Carnegie-Mellon University and
United States Energy Research and
Development Administration
Contract No. E(11-1)-3236
Report No. C00-3236-10

NUCLEAR CHEMISTRY AND GEOCHEMISTRY
RESEARCH

CARNEGIE INSTITUTE OF TECHNOLOGY
AND

CARNEGIE-MELLON UNIVERSITY

1950 - 1976

Truman P. Kohman, Principal Investigator

SUMMARY REPORT

Sponsor:

United States Atomic Energy Commission
and

United States Energy Research and Development Administration

CARNEGIE-MELLON UNIVERSITY
Department of Chemistry

MASTER

1976 May 28

Pittsburgh, Pennsylvania 15213

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

NOTICE
This report was prepared as an account of work
sponsored by the United States Government. Neither
the United States nor the United States Energy
Research and Development Administration, nor any of
their employees, nor any of their contractors,
subcontractors, or their employees, make any
warranty, express or implied, or assumes any legal
liability or responsibility for the accuracy, completeness
or usefulness of any information, apparatus, product or
process disclosed, or represents that its use would not
infringe privately owned rights.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

C O N T E N T S

ABSTRACT AND HIGHLIGHTS.....	141
I. INTRODUCTION.....	1
II. PERSONNEL.....	3
A. Supervision.....	3
B. Graduate Students.....	3
C. Postdoctoral Research Associates.....	4
D. Other Research Associates, Research Assistants, and Technicians..	5
E. Undergraduate Students.....	5
F. Supporting Personnel.....	6
III. FACILITIES.....	7
A. Laboratories.....	7
B. Bombardment and Irradiation Facilities.....	8
C. Radioactivity Measurement Instruments.....	8
D. Mössbauer Spectrometers.....	10
E. Mass Spectrometers.....	11
F. Computation Facilities.....	12
G. Other Facilities.....	13
IV. PUBLICATIONS AND REPORTS.....	14
J. Journal and Conference Proceedings Publications.....	14
R. Reviews.....	19
M. Miscellaneous Publications.....	19
D. Doctoral Dissertations.....	19
S. Special Reports.....	20
P. Progress Reports.....	21
V. RESEARCH ACTIVITIES AND RESULTS.....	36
A. High-Energy Nuclear Reactions.....	36
B. Artificial Radionuclides.....	38
C. Primary Natural Radioactivity.....	42
1. Beta Activity.....	42
2. Search for Certain Natural Beta Activities by Active-Daughter Extraction.....	44
3. Alpha Activity.....	45
4. Anomalous Alpha Activity.....	46
D. Extinct Natural Radioactivity.....	47
E. Cosmic-Ray-Induced Radioactivity in Meteorites.....	52
F. Radioactive Dating.....	55
1. Methods Based on Primary Natural Radioactivity.....	56
2. Methods Based on Secondary Natural Radioactivity.....	59
3. Methods Based on Induced Natural Radioactivity.....	61
4. Methods Based on Extinct Natural Radioactivity.....	62
G. Chronology of Nucleosynthesis.....	63
H. Tektite Studies.....	66
I. Neutron-Activation Analysis.....	69
J. Mössbauer Spectroscopy of Natural Materials.....	71
K. Use of Our Facilities by Others.....	74
L. Chemical and Radiochemical Procedures.....	75
M. Instrumental Developments.....	76
N. Miscellaneous Activities.....	79

ABSTRACT AND HIGHLIGHTS

This Report summarizes the activities and results of research in nuclear chemistry, nuclear geochemistry, nuclear cosmochemistry, and some incidental areas carried out at Carnegie Institute of Technology (1950-1967) and Carnegie-Mellon University (1967-1976) with the support of the United States Atomic Energy Commission (1950-1974) and the Energy Research and Development Administration (1975-1976) under the principal supervision of Professor Truman P. Kohman. During leaves-of-absence supervision was assumed by Professors Albert A. Caretto, Jr. (1957-1958) and Julian P. Shedlovsky (1962-1963), and Kohman and Caretto were joint supervisors for two years (1959-1961).

The following individuals obtained Ph.D. degrees in Chemistry with dissertations based on research supported by this Project; unless otherwise indicated, the research supervisor was Professor Kohman: George I. Mulholland (1952), Henry Selig (1954), John W. Jones (1956), Ward S. Diethorn (1956), Robert A. Rightmire (1957), William D. Ehmann (1957), Ronald D. Macfarlane (1959), Ismael Almodovar (1960), David L. Morrison (1961, Caretto), Parmatma S. Goel (1962), John H. Gray (1962, Caretto), James H. Kaye (1963, Kohman and Shedlovsky), Donald J. Reuland (1963, Caretto), William J. Treytl (1963, Caretto), Philip J. Cressy (1964), Tadepalli P. Sarma (1964), and James M. Huey (1972). Other personnel associated with the Project in various capacities are listed.

The facilities available and used are described. The 440-MeV proton synchrocyclotron of the Carnegie Nuclear Research Center was used for much of the research from 1952 until the early 1960's. Low-level radioactivity instrumentation, embodying a number of original developments, was essential to many of the investigations of the early and middle years of the Project. Since the late 1960's a solid-source mass spectrometer has been the central facility for much of the research.

A complete listing is given of the publications, doctoral dissertations, and reports resulting from this research, coded for reference in the narrative section of the Report. The chronological lists provide an overview of the activities at any particular time.

The major part of the Report is a review of the research activities and results, by major categories as indicated by the Table of Contents. The following are some highlights of the accomplishments.

Among the studies of excitation functions for high-energy proton-induced nuclear reactions was a comprehensive study of (p,xp) reactions with $x = 2-5$. The results enabled the relative importance of knock-on cascades and compound-nucleus evaporation as mechanisms to be deduced. The systematics and theory have been found useful in the practical production of neutron-rich nuclides by high-energy accelerators.

The radiochemical recoil technique of studying high-energy reactions was refined so as to yield information on the energy distribution and angular distribution of recoil nuclei. This provided further important contributions to the understanding of the detailed mechanisms involved in the production of specific products in a given interaction.

12

The development of a large low-background internal-sample proportional counter led to the discovery of β^- radiation from the natural radionuclide ^{138}La . Limits were set on the possible natural radioactivity of a number of other nuclides.

Natural alpha activity of ^{144}Nd was discovered by the use of nuclear emulsions and confirmed by measurements with a large internal sample ionization counter. Alpha activity was discovered or confirmed in ^{152}Gd , ^{176}Hf , and ^{190}Pt , and upper limits were established in several cases where activity had been claimed by others.

Searches for unknown long-lived radionuclides culminated in the discovery of ^{26}Al and ^{60}Fe . ^{26}Al provided isotopic tracing possibilities for the last of the chemical elements, and this nuclide has subsequently been found to have a widespread natural occurrence and cosmochemical significance.

^{10}Be , ^{46}Al , and ^{60}Co were detected in meteorites as a result of cosmic-ray bombardment. A number of other radionuclides were subsequently similarly found, of particular significance being ^{14}C . Cosmic-ray induced radioactivity has provided much information about the histories of meteorites before their fall on Earth, and has indicated terrestrial ages of some finds.

Cosmic-ray irradiation of meteorites was simulated by proton-beam bombardment of thick targets at accelerators, with radiochemical determination of product distributions. Mathematical methods were developed for systematizing the data and applying them in the calculation of production rates in solid bodies exposed to cosmic radiation in space.

Improvements were made in the chemical and instrumental techniques of radio-carbon dating and of protactinium-ionium dating of ocean sediments. The mixing of top sediment layers by submarine currents and animal activity was modelled mathematically as a diffusion-like process, and it was shown how this "pseudo-diffusion" affects the apparent sedimentation rates and sediment ages for any method involving the sedimentation of a short-lived radionuclide.

Measurements of the isotopic composition of lead from chondrites led to an improved ^{207}Pb - ^{206}Pb isochron for this class of meteorites, the corresponding mean age being 4.505 ± 0.008 Gy. Individual chondrites, however, had distinctly different single-stage-model ages, with a range of ~ 50 My about the mean. The model ages were found to be negatively correlated with metamorphic grade, implying that parent-body heating caused both volatile-element loss and metamorphism in chondrites. An achondrite was observed to be ~ 50 My older than the chondrite mean.

