficulties were encountered in preparing a unified account of Atomic Energy Project work. For example, the Project Editorial Advisory Board was the first committee ever organized with representatives from every major installation of the Atomic Energy Project. Compartmentation for security was so rigorous during the war that it had been considered necessary to allow a certain amount of duplication of effort rather than to permit unrestricted circulation of research information between certain installations. As a result, the writing programs of different installations inevitably overlap markedly in many scientific fields. The Editorial Advisory Board has exerted itself to reduce duplication in so far as possible and to eliminate discrepancies in factual data included in the volumes of the NNEs. In particular, unified Project-wide volumes have been prepared on Uranium Chemistry and on the Analysis of Project Materials. Nevertheless, the reader will find many instances of differences in results or conclusions on similar subject matter prepared by different authors. This has not seemed wholly undesirable for several reasons. First of all, such divergencies are not unnatural and stimulate investigation. Second, promptness of publication has seemed more important than the removal of all discrepancies. Finally, many Project scientists completed their contributions some time ago and have become engrossed in other activities so that their time has not been available for a detailed review of their work in relation to similar work done at other installations.

The completion of the various individual volumes of the Series has also been beset with difficulties. Many of the key authors and editors have had important responsibilities in planning the future of atomic energy research. Under these circumstances, the completion of this technical series has been delayed longer than its editors wished. The volumes are being released in their present form in the interest of presenting the material as promptly as possible to those who can make use of it.

The Editorial Advisory Board

PLUTONIUM PROJECT RECORD FOREWORD

This report is a technical account of information collected while developing methods for producing plutonium. Some of the information deals directly with nuclear physics and chemistry. Most of it is related rather to technical processes that needed to be performed in preparation for making the plutonium. These publications represent selections from the great mass of current reports, made on the basis of their value to basic science and technology.

The current technical reports, written during the war years, were essential to the active work of the plutonium project. They supplied needed data and calculations to those who were planning the new processes. Selecting from this mass of records the most reliable data and presenting them in a useful form has been an enormous task, for which the writers and editors of these volumes deserve the sincere thanks of their scientific colleagues. Many fields of science and technology will develop more rapidly because of this knowledge.

The efforts of the men who did this research resulted in the successful production of atomic bombs, which shortened the war and saved the lives of many of their comrades. But in the long view of history it is probable that the major human heritage from their work will not be this quick victory. It may not even be the useful applications of atomic energy, which was first presented as a Promethean gift to man. It is not unlikely that the scientific information in these pages may be the starting point to new reaches of knowledge, which will give to man an understanding that will truly enrich his life.

Arthur H. Compton

INTRODUCTORY NOTE ON THE PLUTONIUM PROJECT RECORD

Organization and Record of the Metallurgical Project. The Plutonium Project Record, which forms Division IV of the National Nuclear Energy Series (NNES) is the scientific and technical record of the former Metallurgical Project. The project had its origin in work carried on in 1940-1941, mainly at Columbia and Princeton on the development of the chain-reacting pile and at the University of California at Berkeley on the production and chemistry of transuranic elements. In January 1942 this work was concentrated in the newly organized Metallurgical Laboratory at Chicago under the leadership of A. H. Compton. The Metallurgical Project grew out of the Metallurgical Laboratory. The initial objectives of the Metallurgical Laboratory were (1) to develop chain-reacting piles to produce plutonium and (2) to develop fission bombs. Major associated units were organized in 1942 at Iowa State College at Ames, Iowa (chemistry and metallurgy) under F. H. Spedding; at the University of California at Berkeley, Calif. (chemistry) under W. M. Latimer and E. D. Eastman, continuing the previous work there; and at Massachusetts Institute of Technology (metallurgy) under J. Chipman and later M. Cohen. Early in 1943 the work on fission bombs was transferred to an independent project at Los Alamos.

After the successful demonstration of a nuclear chain reaction in the West Stands pile at Chicago in December 1942, the Argonne Laboratory with its experimental pile was built west of Chicago, and the Clinton Laboratories with their pilot-plant pile were built at Oak Ridge, Tenn.—both in 1943. The three major laboratories at Chicago, Argonne, and Clinton, the associated laboratories at Ames, Berkeley, and M.I.T., and some seventy other cooperating groups then constituted the Metallurgical Project, under A. H. Compton as Project Director. Closely cooperating in the transition from laboratory and pilot-plant to large-scale operation was E. I. du Pont de Nemours & Company, which was made responsible for the design and construction of the Clinton pile and for the design, construction, and operation of the Hanford Plutonium Plant. The Project continued as such until June 30, 1945, when it was dissolved.