Searches for cases of extinct natural radioactivity with non-gaseous decay products have not yet revealed any, but it was established that the early Solar-System abundance of ^{205}Pb was less than 10^{-4} times that of ^{204}Pb .

From the abundances in meteorites of the extinct nuclides ^{129}I and ^{244}Pu and the extant nuclides ^{127}I , ^{232}Th , ^{235}U , and ^{238}U , deductions were made about the chronology of heavy-element nucleosynthesis in the Galaxy prior to the formation of the Solar System. A terminal "spike" due to one or more local supernovae was shown to be likely. These calculations were supplemented and extended to the present by consideration of heavy-element abundances in stars of known ages.

A theory of proportional-counter behavior yielded a successful formula for the gas amplification in cylindrical counters of arbitrary dimensions, pressure, and applied voltage, having two parameters characteristic of the gas. These physically significant parameters can be evaluated from a few measurements on a counter filled with the particular gas.

Mössbauer spectroscopy of iron in coal revealed a form of Fe^{II} with a unique Mössbauer spectrum. It was at first thought that this could be due to organically bound iron, but subsequent work showed that it was inorganic, almost certainly in a phyllosilicate, and probably in a mineral of the chlorite group. A mineral different from any hitherto characterized may be involved.

Our facilities were frequently made available to a number of other researchers at the University. Most significant was a thorough study of the isotopic composition of lead in works of art, particularly paintings. Correlations with the isotopic composition of commercial lead of various times provided significant information on the authenticity of certain paintings.

Among developments not pertaining directly to objectives of the Project, but made possible by the enlightened AEC support, was the formulation of a universal system of magnitudes for astronomical objects based on power fluxes. This facilitates meaningful intercomparisons of power fluxes and magnitudes in all parts of the electromagnetic spectrum from radio to gamma radiation.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY
RESEARCH

CARNEGIE INSTITUTE OF TECHNOLOGY
AND

CARNEGIE-MELLON UNIVERSITY

1950 - 1976

SUMMARY REPORT

Truman P. Kohman, Principal Investigator

I. INTRODUCTION

Instruction and research in nuclear chemistry at Carnegie Institute of Technology began in 1948, when the writer was brought into the Chemistry Department by Professor J. C. Warner, then Head of the Department and soon thereafter (1950-1963) President of the Institute, which was later (1967) to become Carnegie-Mellon University. At that time the Physics Department was developing the Carnegie Nuclear Research Center at Saxonburg, Pennsylvania, where a 140-inch 440-MeV synchrocyclotron was nearing completion under the direction of Professor Edward C. Creutz. A "hot" cell for high-level radiochemical work at Saxonburg was being planned, and other laboratory space was designated for nuclear chemistry research at the Center and in Engineering Hall (later named Doherty Hall) on the campus. Courses in nuclear chemistry and radiochemical techniques were inaugurated by the writer.

From the beginning of support by the Atomic Energy Commission in 1950 May the nuclear chemistry research program involved two broad areas, (1) induced nuclear reactions and artificial radioactivity and (2) nuclear phenomena in nature and the use of nuclear techniques in the study of natural systems. There was considerable overlap and interaction, however, as in the search for and characterization of radionuclides which might be observable in nature, and in the laboratory simulation of cosmic-ray-induced nuclear reactions. This was recognized somewhat belatedly in 1963 by the change in title of the Project from "Nuclear Chemistry Research" to "Nuclear Chemistry and Geochemistry Research."

In 1957-1958, while the writer was on a sabbatical leave, the supervision of this research program and teaching in the area were taken over by Albert A. Caretto, Jr., on a one-year faculty appointment. Professor Caretto returned as a regular faculty member in the spring of 1960. The Project was under the joint supervision of Caretto and the writer until the fall of 1961, when he began receiving separate support from the AEC. Professor Caretto's research was and is strongly oriented towards high-energy nuclear reactions, and from 1961 on this Project became more strongly oriented towards geochemistry including cosmochemistry.

During another sabbatical leave of the writer, in 1962-1963, the Project was supervised by Julian P. Shadlovsky, formerly a postdoctoral research associate, on a one-year faculty appointment.

A further permanent increase in the nuclear chemistry staff of the Chemistry Department was made possible in 1968 by a Science Development grant by the National Science Foundation. Paul J. Karol and Morton Kaplan joined the Chemistry Department faculty in 1969 and 1970, respectively. Professor Karol became a Co-Investigator with this Project from 1970 to 1973. Professor Kaplan has been receiving support from the AEC since his arrival, in continuation of that he received previously at Yale. Thus, largely because of support by the Commission and its successor, the Energy Research and Development Administration, Carnegie-Mellon University has become a leading center of research and instruction in nuclear chemistry.

During the period of AEC support the writer has received additional research support for related activities in the geochemistry and cosmochemistry area from the Petroleum Research Fund administered by the American Chemical Society (1957-1960 and 1962-1965, for work on dating of marine sediments), the Division of Biology and Medicine of the AEC (1965-1967, for work on natural ^{129}I principally by Dr. Raymond R. Edwards), and by the National Aeronautical and Space Administration (1968-1970, for work on lunar samples).

In 1964 Carnegie Institute of Technology, under the farsighted leadership of President Warner, inaugurated an Interdisciplinary Program in Earth and Astronomical Sciences, with the writer as Program Director. Among the new faculty members brought to Carnegie in 1965 under this Program was Ronald G. Ostic as Assistant Professor of Geophysics. Ostic became informally associated with this Project during the academic years and formally as a research associate during the summers. His presence and efforts were important factors in the awards of a sizeable grant from the National Science Foundation towards the acquisition of a solid-source isotope-ratio mass spectrometer and of the contract from NASA for radiogenic isotope studies of lunar materials. Unfortunately, following President Warner's retirement in 1965, the Interdisciplinary Program in Earth and Astronomical Sciences was abruptly terminated in 1967 in spite of its successful development of instruction in the earth sciences, attraction of a number of new graduate students, and obtaining of research and fellowship-trainee support from the NSF, NASA, and NDEA. Professor Ostic left in 1968, and the NASA support was terminated in 1970. The present AEC Project has, however, continued to benefit from the excellent mass spectrometer which was acquired under that Program and installed by Professor Ostic.

In this Report, following summaries of personnel associated with this Project and facilities used by it, is a coded listing of publications and reports, and then a narrative summary of the research activities and results obtained with the AEC-ERDA support.

II. PERSONNEL

A. Supervision

Throughout the period of this project except for two sabbatical leaves, the writer has been the Principal Investigator (to use current terminology). As explained in the Introduction, Professors Albert A. Caretto, Jr., and Julian P. Shedlovsky assumed active supervision during the 1957-1958 and 1962-1963 academic years, respectively. Professors Caretto and Paul J. Karol were Co-Investigators during the periods 1960-1961 and 1970-1973, respectively.

B. Graduate Students

The largest contribution of any group to the research activity and results of this Project was that of the graduate students who completed Ph.D. theses with the writer and Professor Caretto. The commitment to completion of the degree requirements, the interest which intense research involvement generates, and the imposition of academic standards all contribute to the quantity and quality of their output.

Many graduate students had a shorter involvement with the Project for one reason or another. Even when their work did not lead to publishable results their support was justified by the training in research which they secured from this association, which in many cases was of value in their attainment of the Ph.D. with some other supervisor or even in some other area of chemistry. Most of them received thoroughgoing and effective tutelage in scientific writing, especially after the adoption of the present form of annual Progress Report: the 1959-1960 and subsequent Reports are composed mainly of individually authored contributions by the research workers themselves.

1. Graduate Students who Attained Ph.D. Degrees with Professor Kohman

George I. Mulholland: 1950-1952
 Henry Salig: 1950-1954
 John W. Jones: 1950-1956
 Ward S. Diethorn: 1952-1956
 Robert A. Rightmire: 1954-1957
 William D. Ehmann: 1954-1957
 Ronald D. Macfarlane: 1955-1959
 Ismael Almódovar: 1956-1960
 Parmatma S. Goel: 1958-1962
 James H. Kaye: 1959-1963
 Philip J. Cressy: 1959-1964
 Tadepalli P. Sarma: 1960-1964
 James M. Huey: 1966-1972

2. Graduate Student Still Working Toward Ph.D. Degree with Professor Kohman

Chia-Cheng Lin: 1973-

3. Graduate Students Who Attained Ph.D. Degrees with Other Faculty of the Department

L. Guy Donaruma: 1951-1952 (1953, Southwick)
 Peter J. Lykos: 1952 Summer (1954, Parr)

Hubert W. Joy: 1953 Summer (1957, Parr)
 David L. Morrison: 1958-1961 (1961, Caretto)
 John H. Gray: 1958-1961 (1962, Caretto)
 Donald J. Reuland: 1960-1961 (1963, Caretto)
 William J. Treytl: 1960-1961 (1963, Caretto)
 Pauline M. Zwickowitz: 1960-1961 (1964, Rubin)
 F. Michael Kiely: 1963 Spring (1967, Caretto)
 Narayanasamy Viswanathan: 1963-1966 (1968, Van Dyke)
 Roy F. Schall: 1963-1966 (1968, Caretto)
 Agnes Tze-Wen Kuo: 1964-1966 (1970, Kurland)
 Edgar A. Paez: 1966-1969 (1971, Weaver)
 Muppalia Rama Rao: 1967 (1972, Bothner-By)
 Thomas J. Thomas: 1968-1969 (1975, Carlin)

4. Graduate Students Who Left CIT-CMU Without Attaining Ph.D. Degrees

Esther Waldron: 1951-1953
 David H. Freeman: 1953-1954 (Ph.D. 1957, Massachusetts Institute of Technology)
 Alton L. Long: 1953-1954
 Victor C. Solomon: 1953-1956
 F. William Walker: 1957-1960
 Sachiko Nakagawa: 1957-1959
 Hans Hamster: 1958-1960
 B. Diane Wassil: 1961 Spring and Summer
 Andrea F. (née Henry) Hough: 1962-1964
 Aziza Ragai: 1964-1965
 Shrinivas Iyer: 1965 Spring
 Samir Sarkar: 1965 Summer
 James H. Luetgert: 1966 Summer, 1967 Summer
 James R. Noyce: 1967-1968 (Ph.D. 1972, University of Arkansas)
 Vassiliki Tsomi: 1967-1968
 Ramzi Y. Saleh: 1968-1970 (Ph.D. 1975, University of Pittsburgh)
 Nancy Man-Na (née Kuo) Kan: 1969-1971

C. Postdoctoral Research Associates

This list does not include graduate students who remained for a few weeks or months after receiving their Ph.D. degrees, preparing their work for publication, etc.

Professor Thomas R. Alexander, Associate Professor of Chemistry, Carnegie Institute of Technology: 1953-1954 (part-time).
 Professor Nobufuso Saito, Assistant Professor of Chemistry, University of Tokyo: 1952-1954.
 Jean-Claude Roy, from University of Notre Dame, Notre Dame, Indiana: 1954-1955.
 Dr. Hamed M. El-Badry, Senior Lecturer, Cairo University: 1955-1957.
 Dr. Erich Merz, from Max-Planck Institut für Chemie, Mainz, Germany: 1958-1960.
 Dr. Madan W. Chakrabarty, Lecturer in Inorganic Chemistry, Jadavpur University, Calcutta: 1959-1961.
 Dr. Julian P. Shedlovsky, from Gulf Research & Development Laboratories, Harnarville, Pennsylvania: 1960-1962 (see II.A.).
 Dr. Garimella V. S. Rayudu, from McGill University, Montreal: 1961-1964.
 Dr. Shiv Nath Tandon, from Indian Institute of Technology, Kanpur, India: 1964-1966

Dr. Charles F. Beck, Jr., Instructor of Chemistry, Mount Lebanon High School, Mount Lebanon, Pennsylvania, 1965, Summer.

Dr. Raymond R. Edwards, from Nuclear Science and Engineering Corporation, Pittsburgh: 1965 Summer and Fall.

Dr. John F. Lefelhocz, from University of Notre Dame, Notre Dame, Indiana: 1966-1967.

Dr. James F. Tanner, from University of Kentucky, Lexington: 1966-1968.

Professor Ronald G. Ostic, from University of California at San Diego, La Jolla; Assistant Professor of Geophysics at CIT-CMU, 1965-1968: Summers of 1965, 1966, 1967 (see Introduction).

Dr. Brij M. P. Trivedi, from Indian Institute of Technology, Kanpur, India: 1969-1971.

Dr. Brij M. Bansal, from University of California at Berkeley and NASA-supported lunar samples program at CMU: 1971 May-July.

Dr. Michael S. Lancet, from University of Chicago: 1972-1974.

D. Other Research Associates, Research Assistants, and Technicians

1. Special Research Associate

Peter Strohail, Institute Rudjer Bosković, Zagreb, Yugoslavia; visiting scientist under auspices of U.S. International Cooperation Administration and Government of Yugoslavia: 1958-1960.

2. Instrumentation and Electronics

John E. Thomas: 1950-1951

James R. Simanton: 1952-1956

Franc Mesojedec (changed to Meno): 1957-1964

Michael J. Toia: 1964-1969

Mark W. Harsenic: 1969-1972

3. Chemistry

Doris D. Jones: 1950-1951

Esther C. Waldron: 1951 Summer

John W. Cooper: 1951-1952

Lou Ann Stone: 1957-1958

Heidi B. Fenton: 1962

Geoffrey W. White: 1963

Ronald D. Allerton: 1966 Summer

Elizabeth Y. Fang: 1966-1968

Maria I. C. de Carvalho: 1970

E. Undergraduate Students

Throughout the period of this Project, undergraduate students have been associated with the research work in a variety of ways (part-time employees during the academic year, full-time summer employees, academic-credit research participants, and others) and in a variety of activities (chemical or instrumental laboratory assistance, drafting, computation, and semi-independent research participation). The following list is incomplete and does not distinguish between categories of participation. For the period 1950-1959, only those who

participated to a substantial degree during an academic year or full-time during a summer as a minimum are listed. For the period 1960-1974, only those who made written contributions to the annual Progress Reports are included. Students who were employed immediately after their graduation are also included here.

For many of those students, affiliation with this Project was a worthwhile educational and work experience, and for some of them a valuable introduction to scientific research and writing.

L. Leroy Lewis: 1950	Michael L. Bender: 1964-1965
Robert W. McKelvey: 1950	Sanford J. Catz: 1964-1965
Donald G. Schindler: 1951-1952	Frederick J. Van Itallie: 1965-1967
R. Joseph Ransil: 1951-1952	Richard G. Sextro: 1965-1966
John W. Cooper: 1951-1952	Harold J. Konish: 1969
Virginia A. Schultz: 1952-1955	Mark J. Yeager: 1969-1971
James I. Nixon: 1953-1955	Nina A. Landes: 1969-1970
Gaile R. Ventresca: 1956	James D. Ulmer: 1970
Philip M. Tischler: 1956-1957	John A. Yokum, Jr.: 1970
Glenn I. Henrickson: 1956-1957	John F. McNahan: 1970
Michael J. Toia: 1957-1959	Charles A. Ritter: 1970-1972
Thomas A. Becker: 1957-1959	Peter Kamarchik: 1971-1972
Jane S. Potter: 1958-1961	Stanley B. Nachimson: 1972-1973
Edward A. Ondovchik: 1959	Robert A. Witter: 1973-1974
Mark I. Appelbaum: 1961	Daniel L. Menis: 1973-1974
Martha R. Miller: 1962-1963	D. Lynn Svec: 1973-1974

In addition to these CIT/CND students, Paulette L. Rohman, an undergraduate at Wesleyan University, Middletown, Connecticut, was a volunteer research worker during the summer of 1972.

F. Supporting Personnel

Persons employed or supported by the Project who performed essential services while not actually participating in the research included health physicists, machinists, glassblowers, general laborers, secretaries, stenographers, and clerks.

The secretaries who were most closely involved in the handling of technical correspondence and typing of technical reports and papers were:

Sylvia Leggett: 1953-1954	Gail Chasey: 1969-1970
Blanche Murphy: 1954-1963	Judith Watt: 1970-1972
Dorothy Ponsetto: 1961; 1976	Anne Spudich: 1972-1973
Anneli Ebner: 1964	Charlotta Thunander: 1973
Janet Muder: 1964-1969	Lois Sebatane: 1973-1974
June Johnson: 1964-1967	Bernetta Spector: 1974-1976

III. FACILITIES

A. Laboratories

At the initiation of this work in 1950 the Chemistry Department was housed in Engineering and Science Hall (later named Doherty Hall). Expansion of the department and its research activities necessitated conversion of attic space into laboratories, and two rooms were assigned to the writer, one as a chemistry laboratory and one as a combined instrument laboratory and office.