The Plutonium Project Record (PPR) covers most of the scientific and technical work of the Metallurgical Laboratory and the Metallurgical Project up to the date of the dissolution of the Project, and also

the continuation of this work in the successor laboratories up to approximately Jan. 1, 1946, or in some cases to a later date. In addition, the PPR covers in part the pre-1942 work at Columbia, Princeton, and Berkeley. The record of the work directly leading up to the Los Alamos Project, however, is omitted. Nevertheless the PPR and the Los Alamos Technical Series (Division V of the NNES) cover closely related and in part overlapping subject matter in some of their volumes, particularly in nuclear physics and in chemistry and metallurgy of plutonium.

Important phases of the work of the Metallurgical Project that are not reported in the PPR but will be reported elsewhere in the NNES are as follows: (1) Division VII, the report of the Materials Procurement Project, includes certain early work on process metallurgy, which was initiated largely by the Metallurgical Laboratory. (2) The Division VIII NNES volumes on Analytical Chemistry, which developed from two volumes originally planned as part of the PPR, contain much Metallurgical Project work, including one complete Collected Papers volume. (3) The Division VIII NNES volumes on Uranium Chemistry, which were planned and carried out under the supervision of the PPR editorial group, likewise contain much Metallurgical Project work, including one complete Collected Papers volume.

History and Plan of the Plutonium Project Record. During the war years the scientific and technical work of the Metallurgical Project and its associated laboratories was described currently in a series of reports called the "C reports." The work up to July 1, 1945 was described in some 3,000 reports. After that date the Clinton Laboratories reports became a separate series, but reports of the other units of the former Metallurgical Project continued to be issued as C reports. Most of the C reports were preliminary or semifinal reports. The main consideration during the wartime development was speed of issue and distribution.

As the mass of scientific and technical knowledge obtained on the Project piled up, an increasing need was apparent for its digestion into survey or summary form. In partial answer to this need, an editorial group was set up in the spring of 1943 to organize a Project Handbook. Although never fully completed because of the engrossment of authors in immediately urgent tasks, and because of the transfer of many of them to other sites, enough of the Project Handbook was finished to be of real value.

By the summer of 1944, the Metallurgical Project had largely concluded its major task, that of providing the scientific and pilot-plant know-how for the design of the large-scale Hanford Plutonium Plant. The time seemed ripe to plan a series of volumes in which the Project's fund of accumulated scientific and technical knowledge would be

recorded. These would replace the often sketchy and sometimes mutually contradictory C reports and fill many gaps of unwritten knowledge. In the early planning, Laurence L. Quill as Chief of the Editorial Section of the Project Information Division during the summer of 1944, Eugene Rabinowitch, and H. H. Goldsmith made important contributions. After several committee meetings, a plan for the preparation of a Metallurgical Project Record was approved by the Project Director in the fall of 1944. Later, in 1945, the name was changed to Plutonium Project Report or Record (PPR).

When the PPR was organized, rigid compartmentation was still in effect between the Metallurgical Project and the other Manhattan District projects. Members of each project were in general not supposed to know even the major objectives or main outlines of the other projects. The PPR had therefore to be planned as an independent entity. Nevertheless, at its inception the idea was firmly held that later on the Record should become part of a larger series covering the work of all the atomic energy projects. This idea was repeatedly advocated and led in late 1945 to the plan for the Manhattan Project Technical Series (MPTS), a name which was finally revised to the present designation of National Nuclear Energy Series (NNES).

The general plan of organization of the PPR was that of a series of some twenty Survey volumes, called "A volumes," each documented by a like-numbered Collected Papers volume (or volumes); these were called "B volumes." In general, following somewhat a pattern set by the Project Handbook, a Survey volume was planned for each scientific or technical subject to which the Metallurgical Project had made sufficiently major contributions. Each Survey volume was intended to be a fairly complete review or monograph (or else a collection of review chapters) on the subject field. It was planned to cover work done both within and outside the Metallurgical Project, though with primary emphasis on the former, outside work being included only for the sake of accuracy and completeness.

In contrast to the Survey volumes, each Collected Papers volume was designed to consist of individual papers, mostly from individual laboratories and more or less similar to articles in the scientific journals; they were to include only work done within the Project. In planning the PPR, it was realized that some of the Survey volumes would overlap with possible volumes of other projects, but because of compartmentation restrictions, it was decided to proceed in general with the plan as outlined. An exception was the field of uranium chemistry, where it was obvious that all the major projects were making important contributions. In this field, a Handbook of Uranium Chemistry was planned early in 1944, to be edited and written at the Metallurgical Laboratory at Chicago, but as a cooperative effort of all the

projects, and based on a full interchange of information among them. When the Record was organized, this volume was tentatively included as one of the PPR Survey volumes, to be accompanied by a corresponding Collected Papers volume covering Metallurgical Project work only. Later, when the MPTS (now NNES) was organized, these volumes, with the addition of Collected Papers from the other projects, were transferred to the over-all Division (Division VIII) of the technical series. In the field of analytical chemistry, a Survey volume and a Collected Papers volume were planned for the PPR and were well on their way toward completion. When the MPTS was organized, the content of these volumes was pooled with the work of other projects of the Manhattan District to form Survey and Collected Papers volumes of Division VIII of the MPTS. In certain other fields, pooling of material from the different projects was also considered, but was felt to involve too large a task of reorganization.

Because of the wide variety of subject matter, the organization of the PPR into Survey volumes, each accompanied by one or more Collected Papers volumes, is not always consistently followed. There are a few Collected Papers volumes without corresponding Survey volumes, and the converse is also true. Furthermore, the form of organization varies considerably from one volume to another because of varying subject matter and the preferences of the different volume editors and committees.

When the PPR plans were approved toward the end of 1944, the completion deadline for the manuscripts was set for June 30, 1945, the date of dissolution of the Metallurgical Project. Most of the PPR volumes were organized into three groups: (1) chemistry and metallurgy; (2) physics and related engineering; (3) biology and medicine. The first task was to obtain volume editors and editorial committees for the various volumes, to plan the contents, and to find authors. John C. Warner, as chemistry editor of the PPR and Chief of the Editorial Section of the Project Information Division from December 1944 to June 30, 1945, made decisive contributions to the chemistry and metallurgy volumes and to the general planning of the PPR.

The organization of the volumes on physics and on biology and medicine went more slowly, partly because the subject matter was then less ripe for writing than was that on chemistry and metallurgy, partly because of the demands for continuing research and, in the field of instrumentation, for production of instruments to be used at Los Alamos, Hanford, and other sites. Eugene P. Wigner, Frederick Seitz, and H. H. Goldsmith took an active part in the early organization of the physics volumes. Plans for the volumes on biology and medicine were very effectively organized by Raymond E. Zirkle as PPR editor for these fields, with the backing of Robert S. Stone as Associate

Project Director for Health. Hoylande D. Young entered the PPR program as Technical Editor in charge of final editing and processing of manuscripts, and after June 30, 1946, became General Editor.

After the organization of the PPR, steady progress was made in the work of writing and editing, but at a slower pace than was originally hoped. The dissolution of the Project on June 30, 1945, with the readjustments and administrative problems involved in a 50 per cent cut of total personnel; the end of the war after the bomb was dropped in August and the subsequent deep preoccupation and extensive activities of Project personnel in connection with the social and political implications of atomic energy and atomic warfare; new research and planning directed toward the postwar continuation of the atomic energy program; all these slowed the progress of the PPR writing program. During this difficult period, invaluable encouragement and support of the PPR program came from, among others, Norman Hilberry, Associate Director of the Metallurgical Project up to the time of its dissolution, and Farrington Daniels, Director of the Metallurgical Laboratory in 1945-1946.

Meantime, other projects in the Manhattan District group began the preparation of final accounts of their work. In particular, the Los Alamos Technical Series was begun in 1945. Finally, the MPTS (now the NNES) was organized under the Manhattan District Editorial Advisory Board late in 1945. Under the chairmanship of Alberto F. Thompson, as Chief of the Publications Section of the Research Division of the District, this group began the task of coordinating existing writing activities and filling the gaps in these, with the objective of producing a reasonably well-rounded series of volumes covering the work of the entire District. During early 1946, rules for declassification were set up, and the editors of the MPTS volumes faced the difficult task of dividing the subject matter of their volumes into declassifiable parts, publishable immediately, and classified parts, for which publication must be deferred. In June 1947 the completion of the editorial work of the PPR, as part of the NNES, was taken over by the Technical Information Division of the Atomic Energy Commission, at Oak Ridge, Tenn.