In 1950 a new wing was added to this building, and the research group supported by this contract was assigned laboratory and office space on the third floor. This space was expanded from time to time, partly by conversion of hall space to a combined secretarial office and library. A radiochemistry laboratory and counting room on the second floor used mainly for instruction was also used for some research. These laboratories are described with illustrations in the 1959-1960 Progress Report.

In 1963 a large basement-level room was acquired and air-conditioned for instruments, especially low-background counters with massive shielding. This is illustrated in the 1962-1963 Progress Report. When the mass spectrometer was acquired in 1966 it was put into this room.

The "hot" cell at the Nuclear Research Center, which was designed by the writer, was completed in 1952. Ordinary laboratory space was also made available for target preparation, counting, and "warm" work with the cyclotron. The Nuclear Research Center and some of the radiochemical work space is illustrated in the 1959-1960 Progress Report. These laboratories were used for work supported by this project until 1963.

Following the formation of Carnegie-Mellon University in 1967, the research activities and facilities of the Chemistry Department were moved from Doherty Hall on the main campus to the Mellon Institute Building. In 1970 the writer made this move. Nearly ideal space was provided, rather extensive remodeling being required for some of it. The following description, which is still applicable, is taken from the 1970-1971 Progress Report:

The space, designated as the Nuclear Geochemistry Laboratory, is comprised of the following units:

- 519: Entrance, wall displays, natural materials collection.
- 521: Small instrument room, temporarily used for storage.
- 523: Electronics and instrument workshop.
- 525: Postdoctoral associates' office.
- 527: Professor's office.
- 529: Secretary's office and files.
- 531: Library and conference room.
- 533: Clean laboratory, within which are a walk-in tunnel with HEPA (High-Efficiency Particulate-Air) filters and horizontal laminar air flow and a fume hood with HEPA filters and vertical laminar air flow.
- 535: Chemical laboratory, with four benches and student desks.
- 537: Chemical laboratory, intended for intermediate-level radiochemical operations.
- 539: Radiochemistry laboratory, intended for moderate-level radiochemical operations and storage of radioactive materials.

116-B: Instrument laboratory, partitioned by five-foot-high walls into three sections with separate doors, housing the following:

West: Alpha, beta, and gamma counting equipment, Mössbauer spectrometer.

Center: Mass spectrometer, source preparation facilities, remote computer station teletype.

East: Low-level counting instruments.

All of the space is air-conditioned, and the clean laboratory is provided with pre-filtered and electrostatically cleaned air in addition to the HEPA filters.

B. Bombardment and Irradiation Facilities

The Carnegie proton synchrocyclotron at the Nuclear Research Center was used on a number of occasions at energies from 45 to 440 MeV.

In some studies bombardments were made with the University of Pittsburgh cyclotron (15-MeV deuterons), the McGill University synchrocyclotron (450-MeV protons), the Brookhaven National Laboratory Cosmotron, (protons of 0.5, 0.8, 1.5, 2.2, 2.9, and 3.0 GeV), the University of California at Berkeley Bevatron (6.1-MeV protons), and the Oak Ridge National Laboratory cyclotron (21-MeV protons).

Neutron irradiations were made at the Canadian Chalk River NEX reactor, the Argonne National Laboratory CP-5 heavy-water-moderated reactor, and the Nuclear Engineering Test Facility at Wright-Patterson Air Force Base, near Dayton, Ohio.

C. Radioactivity Measurement Instruments

1. Counters and Spectrometers

Throughout the period of this research a number of standard beta counters were maintained, for some time in connection with the course in Radiochemical Techniques. The original sealed mica-window Geiger counters with binary scalers were later replaced with aluminized-mylar-window methane-flow proportional counters with decade scalers and automatic timers.

An internal-sample 4π Geiger counter was built and used in the characterization of ^{26}Al in 1966-1967.

Anthracene and plastic scintillators were used occasionally for beta spectrometry, either alone or in coincidence assemblies.

Rather early in this period NaI(Tl) scintillation detectors came into use for gamma measurement and spectrometry, and a number of these were acquired and used, singly and in coincidence assemblies. In 1967-1968 a 4-inch x 4-inch well-type detector was acquired and is still in operating condition.

In 1968-1969 a 30-cm^3 Ge(Li) gamma detector was acquired and incorporated into a gamma spectrometer. This is still in frequent use.

A Frisch-grid alpha ionization chamber was constructed in 1957, and a commercial chamber of this type was acquired in 1959-1960. Semi-conductor alpha detectors replaced these beginning in 1961-1962, and two are maintained in operating condition.

The first multichannel pulse analyzer used was a 48-channel electromechanical analyzer purchased in 1950 from the University of Chicago, where it had been used by the writer. This was converted to a faster and more reliable 24-channel analyzer in 1955 and used for a number of purposes for several years. A single-channel analyzer, a "gray-wedge" analyzer, and a scanning single-channel analyzer were also used for a while. The first electronic multichannel analyzer (256 channels) was acquired in 1959-1960. At present we have two 256-channel analyzers, one 128-channel analyzer, and one 512-channel analyzer, all made by Nuclear Data.

2. Low-Level Counters

Much of the work of this Project had to do with very weak natural radioactivities of various types, and a number of low-level counting systems were acquired, mostly by building or assembling them ourselves.

In the first year a large proportional counter for internal solid samples of 1500 cm² area was constructed and mounted with a surrounding sheath of Geiger counters in anticoincidence in a heavy shield of lead bricks. The output was recorded by the 48-channel analyzer with anticoincidence control. The shielding was later improved by surrounding the counter by a 1-inch mercury cylinder and adding a 2-inch iron liner to the pile of lead bricks. This was used for beta-particle and X-ray detection and spectrometry in samples of low specific activity. The counter was converted to an ionization alpha counter in 1956-1957 by enlarging the counter wire and increasing the external amplification. It was later reconverted to a proportional counter for X-ray measurements.

For low levels of beta activity in samples of small mass, special small Geiger counters were designed and built. The most successful were made of lucite with a side window of aluminized mylar. Four of these were operated inside an anticoincidence-counter sheath, all inside a massive shell of steel bricks. For samples of intermediate specific activity somewhat larger cylindrical gold-foil-wall counters were built and mounted in a similar assembly. By using a high-argon counting gas, these could be used for X-ray-emitting samples. These systems have not been reassembled since their dismantling for the move to the Mellon Institute Building.

A commercially built (Sharp Laboratories) Low-Beta unit with two circular-window Geiger counters in a common anticoincidence-copper-lead shield was acquired about 1961. It is still functional.

A system for measuring natural ¹⁴C for radiocarbon dating was built around a one-liter methane proportional counter, which was operated at up to 5 atmospheres pressure and could probably be operated at up to 20 atmospheres. Several smaller gas proportional counters operating with highly purified CO₂ were used for the measurement of cosmic-ray-produced ¹⁴C in meteorites.

Large (50-liter) and small (2 cm³) gas X-ray counters were built and partially tested in low-background assemblies, but were never put into active service.

Several coincidence-counting assemblies described in the next subsection were also used for low levels of radioactivity, since coincidence and energy selection eliminates much of the background due to cosmic radiation and radioactive contamination.

3. Coincidence-Counting Assemblies

For absolute standardization of β - γ emitters and for disintegration-scheme studies, a 1 3/4-inch x 1/2-inch anthracene scintillator and a 3-inch x 3-inch NaI(Tl) scintillator were used in a coincidence assembly.

A γ - γ coincidence assembly of two 2-inch x 2-inch NaI(Tl) crystals in a heavy shield was used for low-level samples of γ - γ , β^+ , and γ - β^+ emitters. This was replaced in 1964 by a system with two 3-inch x 3-inch NaI(Tl) crystals, in a large cylindrical plastic scintillator viewed by six photomultipliers in anticoincidence, all inside a massive shield of iron and lead bricks. A versatile circuit provides for a variety of coincidence and anticoincidence counting modes, and the recording circuitry includes a dedicated 128-channel pulse analyzer. This was reassembled, tested, and calibrated after the move to the Mellon Institute Building, and is still serviceable.