In addition to those named above, many other project members worked together in planning the PPR. After the general plans were made, the actual work of preparing the various volumes was in the hands of the volume editors, volume editorial committees, and authors, as described in the prefaces of the individual volumes.

Robert S. Mulliken
Editor-in-Chief,
Plutonium Project Record

The Manhattan Project Technical Section of the National Nuclear Energy Series is intended to be a comprehensive account of the scientific and technical achievements of the United States program for the development of atomic energy. It is not intended to be a detailed documentary record of the making of any inventions that happen to be mentioned in it. Therefore, the dates used in the Series should be regarded as a general temporal frame of reference, rather than as establishing dates of conception of inventions, of their reduction to practice, or of occasions of first use. While a reasonable effort has been made to assign credit fairly in the NNES volumes, this may, in many cases, be given to a group identified by the name of its leader rather than to an individual who was an actual inventor.

VOLUME EDITORS' PREFACE

This volume consists of more than 150 original research papers dealing with the transuranium elements and cognate topics. The papers are intended to be of the same type as those ordinarily appearing in chemistry and physics journals. They are published in this rather unorthodox way in order to lend a certain measure of coherence to a subject which, to a large extent, has developed in an unusual way under the cloak of secrecy. It is also to be hoped that publication as a unit will aid in the fair distribution of credit to the many workers who have contributed to this subject.

The papers in this volume cover work begun as early as 1940. Although the original intention was to include only work accomplished during the war years, a number of communications in this volume deal with work done in the very recent past. It was in many cases difficult to draw a line between work done during the war years and research of a more recent date; this was particularly true in the many instances when postwar work depended directly on work of the war years. The end of the war found the contributors to this volume scattered over the entire country. As a consequence the preparation of this volume has been extended over a period of about four years. This has resulted in some inconsistency in the degree of detail with which the individual papers cover their subjects.

In general, the editors have refrained from extensive editorial work on the individual papers. It is believed that a large proportion of the papers meet the standards required by scientific journals for publication, but it is also acknowledged that some of the papers may fall below these standards for one reason or another. Nevertheless, it has been the intention of the editors that a rather liberal policy be followed with respect to the inclusion of papers in the volume. The authors of the papers must accept primary responsibility for presentation, accuracy of results, and conclusions. The editors have made a serious effort to eliminate errors from the text, but discrepancies are known to occur in various papers treating the same subject. In some cases these are the result of lack of definitive work. Included in the volume are a number of papers of historical interest which give nuclear constants now superseded by more accurate values. It will be clear from the text which values are to be considered the most reliable. It is planned to correlate these data in a more definitive

way in a survey volume on the transuranium elements which is now in active preparation.

The papers are devoted to the four known transuranium elements: neptunium (atomic number 93), plutonium (number 94), americium (number 95), and curium (number 96). There are, of course, many more papers concerning plutonium than there are concerning the other transuranium elements and, in turn, more papers about neptunium than there are about americium and curium. There are several papers concerning radium, actinium, thorium, protactinium, and uranium, which are included here for reference convenience. The papers concerning these other heavy elements are being published for the most part in other volumes of the National Nuclear Energy Series.

Although the papers included in this volume give on the whole a fairly representative picture of wartime work with the transuranium elements, there are a number of omissions that should be mentioned. None of the work in connection with the development of the nuclear weapon itself, which was done at the Los Alamos Laboratory in New Mexico, has been covered. Similarly, the development of plant processes for the separation of plutonium from uranium and the fission products is not described in these papers.

In order to provide some background, which may not be obtainable from a reading of the papers alone and which may help in giving perspective to the individual papers, it has seemed worth while to include here a brief historical survey of the outstanding developments concerning the transuranium elements as they took place at the various laboratories. This brief survey deals with the development of knowledge of the basic scientific facts about these elements and does not touch upon the technological information that was gathered by another group, fully as large and as diligent as the group concerned with the basic aspects of the work. For clarity of presentation it has seemed desirable to describe the historical development of our knowledge of the basic facts concerning each of the transuranium elements separately rather than to attempt to give data for the entire group in exact chronological order. In the following paragraphs, therefore, the story of each of these elements is told separately.