In 1966-1967, with the assistance of the AEC Division of Biology and Medicine contract for the study of natural and environmental ^{129}I , an assembly of a 4 π gas-flow beta proportional counter and two 4-inch x 4-inch NaI(Tl) scintillators inside a massive shield was constructed for the measurement of neutron-induced ^{130}I . This was calibrated for ^{130}I and tested for discrimination against interfering ^{126}I . However, following termination of that contract, this was disassembled and shipped to Dr. Edward's new location (Chapman College, Orange, California). It is not known whether it was ever reassembled and used.

Another γ - γ system consisting of two 2-inch by 2-inch NaI(Tl) scintillators with adjustable spacing, a fast coincidence circuit, and a versatile logic system was constructed in 1968, for use principally by Dr. Tanner. This has been idle since his disassociation from this Project later that year.

4. Miscellaneous

Several portable survey meters were used, and one portable Geiger counter is still in use.

A recording scaler, in which pulses from the different digits of a decimal scaler produced pulses of different lengths on a paper-chart recorder, was built in 1959-1960 and used for a while for unattended recording of decay curves, etc.

A magnetic positron-electron separator was built in 1959-1960 and used in disintegration-scheme studies.

A system for measuring the energy and time delay of microsecond-millisecond alpha particles produced in pulsed synchrocyclotron bombardments was designed and partially built and tested in 1961-1963, but not completed and made functional.

D. Mössbauer Spectrometers

For initial Mössbauer experiments in 1963-1964, a spectrometer of the Physics Department was used. Again in 1968-1969 a Physics Department Mössbauer spectrometer was used.

In 1964 a constant-velocity mechanical drive (Model B) was acquired from the Nuclear Science and Engineering Corporation. This was modified and equipped with a laboratory-built control and recording system using some obsolete electronic and mechanical components in 1965-1966. Although this did yield publishable results, inherent limitations and frequent service requirements led to its retirement in 1968.

In 1968-1969 a complete system with a constant-acceleration drive (Model NS-1) was acquired from the International Chemical and Nuclear Corporation by the Chemistry Department through a National Science Foundation Science Development grant. This presented substantial deficiencies which could not and would not be remedied by the supplier. They were finally remedied in our own laboratory in 1971, and the system was used until 1973, when the proportional counter was damaged as a result of a power interruption.

Only ^{57}Fe studies were carried out with these instruments, using several ^{57}Co sources, all of which are now too weak for use.

The system can be reactivated by repair or replacement of the detector and supply of an appropriate radiation source or sources. A liquid-helium cryostat and a 1.2-kG electromagnet are also available for varying the absorber temperature and subjecting it to a variable magnetic field.

E. Mass Spectrometers

In collaboration with Professor Joseph I. Lipson of the Earth and Planetary Science Department of the University of Pittsburgh, an attempt was made in 1963-1964 to reconstruct and up-date an Army surplus General Electric 6-inch-radius mass spectrometer as a thermal-emission solid-source isotope-ratio instrument. Although minimal operation was achieved, this project was abandoned when Professor Lipson left the EPS Department at Pitt and the CIT graduate student involved abandoned her studies.

As mentioned in the Introduction, an excellent commercially built mass spectrometer (Nuclide Corporation, SU-2.4, 12-inch-radius, 90° -deflection) was acquired in 1966 under the Interdisciplinary Program in Earth and Astronomical Sciences with the assistance of a grant from the National Science Foundation. This was installed under and made operational by Professor Ronald G. Ostic. It has two interchangeable source assemblies with receptacles for a center filament and two side filaments. The detector has an optional ion-electron multiplier and a vibrating reed electrometer. The original equipment included an expanded-scale chart recorder and a 12-position array of potentiometers for stepped variation of the accelerating potential with a timer-controlled cycling switch.

Under the NASA-sponsored program for lunar samples research, this was modified in 1969-1970 for digital output by provision of a digital voltmeter consisting of a Hewlett-Packard 2212A voltage-to-frequency converter, a Monsanto 1510A counter-timer, and a Hewlett-Packard 5050B digital printing recorder. The peak-selecting arrangement was modified to permit manual switching between two values of the magnet current with constant accelerating potential. An optical pyrometer (Pyrometer Instrument Co., Model 95) was provided for monitoring and controlling the filament temperature.

In 1971-1972 the timer-driven peak-selecting arrangement was modified for automatic cyclic switching between up to twelve values of the magnet current. In 1972-1973 a new set of high-current magnet coils with suitable power supply, a Hall-effect magnet-field sensor, and a field-control system for the magnet current was installed by the Nuclide Corporation (MR-13/MS-13 system). The cyclic control was modified for automatic cycling between pre-set magnet-field values selected to center on flat-topped peak positions and at background positions. The peak switching and field stabilization now requires a small rather than a large fraction of the time for a run, and cycling among all peaks of an element is much more time-efficient than measuring only one ratio between two peaks at a time.

In 1973-1974 the recording system was modified by adding an electronic interface (DATAcap, Inc., Model D102 Coupler with 33 ASR Teletype Driver) between the counter-timer and the teletypewriter which had been used for manually transmitting the printed output to the Computation Center for processing. A level-matching circuit for 33 signal channels had to be constructed. During operation of the mass spectrometer, the output is punched on a paper tape by the teletypewriter while off-line from the computer. Initially, the tape was fed back to the teletypewriter when on-line with the computer for processing. In 1975 a separate Teletype 33 was attached permanently to the mass spectrometer for dedicated use with it.

In 1974 a switching device was added to facilitate the location of the magnet field corresponding to the center of each peak.

The mass spectrometer was moved from the main campus to the present location in 1970 with little difficulty in realignment and start-up. It is fully operational at present.

F. Computation Facilities

The CIT-CMU Computation Center has been developing throughout the lifetime of this Project, and from the start has been one of the best university computation facilities. For the past few years the principal systems have been a UNIVAC 1108 and an IBM 360/67 time-sharing system with many remote terminals.

A number of participants in this research, including the writer, have made frequent use of the available facilities. Through about 1970 this was done mostly by batch jobs with punched cards, with each user providing his own "courier" service, although some use was made of "public" remote terminals.

In the 1970-1971 year, following the move to the Mellon Institute Building, a Dataphone and Teletype 33 were installed adjacent to the mass spectrometer. This has been used extensively for access to the 360/TSS both for mass-spectrometer data reduction and for other computations.

In 1974 a modification was made to the mass spectrometer allowing it to transmit its output to the same teletypewriter in order to record the data on punched paper tapes, the teletypewriter being switched between the mass spectrometer and the Dataphone. In 1975 a separate teletypewriter was provided for the mass spectrometer, so the Dataphone-teletypewriter combination is now always available for remote computations.

The UNIVAC 1108 is still used for large batch jobs, since it is somewhat faster, has a larger memory, and provides outputs via line printer and/or CALCOMP plotter.

G. Other Facilities

A variety of facilities for general laboratory and chemical operations were provided by the Chemistry Department or acquired with AEC funds: balances, ovens, furnaces, pH meters, centrifuges, vacuum pumps and gauges, desk calculators, microscopes, etc.

A variety of electronic test and service instruments were also provided: voltmeters, multimeters, pulse generators, tube and transistor testers, oscilloscopes, oscilloscope cameras, etc.

A lead chest was constructed for storage of radioactive materials. A large supply of lead and iron bricks is available for radiochemical work and counter shields.

IV. PUBLICATIONS AND REPORTS

J. Journal and Conference Proceedings Publications

- J52a POSSIBLE NATURAL RADIOACTIVITY OF NEODYMIUM.
G. I. Mulholland and T. P. Kohman.
Phys. Rev. 85, 144-145 (1952).
- J52b THE PAIRING EFFECT IN NUCLEI AND THE BETA-LABILE ELEMENTS.
T. P. Kohman.
Phys. Rev. 85, 530-531 (1952).
- J52c SEARCH FOR NATURAL RADIOACTIVITY OF CALCIUM 48.
J. W. Jones and T. P. Kohman.
Phys. Rev. 85, 941 (1952).
- J52d NATURAL RADIOACTIVITY OF LANTHANUM 138.
G. I. Mulholland and T. P. Kohman.
Phys. Rev. 87, 681-682 (1952).
- J53a SYNCHROCYCLOTRON PRODUCTION AND PROPERTIES OF MAGNESIUM 26.
J. W. Jones and T. P. Kohman.
Phys. Rev. 90, 495-496 (1953).
- J54a NATURAL ALPHA ACTIVITY OF NEODYMIUM.
E. C. Waldron, V. A. Schultz, and T. P. Kohman.
Phys. Rev. 93, 254-255 (1954).
- J54b GEOCHRONOLOGICAL SIGNIFICANCE OF EXTINCT NATURAL RADIOACTIVITY.
T. P. Kohman
Science 119, 815-852 (1954).
- J54c POSSIBLE GEOLOGICAL SIGNIFICANCE OF BOUND BETA DECAY.
T. P. Kohman.
Proc. Conf. on Nuclear Processes in Geologic Settings, Williams Bay, Wisconsin, 1953 September, 10-13 (1954).
- J54d SEARCH FOR NEW NATURAL RADIOACTIVITIES.
T. P. Kohman.
Proc. Conf. on Nuclear Processes in Geologic Settings, Williams Bay, Wisconsin, 1953 September, 14-16 (1954).
- J54e EXTINCT NATURAL RADIOACTIVITY: AN EXPERIMENTAL APPROACH.
T. P. Kohman.
Proc. Conf. on Nuclear Processes in Geologic Settings, Williams Bay, Wisconsin, 1953 September, 18-20 (1954).
- J54f LONG-LIVED RADIOACTIVE ALUMINUM 26.
J. R. Simanton, R. A. Rightmire, A. L. Long and T. P. Kohman.
Phys. Rev. 96, 1711-1712 (1954).
- J57a IRON 60.
J.-C. Roy and T. P. Kohman.
Canadian J. Phys. 35, 649-655 (1957).

- J57b PRODUCTION OF CARRIER-FREE ALUMINUM-26 AND SODIUM-22.
R. A. Rightmire, T. P. Kohman, and A. J. Allen.
Journal of Applied Radiation and Isotopes, 2, 274-279 (1957).
- J58a ARE TEKTITES EXTRA-SOLAR-SYSTEM METEORITES?
T. P. Kohman.
Nature 182, 252-253 (1958).
- J58b ALUMINUM 26: PROPERTIES, PRODUCTION, ASSAY, NATURAL OCCURRENCE, AND POTENTIAL USEFULNESS.
T. P. Kohman, R. A. Rightmire, W. D. Ehmann, and J. R. Simanton.
Proceedings of the International Conference on Radioisotopes in Scientific Research (UNESCO, Paris, 1957 September), Volume I, 1-18(1958).
- J58c COSMIC-RAY-INDUCED RADIOACTIVITY IN METEORITES AND TEKTITES.
T. P. Kohman and W. D. Ehmann.
Proceedings of the International Conference on Radioisotopes in Scientific Research (UNESCO, Paris, 1957 September), Volume II, 661-680 (1958).
- J58d ÜBER DIE HALBWERTSZEIT DES LANGLEBIGEN ^{26}Al .
R. A. Rightmire, T. P. Kohman, and H. Hintenberger.
Z. Naturforsch. 13a, 847-853 (1958).
- J58e COSMIC-RAY-INDUCED RADIOACTIVITIES IN METEORITES - I: CHEMICAL AND RADIOMETRIC PROCEDURES FOR ALUMINUM, BERYLLIUM, AND COBALT.
W. D. Ehmann and T. P. Kohman.
Geochim. Cosmochim. Acta 14, 340-363 (1958).
- J58f COSMIC-RAY-INDUCED RADIOACTIVITIES IN METEORITES - II: Al^{26} , Be^{10} , AND Co^{60} IN AEROLITES, SIDERITES, AND TEKTITES.
W. D. Ehmann and T. P. Kohman.
Geochim. Cosmochim. Acta 14, 364-379 (1958).
- J59a DISINTEGRATION SCHEME OF LONG-LIVED ALUMINUM-26.
R. A. Rightmire, J. R. Simanton, and T. P. Kohman.
Phys. Rev. 113, 1069-1077 (1959).
- J60a NEW NUCLIDE, KRYPTON-74
J. H. Gray, III, H. L. Hamester, and A. A. Caretto, Jr.
Phys. Rev. 120, 977-979 (1960).
- J60b THORIUM ISOTOPES METHOD FOR DATING MARINE SEDIMENTS.
I. Almodóvar and T. P. Kohman.
Preprints of General Papers, Division of Petroleum Chemistry,
American Chemical Society 5, (3) 165-169 (1960).
- J61a CHRONOLOGY OF NUCLEOSYNTHESIS AND EXTINCT NATURAL RADIOACTIVITY.
T. P. Kohman
J. Chem. Educ. 38, 73-82 (1961).

- J61b COMMENTS ON THE TIME DEPENDENCE OF NUCLEOSYNTHESIS.
T. P. Kohman.
Proc. Conf. on Problems Related to Interplanetary Matter,
Highland Park, Illinois, 1960 June, pages 7-8 (1961).
- J61c NATURAL ALPHA RADIOACTIVITY IN MEDIUM-HEAVY ELEMENTS.
R. D. Macfarlane and T. P. Kohman.
Phys. Rev. 121, 1758-1769 (1961).
- J61d EXCITATION FUNCTIONS OF (p,2 NUCLEON) REACTIONS.
P. P. Strohal and A. A. Caretto, Jr.
Phys. Rev. 121, 1815-1822 (1961).
- J61e NEUTRON-DEFICIENT NUCLIDES OF HAFNIUM AND LUTETIUM.
E. R. Merz and A. A. Caretto, Jr.
Phys. Rev. 122, 1558-1563 (1961).
- J62a COSMIC-RAY-INDUCED RADIOACTIVITIES IN METEORITES - III: CALCULATION OF
PRODUCTION RATES OF SPECIFIC NUCLIDES IN IRON METEORITIDS.
P. S. Goel.
Researches on Meteorites (C. B. Moore, Ed.; John Wiley and Sons, Inc.,
New York) 36-67 (1962).
- J62b COSMIC-RAY-INDUCED RADIOACTIVITIES IN METEORITES - IV: CARBON-14 IN A
STONE AND AN IRON.
P. S. Goel and T. P. Kohman.
Researches on Meteorites (C. B. Moore, Ed.; John Wiley and Sons, Inc.,
New York) 100-106 (1962).
- J62c COSMOGENIC CARBON-14 IN METEORITES AND TERRESTRIAL AGES OF "FINDS"
AND CRATERS.
P. S. Goel and T. P. Kohman.
Science 136, 875-876 (1962).
- J62d RECOIL BEHAVIOR OF THE $\text{Cu}^{65}(\text{p,pn})\text{Cu}^{64}$ REACTION.
E. R. Merz and A. A. Caretto, Jr.
Phys. Rev. 126, 1173-1180 (1962).
- J62e EXCITATION FUNCTIONS OF (p,xp) REACTIONS.
D. L. Morrison and A. A. Caretto, Jr.
Phys. Rev. 127, 1731-1738 (1962).
- J63a TERRESTRIAL AGES OF METEORITES FROM COSMOGENIC CARBON-14.
T. P. Kohman and P. S. Goel.
Radioactive Dating (IAEA, Vienna) 395-411 (1963).
- J63b COSMIC-RAY-EXPOSURE HISTORY OF METEORITES FROM COSMOGENIC CHLORINE-36.
P. S. Goel and T. P. Kohman.
Radioactive Dating (IAEA, Vienna) 413-432 (1963).
- J63c RADIOACTIVE NUCLIDES PRODUCED BY COSMIC RAYS IN SPUTNIK 4.
J. P. Shedlovsky and J. H. Kaye.
J. Geophys. Res. 68, 5069-5078 (1963).