Neptunium. The first transuranium element was discovered by E. M. McMillan and P. H. Abelson at the University of California in May 1940. This research had no connection with the wartime program and was published at that time in the *Physical Review*. Using the neutrons from the 60-in. cyclotron, the investigators were able to show with the help of chemical work that a radioactivity of 2.3 days half life formed during the irradiation of uranium is due to the isotope

${}_{93}^{239}$, which is the decay product of the 23-min ${}_{93}^{239}$ formed by radioactive neutron capture in ${}_{92}^{238}$. Because of the similarity in chemical properties of element 93 to uranium, the discoverers of the new element named it "neptunium" after Neptune, the planet immediately beyond Uranus, which had given its name to uranium.

For the next few years the chemical properties of neptunium were studied exclusively by the tracer technique, using this isotope. Following this original work, in addition to the secret wartime work of G. T. Seaborg and A. C. Wahl, represented by a paper in this volume, there were a number of German investigations, descriptions of which were published in the German scientific journals. Later the tracer chemistry of neptunium was studied at a number of sites connected with the Plutonium Project, and some papers covering this work are included here.

Another isotope of neptunium, Np^{237} , was discovered early in 1942 as a result of irradiating uranium with fast neutrons from the Berkeley cyclotron. This isotope is the decay product of the previously known 7-day β -particle-emitting ${}_{92}^{237}$, which is formed as a result of an $(n,2n)$ reaction on ${}_{92}^{238}$. The isotope Np^{237} is of particular importance because it has a very long life, with a half life of 2.20×10^6 years, and because it is available in weighable amounts. It is produced during the operation of the large uranium chain-reacting units, a very fortunate circumstance, for otherwise it is probable that the element neptunium would not be available for thorough study in the macroscopic state.

The element neptunium in the form of pure compounds was first isolated by L. B. Magnusson and T. J. LaChapelle in July 1944 at the wartime Metallurgical Laboratory of the University of Chicago (now the Argonne National Laboratory). This work was done by using the techniques of ultramicrochemistry and involved amounts of the order of micrograms formed largely by cyclotron bombardments.

A few months later this isotope began to become available as a by-product from the operation of the uranium chain-reacting piles. The first few milligrams from this source were isolated near the end of 1944 as a result of special runs, designed for the extraction of neptunium, in the chemical-extraction plant that was operated in conjunction with the pile at the wartime pilot plant of the Clinton Laboratories (now the Oak Ridge National Laboratory) in Tennessee. The Np^{237} from this source was concentrated and isolated in pure form by a group of chemists who came to the Clinton Laboratories from the Metallurgical Laboratory in a collaborative effort of the two laboratories. This group was headed by J. J. Katz and consisted of L. B.

Magnusson, T. J. LaChapelle, J. R. Gilbreath, and W. C. Beard. These initial few milligrams served as the supply which made it possible to determine many of the chemical properties of neptunium on the microchemical scale of investigation.

In the following year it became possible to obtain larger amounts of neptunium, amounting to some hundreds of milligrams, as a by-product of the operation of the plutonium-production plant at the Hanford Engineer Works in the state of Washington. A group of chemists at the Metallurgical Laboratory, working under the direction of F. W. Albaugh and R. C. Thompson, devised a process for the extraction of neptunium at the Hanford Engineer Works which was found to be successful. Using the material from these sources, a study of the chemistry of neptunium was carried on at the Metallurgical Laboratory. J. C. Hindman was the leader of a group including L. B. Magnusson and T. J. LaChapelle, which investigated the chemical, including the thermodynamic, properties of this element in aqueous solution. A study of the compounds that could be prepared in a dry way by the use of vacuum-line techniques was made by S. Fried and N. R. Davidson. There has been experimental work concerning this element with the isotope Np^{237} at other sites, notably the work of K. A. Kraus and coworkers at the Clinton Laboratories and, more recently, work at the Radiation Laboratory at Berkeley, California.

Papers describing most of these experiments on neptunium are included in this volume. Work is continuing, and there will certainly be many more publications in the future.