- J63d SEARCH FOR COSMIC-RAY-INDUCED RADIOACTIVITY IN TEKTITES.
T. P. Kohman, M. M. Chakrabartty, P. S. Goel, and G. V. S. Rayudu.
Incomplete manuscript.
Abstract: Second International Symposium on Tektites (University of Pittsburgh, 1973 September), Program and Abstracts, No. 28, page 24; presented by Rayudu. (See *Geochim. Cosmochim. Acta* 28, 753 (1964)).
- J64a FORMATION CROSS SECTIONS OF VARIOUS RADIONUCLIDES FROM Ni, Fe, Si, Mg, O, AND C FOR PROTONS OF ENERGIES BETWEEN 130 AND 400 MEV.
G. V. S. Rayudu
Canadian J. Chem. 42, 1149-1154 (1964).
- J64b RECOIL STUDY OF THE $Zn^{68}(p,2p)Cu^{67}$ REACTION.
D. L. Morrison and A. A. Caretto, Jr.
Phys. Rev. 133B, 1165-1170 (1964).
- J64c 2π -DIFFERENTIAL RECOIL STUDY OF THE $Cu^{65}(p,pn)Cu^{64}$ REACTION.
D. J. Reuland, N. K. Ganguly, and A. A. Caretto, Jr.
Phys. Rev. 133B, 1171-1177 (1964).
- J64d RADIONUCLIDE PRODUCTIONS IN THICK IRON TARGETS BOMBARDED WITH 1- AND 3-GEV PROTONS.
J. P. Shedlovsky and G. V. S. Rayudu
J. Geophys. Res. 69, 2231-2242 (1964).
- J64e CHLORINE-36 PRODUCED BY NEUTRON CAPTURE IN METEORITES.
P. S. Goel.
Nature 203, 1162-1163 (1964).
- J64f ON THE DECAY OF 1.25-MIN Rb^{82} .
S. Jha, T. D. Nainau, A. S. Johnston, D. R. Reddy, and G. V. S. Rayudu.
Bull. Am. Phys. Soc. (II) 9, 472 (1964) (Abstract).
- J65a HALF-LIFE OF MANGANESE-53 FROM METEORITE OBSERVATIONS.
J. H. Kaye and P. J. Cressy.
J. Inorg. Nucl. Chem. 27, 1889-1892 (1965).
- J65b METHOD FOR THE ISOLATION OF THORIUM FROM SILICEOUS MATERIALS.
I. Almodóvar,
Anal. Chim. Acta 33, 426-433 (1965).
- J65c COSMOGENIC RADIONUCLIDES IN THE BONDUC METEORITE.
P. J. Cressy, Jr., and J. P. Shedlovsky.
Science 148, 1716-1717 (1965).
- J66a STUDY OF (p,n) REACTIONS BETWEEN 100 AND 400 MEV.
W. J. Treytl and A. A. Caretto, Jr.
Phys. Rev. 146, 836-840 (1966).
- J66b PSEUDO-DIFFUSION IN OCEAN SEDIMENTS.
T. P. Sarma and T. P. Kohman.
All India Symposium on Radioactivity Metrology and Radionuclides (Bombay, 1966), 308-318. (*Chem. Abst.* 67, 66622 (1967)).

- J67a COSMOGENIC RADIOACTIVITIES IN THE PEACE RIVER AND HARLETON CHONDRITES.
J. P. Shedlovsky, P. J. Cressy, Jr., and T. P. Kohman.
J. Geophys. Res. 72, 5051-5058 (1967).
- J67b MÜSSBAUER SPECTROSCOPY OF IRON IN COAL.
J. E. Lefelhocz, A. F. Friedel, and T. P. Kohman.
Geochim. Cosmochim. Acta 31, 2261-2273 (1967).
- J67c NUCLIDE PRODUCTION BY COSMIC RAYS IN METEORITES AND ON THE MOON.
T. P. Kohman and M. L. Bender.
High Energy Nuclear Reactions in Astrophysics (B. S. P. Shen, Ed.;
W. A. Benjamin, Inc., New York), 169-245 (1967).
- J68a FORMATION CROSS SECTIONS OF VARIOUS RADIONUCLIDES FROM Ni, Fe, Si, Mg,
O, AND C FOR PROTONS OF ENERGIES BETWEEN 0.5 AND 2.9 GEV.
G. V. S. Rayudu.
J. Inorg. Nucl. Chem. 30, 2311-2315 (1968).
- J68b NUCLEAR REACTIONS FOR THE STUDY OF SOLAR FLARE PROTONS.
G. V. S. Rayudu.
Trans. Am. Nucl. Soc. 11, 21-22 (1968).
- J69a ISOTOPIC COMPOSITION OF METEORITIC THALLIUM.
R. G. Ostic, H. M. El-Badry, and T. P. Kohman.
Earth and Planetary Science Letters 7, 72-76 (1969).
- J70a LEAST-SQUARES FITTING OF DATA WITH LARGE ERRORS.
T. P. Kohman.
J. Chem. Ed. 47, 657-658 (1970).
- J70b A UNIVERSAL MAGNITUDE SCALE FOR ASTRONOMICAL OBJECTS.
T. P. Kohman.
Bull. Amer. Astron. Soc. 2, 326 (1970).
(Abstract of paper presented at Boulder, Colorado, meeting of AAS,
1970 June).
- J72a SEARCH FOR EXTINCT NATURAL RADIOACTIVITY OF ^{205}Pb VIA THALLIUM-ISOTOPE
ANOMALIES IN CHONDRITES AND LUNAR SOIL.
J. M. Huey and T. P. Kohman.
Earth and Planetary Science Letters 16, 401-412 (1972).
- J73a ^{207}Pb - ^{206}Pb ISOCHRON AND AGE OF CHONDRITES.
J. M. Huey and T. P. Kohman.
J. Geophys. Res. 78, 3227-3244 (1973).
- J73b CHRONOLOGY OF GALACTIC HEAVY-ELEMENT NUCLEOSYNTHESIS.
T. P. Kohman and J. M. Huey.
Cosmochemistry (A. G. W. Cameron, Ed.; D. Reidel Publ. Co., Dordrecht-
Holland), 165-166 (1973).
(Abstract of paper presented at Symposium on Cosmochemistry, Cambridge,
Mass., 1972 August.)
- J74a PREPARATION OF HIGH PURITY CONCENTRATED HYDROFLUORIC ACID.
M. S. Lancet and J. M. Huey.
Anal. Chem. 46, 1360-1361 (1974).

- D57b COSMIC-RAY-INDUCED RADIOACTIVITIES IN METEORITES AND TEKTITES.
W. D. Ehmann (with T. P. Kohman).
NYO-6634 (1957 June 3).
- D59a NATURAL ALPHA RADIOACTIVITY IN MEDIUM-HEAVY ELEMENTS.
R. D. Macfarlane (with T. P. Kohman).
NYO-7687 (1959 May 28).
- D60a THORIUM ISOTOPES METHOD FOR DATING MARINE SEDIMENTS.
I. Almodovar (with T. P. Kohman).
NYO-8919 (1960 May 31).
- D61a A SYSTEMATIC STUDY OF (p,xp) REACTIONS IN THE 100-400 MEV REGION.
D. L. Morrison (with A. A. Caretto, Jr.).
NYO-8921 (1961 June 2).
- D62a COSMOGENIC CARBON-14 AND CHLORINE-36 IN METEORITES.
P. S. Goel (with T. P. Kohman).
NYO-8922 (1962 June 4).
- D63a COSMOGENIC X-RAY AND β -RAY EMITTERS IN IRON METEORITES.
J. H. Kaye (with T. P. Kohman and J. P. Shadlovsky).
NYO-8923 (1963 June 27).
- D64a COSMOGENIC RADIONUCLIDES IN STONE METEORITES.
P. J. Cressy, Jr. (with T. P. Kohman).
NYO-8924 (1964 June 1).
- D64b DATING OF MARINE SEDIMENTS BY IONIUM AND PROTACTINIUM METHODS.
T. P. Sarma (with T. P. Kohman).
NYO-8925 (1964 June 1).
- D72a LEAD AND THALLIUM ABUNDANCES AND ISOTOPIC COMPOSITION IN CHONDRITE
METEORITES
J. M. Huey (with T. P. Kohman).
COO-3236-1

S. Special Reports

THALLIUM IN METEORITES

- S60a I. ISOLATION PROCEDURES
H. M. El-Badry, 1-27.
- S60b II. SPECTROCHEMICAL DETERMINATION.
E. S. Hodge and W. K. Baer, 28-33.
- S60c III. THALLIUM CONTENT OF METEORITES.
H. M. El-Badry and T. P. Kohman, 34-42.

Joint Report of Carnegie Institute of Technology and Mellon Institute.
NYO-8920 (1960 December 20).

S70a TABLES OF URANIUM-THORIUM-LEAD DECAY-GROWTH FUNCTIONS.

T. P. Kohman.

NYO-844-79 (1970 May 15).

S70b A UNIVERSAL MAGNITUDE SCALE FOR ASTRONOMICAL OBJECTS.