Plutonium. Plutonium has been the most thoroughly studied of all the transuranium elements. This element has been the object of investigation at a large number of the sites of the Plutonium Project, which was organized for the purpose of producing the isotope Pu^{239} . Plutonium, the second transuranium element to be discovered, was first produced by G. T. Seaborg, E. M. McMillan, A. C. Wahl, and J. W. Kennedy late in 1940. This investigation took place at the University of California in Berkeley. It was found that the irradiation of uranium with deuterons in the 60-in. cyclotron leads to the production of the neptunium isotope Np^{239} , which decays to the 100-year Pu^{239} . The name "plutonium" was suggested to follow the system used in the naming of uranium and neptunium.

During 1941 and the early part of 1942, the Pu^{239} was used at the University of California to investigate the chemical properties of plutonium by the tracer method. This early work ultimately formed the basis for the later development of the separation process used in connection with the large-scale production of plutonium. One important experiment, not of tracer character, was also made during this period. A macroscopic quantity of the isotope Pu^{239} , amounting

to about $0.5 \mu\text{g}$, was produced and isolated with a very small amount of rare-earth carrier material. This was done at the University of California by J. W. Kennedy, G. T. Seaborg, E. Segrè, and A. C. Wahl, who used this small sample to make the decisive demonstration that the isotope Pu^{239} undergoes fission with thermal neutrons.

Much of the wartime research on plutonium was devoted to working out chemical processes in connection with its production and utilization. Following the initial work at the University of California, the investigations on plutonium spread to other laboratories. Chief among these were the Metallurgical Laboratory, where the program got under way during the first half of 1942; the Los Alamos Laboratory in New Mexico, where work began early in 1943; the Clinton Laboratories in Oak Ridge, Tenn., which opened late in 1943; and the Hanford Engineer Works in Washington, which began operation in the latter half of 1944.

In addition to the program of study by the tracer technique, there was an important period, before the production plants went into operation, when the investigations were conducted with microgram amounts of Pu^{239} on the ultramicrochemical scale of operation. The main reason for this program was to test the proposed plutonium-separation process, tentatively worked out on the tracer scale chiefly by S. G. Thompson, at the actual concentrations that would be used at the Hanford plant.

As a result of this program the first chemically pure plutonium, free from carrier material and all other foreign matter, was prepared by B. B. Cunningham and L. B. Werner at the Metallurgical Laboratory on Aug. 18, 1942. The first weighing of such a compound occurred on Sept. 10, 1942. This was followed by the preparation and identification of many compounds, the study of the basic chemical and thermodynamic properties in solution, and the testing of the separation process at Hanford concentrations. These investigations were all performed on the microgram scale with cyclotron-produced plutonium by Cunningham and his coworkers, including L. B. Werner, M. Cefola, R. Patton, and C. Smith. Considerable preparative work was later done on the milligram scale with pile-produced plutonium by H. H. Anderson. The program of work on plutonium, as well as on the other transuranium elements, at the Metallurgical Laboratory was under the general direction of G. T. Seaborg. Important ultramicrochemical investigations were also carried on during this period by the group that remained in Berkeley with W. M. Latimer, which included A. C. Wahl, J. W. Hamaker, and G. E. Sheline.

Later, as a result of the operation of the chain-reacting piles at Clinton and Hanford, sufficient plutonium became available to make it possible for a number of groups to investigate its chemical proper-

ties. Using plutonium from these sources, a number of groups have investigated intensively the chemical properties of plutonium. The group at Los Alamos under the direction of J. W. Kennedy and C. S. Smith and including A. C. Wahl, C. S. Garner, and I. B. Johns was principally concerned with plutonium chemistry as applied to plutonium purification. W. M. Latimer and E. D. Eastman at Berkeley with R. E. Connick, J. W. Gofman, L. Brewer, and others contributed notably to the purification problem and to the solution chemistry of plutonium, including the oxidation states, potentials, and reaction kinetics. F. H. Spedding, together with A. F. Voigt and F. Wolters at Iowa State College, contributed to the chemistry of complex-ion formation as well as to other phases of plutonium chemistry. The group of chemists at the Metallurgical Laboratory at the University of Chicago made basic solution and dry chemistry studies and investigated the separation process and purification work. Shortly before the Oak Ridge plant was to start operation, a sizable part of the Metallurgical Laboratory staff, under I. Perlman and S. G. English, together with H. S. Brown, V. R. Cooper, R. W. Stoughton, and others, moved to Oak Ridge, where their principal job concerned further intensive work on the plant separation processes and the actual isolation of plutonium that had been decontaminated (separated from fission-product radioactivities) in the separation plant. The first small amounts of plutonium produced at this site were isolated and purified by L. B. Werner, V. R. Cooper, and B. Fries. A little later K. A. Kraus moved to the Clinton Laboratories and initiated a program of investigation on basic solution chemistry. Remaining in Chicago on basic chemistry and purification problems were the several groups under W. M. Manning, E. F. Orlemann, B. B. Cunningham, and N. R. Davidson and the plutonium separations group directed by S. G. Thompson, J. E. Willard, G. W. Watt, F. W. Albaugh, and J. L. Dreher. Of the latter group, all except Albaugh went to the Hanford plant to continue the program of separations investigation. A great deal of work of fundamental importance to the separations program, such as work on fission products and radiation chemistry and chemical and spectrographic analyses, was done by other groups in the Metallurgical Project; this is described in other volumes of the National Nuclear Energy Series.

In addition to the need for work with pure plutonium in connection with the separations process, it was necessary to determine a number of the physical and chemical properties of the dry salts of plutonium and of plutonium metal. Therefore the study on the ultramicro scale had to encompass this field of investigation also. A number of compounds of plutonium were prepared by reactions involving the

solid and gas phases, that is, by dry chemical reactions. This work was done in collaboration with W. H. Zachariasen of the University of Chicago staff, who was able to use his x-ray technique to identify or help identify a number of the compounds that were synthesized and, in many cases, thus to establish their chemical structure. This preparative work was carried on largely by N. R. Davidson, O. C. Simpson, R. E. Heath, S. Fried, E. F. Westrum, F. Hagemann, J. J. Katz, and coworkers. Another feat that was accomplished with only microgram amounts of plutonium was the preparation of plutonium metal and the study of some of its properties. This was done by P. L. Kirk and H. L. Baumbach and the people working with them.

Much of the work described above is reported by papers in this volume, but much of it is of such a nature that it is not included, and therefore this brief mention of it will have to suffice. Following the successful solution of the production and processing problems at the various sites, investigation of the basic chemical properties of plutonium, usually in larger amounts, has continued in a program involving a large number of investigators. Many of the people who have already been mentioned continued on this work, and a number are still active in it.

Americium and Curium. The work on americium and curium has not been nearly so extensive as that on plutonium or on neptunium. Americium, the element with atomic number 95, was the fourth transuranium element to be discovered, its first identification taking place near the end of 1944 and early in 1945. The first isotope of this element was identified in the experiments of G. T. Seaborg, R. A. James, and L. O. Morgan at the Metallurgical Laboratory. The irradiation of U^{238} with high-energy helium ions in the cyclotron leads to the formation of the β -particle-emitting Pu^{241} , from which the isotope Am^{241} is formed as the daughter.

The availability of this isotope of americium made it possible to study the chemical properties of this element, using the tracer technique. This work was done at the Metallurgical Laboratory, chiefly by S. G. Thompson, R. A. James, L. O. Morgan, and I. Perlman, and later at the Radiation Laboratory in Berkeley by these investigators as well as by L. B. Werner.

Americium has been isolated in the form of pure compounds, a feat which Cunningham accomplished in 1945 and early in 1946 at the Metallurgical Laboratory by work on the ultramicrochemical scale. A great deal of work with macroscopic amounts of americium has been done following its initial isolation. Cunningham and coworkers at the University of California have carried on investigations of its solution chemistry. The preparation of compounds by vacuum-line

techniques has been carried on chiefly by Cunningham and Westrum at the University of California and by Fried at the Argonne National Laboratory.

Curium was the third transuranium element to be discovered. The first isotope of this element was Cm^{242} , which was identified in 1944 by G. T. Seaborg, R. A. James, and A. Ghiorso at the Metallurgical Laboratory as a result of its production in the Berkeley 60-in. cyclotron by the irradiation of Pu^{239} with high-energy helium ions.

The chemical properties of this element were subsequently investigated on a tracer scale by substantially the same group as has been mentioned previously in connection with the tracer-scale investigation of americium. Curium was not isolated in the form of a pure compound until the fall of 1947, when Werner and Perlman, working on the ultramicrochemical scale of operation, succeeded in isolating it at the University of California. The investigation of this element on the macroscopic scale is exceedingly difficult owing to its intense specific α -particle radioactivity.

Americium and curium exhibit chemical behavior very much like that of the rare-earth elements, and therefore a strong analogy with the methods of rare-earth chemistry runs through the program of investigation. Thus the chemistry of americium and curium is in a sense not so complicated as that of plutonium and neptunium with their multiple oxidation states. The names "americium" and "curium" stem from the chemical analogy of these elements with the long-known rare-earth elements europium and gadolinium, respectively. Although the separation from each other and from the rare-earth elements is difficult and has formed the basis for much of the investigation, the papers in this volume will show that up to the present time a relatively greater emphasis has been placed on the radioactive properties of these elements than is the case with plutonium and neptunium.

Very important to all this work was the development of instrumental methods, some of the results of which are reported in this volume. Particularly active in this work were A. Ghiorso at the Metallurgical Laboratory and later at the University of California and A. H. Jaffey at the Argonne National Laboratory.

Some mention should be made of the influence these investigations have had on the basic questions of the electronic structure of the transuranium and other heavy elements and their place in the periodic system. Although previous similar suggestions had been made, the investigation of the chemical properties of neptunium by McMillan and Abelson probably gave the best early evidence that a rare-earth

type of transition group was beginning in this region, and they suggested uranium as the prototypical member. Later consideration of the more detailed chemical properties of neptunium and plutonium led Seaborg to the view that this transition group has the element actinium as prototype, in the same formal sense that the rare-earth group has lanthanum as prototype, but with certain expected differences. This view has been borne out by many subsequent investigations, especially those involving the elements americium and curium; this volume includes papers in which this matter is discussed.

The editors wish to thank the individual contributors for their cooperation. Numerous other persons have done much to make this volume possible. In particular, thanks are due Dr. Hoylande D. Young, Director of the Information Division of the Argonne National Laboratory and General Editor of the Plutonium Project Record; without her unflinching cooperation and guidance this volume very likely would never have appeared. To Dr. Warren C. Johnson and Dr. Willard F. Libby we owe our thanks for their careful reading of the manuscript. Dr. Thomas O. Jones and Dr. Gordon R. Leader were active in the early work of compiling this volume; their contributions are acknowledged with gratitude, and we regret the circumstances that made their further participation impossible. Mrs. Carolyn Baer and Mrs. Eleanor Studier gave notable assistance in the technical aspects of manuscript preparation. Finally thanks are due Brewer F. Boardman and Mrs. Martha Gerrard of the Technical Information Branch at Oak Ridge, Tenn., for their cooperation in preparing this volume for publication.

G. T. Seaborg
J. J. Katz
W. M. Manning

August, 1949

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Paper 1.1a

A NEW ELEMENT:

RADIOACTIVE ELEMENT 94 FROM DEUTERONS ON URANIUM†

By G. T. Seaborg, E. M. McMillan, J. W. Kennedy, and A. C. Wahl

We are writing to report some results obtained in the bombardment of uranium with deuterons in the 60-in. cyclotron.

The uranium was bombarded in the form of U_3O_8 , and the deuterons had to pass through a 2-mil thickness of aluminum foil before hitting the uranium target. The carefully purified element 93 fraction contained a β activity whose aluminum-absorption curve (taken on an ionization chamber connected to an FP-54 tube and also on a Lauritsen electroscope) was distinctly different from the absorption curve of a sample of the 2.3-day 93^{239} (formed from uranium plus neutrons) taken under identical conditions. The upper energy limit of the β particles from this new 93 activity is about 1 mev, compared with about 0.5 mev for 93^{239} . The ratio of γ -ray to β -particle ionization is about five times larger than for 93^{239} . The initial part of the absorption curve of this 93 from uranium plus deuterons is very similar to the initial part of the absorption curve of 93^{239} . Of course the production of 93^{239} is expected in the deuteron bombardment of uranium, from the reaction $U^{238}(d,n)93^{239}$. It is impossible to deduce from the absorption curve the relative intensities of the new 93 and of 93^{239} , since the initial parts of the individual absorption curves of these two activities might well be nearly identical. The rate of decay of the high energy β particles (0.5 to 1 mev) and γ rays from the 93 of uranium

†Contribution from the Department of Chemistry and the Radiation Laboratory, University of California, Berkeley.

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