T. P. Kohman.

NYO-844-80 (1970 June 10).

Incomplete manuscript prepared for publication in The Astronomical Journal.

P. Progress Reports

NUCLEAR CHEMISTRY RESEARCH. PROGRESS REPORT FOR CONTRACT TERM 1950-1951.

T. P. Kohman.

NYO-3230 (1951 May 31).

D. Research Progress

- P51a 1. Search for Natural Alpha Radioactivity in Medium-Heavy Elements, 5-6.
- P51b 2. Natural Beta Radioactivity of Rare Earth Elements, 6-7.
- P51c 3. Search for Weak Natural Beta Activities by the Method of Active Daughter Extraction, 7.
- P51d 4. Search for Extinct Natural Radioactivities, 7-8.
- P51e 5. Spectrometry of X-Rays and Low-Energy Gamma Rays, 8-9.
- P51f 6. Influence of Large-Angle Scattering on Alpha-Particle Measurements, 9.
- P51g 7. Nuclear Systematics, 9.

NUCLEAR CHEMISTRY RESEARCH. PROGRESS REPORT FOR CONTRACT TERM 1951-1952.

T. P. Kohman.

NYO-3230 (1952 May 31).

D. Research Progress and Plans

- P52a 1. Studies of Weak Natural Beta Activities, 4-5.
- P52b 2. Search for Weak Natural Beta Activities by Method of Active-Daughter Extraction, 6-7.
- P52c 3. Search for Natural Alpha Activity in Medium-Heavy Elements, 7.
- P52d 4. Search for Extinct Natural Radioactivities, 7-9.
- P52e 5. New Methods of Geological Age Measurement, 9-11.
- P52f 6. High Energy Nuclear Reactions, 12-13.
- P52g 7. Theoretical Work, 13-14.

NUCLEAR CHEMISTRY RESEARCH. PROGRESS REPORT FOR CONTRACT TERM 1952-1953.

T. P. Kohman.

NYO-3621 (1953 May 31).

D. Research Progress and Plans

- P53a 1. Weak Natural Beta Radioactivities, 7-15.
- P53b 2. Weak Natural Alpha Radioactivities, 16-18.
- P53c 3. Search for Extinct Natural Radioactivities, 18-20.
- P53d 4. Induced Radioactivity and Nuclear Reactions, 20-26.
- P53e 5. Methods of Geochronometry, 26-33.
- P53f 6. Theoretical Work, 33-34.

(Progress Report for 1953-1954 cannot be found.)

NUCLEAR CHEMISTRY RESEARCH, 1954-1955.

T. P. Kohman.

NYO-6629 (1955 May 31).

A. SUMMARY OF RESEARCH PROGRESS

- P55a 1. Natural Radionuclides, 1.
- P55b 2. Artificial Radionuclides, 1.
- P55c 3. Nuclear Reactions, 1-2.
- P55d 4. Nuclear Geochemistry, 2.
- P55e 5. Miscellaneous Experimental Activities, 2.
- P55f 6. Nuclear Systematics and Theory, 2.

NUCLEAR CHEMISTRY RESEARCH, 1955-1956.

T. P. Kohman.

NYO-6631 (1956 May 31).

A. RESEARCH PROGRESS SUMMARY

3. Scientific Results and Work in Progress

- P56a a. Artificial Radionuclides, 2.
- P56b b. Primary Natural Radionuclides, 2.
- P56c c. Induced Natural Radionuclides, 2-3.
- P56d d. Extinct Natural Radionuclides, 3.
- P56e e. Nuclear Reactions, 3.
- P56f f. Instrumentation for Radioactivity Measurement, 3.
- P56g g. Miscellaneous Experimental Activities, 3.

NUCLEAR CHEMISTRY RESEARCH, 1956-1957.

T. P. Kohman, Supervisor, 1956-1957; A. A. Caretto, Supervisor, 1957-1958.

NYO-7682 (1957 July 1).

A. RESEARCH PROGRESS SUMMARY

3. Scientific Results and Work in Progress

- P57a a. Artificial Radionuclides, 2-3.
- P57b b. Primary Natural Radionuclides, 3.
- P57c c. Induced Natural Radionuclides, 3-4.
- P57d d. Extinct Natural Radioactivity, 4-5.
- P57e e. Nuclear Reactions, 5.
- P57f f. Geological and Astronomical Applications of Nuclear Chemistry, 5-6.
- P57g g. Radioactivity Measurement instrumentation, 6-7.
- P57h h. Radiochemical Techniques, 7-8.

NUCLEAR CHEMISTRY RESEARCH, 1957-1958.

A. A. Caretto, Jr., Supervisor, 1957-1958; T. P. Kohman, Supervisor, 1958-1959.

NYO-7683 (1958 July 1).

A. RESEARCH PROGRESS SUMMARY

3. Scientific Results and Work in Progress

P58a	a. Artificial Radionuclides, 2.
P58b	b. Primary Natural Radionuclides, 2-3.
P58c	c. Induced Natural Radionuclides, 3.
P58d	d. Extinct Natural Radioactivity, 3.
P58e	e. Nuclear Reactions, 3-4.
P58f	f. Geological and Astronomical Applications of Nuclear Chemistry and Miscellaneous Experimental Activities, 4-5.

NUCLEAR CHEMISTRY RESEARCH, 1958-1959.

T. P. Kohman, Supervisor; A. A. Caretto, Jr., Associate Supervisor.
NYO-7688 (1959 July 30).

A. RESEARCH PROGRESS SUMMARY

4. Scientific Results and Work in Progress

P59a	a. Artificial Radionuclides, 4.
P59b	b. Primary Natural Radionuclides, 5-6.
P59c	c. Extinct Natural Radionuclides, 7.
P59d	d. Induced Natural Radionuclides, 7.
P59e	e. Nuclear Reactions, 8-9.
P59f	f. Geological and Astronomical Applications of Nuclear Chemistry, 9.
P59g	g. Miscellaneous Activities, 9-10.

NUCLEAR CHEMISTRY RESEARCH AT CARNEGIE INSTITUTE OF TECHNOLOGY, 1959-1960. PROGRESS REPORT.

T. P. Kohman and A. A. Caretto, Jr., Supervisors.
TID-6322 (1960 July 31).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Reaction Studies

P60a	1. Mechanism of (p,pn), (p,2p), and (p,2n) Reactions (P. P. Strohal), 26-31.
P60b	2. Excitation Functions of (p,xp) Reactions (D. L. Morrison), 31-32
P60c	3. Excitation Functions of Light-Mass Nuclides from Targets in the Mass Region A = 25-60 (J. H. Gray), 32-33.
P60d	4. Recoil Studies of the $\text{Cu}^{65}(\text{p,pn})\text{Cu}^{64}$ Reaction at Proton Energies from 80 to 440 MeV (E. R. Merz), 33-34.
P60e	5. Practical Production of Carrier-free Magnesium-28 (E. R. Merz), 34-37.
P60f	6. Nuclear Reactions in Thick Targets by 440-MeV Protons (P. S. Goel), 38-41.

B. Properties of Radionuclides

- P60g 1. New Nuclide, Krypton-74 (J. H. Gray and H. L. Hamster), 42-44.
 P60h 2. Disintegration Scheme of Iron-60 (E. R. Merz), 42, 45-48.
 P60i 3. Neutron-Deficient Isotopes of Mercury and Gold
 (E. R. Merz), 42-44.
 P60j 4. Search for Hg²⁰⁶ and Hf¹⁸² (E. R. Merz), 50-51.
 P60k 5. Artificial Alpha Activity in Medium-Heavy Elements
 (E. R. Merz), 51-52.
 P60l 6. Neutron-Deficient Isotopes of Hafnium and Lutetium
 (E. R. Merz), 52-54.

C. Nuclear Geochemistry and Astrochemistry

- P60m 1. Thorium Isotopes Method for Dating Marine Sediments
 (I. Almodovar), 55-61.
 P60n 2. Measurements of Cl³⁶, Si³², and C¹⁴ in Siderites and of C¹⁴
 in Aerolites and Tektites (P. S. Goel), 62-65.
 P60o 3. Studies of Long-Lived Cosmic-Ray-Induced Radioactivity in
 Meteorites and Tektites (M. M. Chakrabartty), 65-68.
 P60p 4. Studies of X-Ray Emitters Produced in Meteorites by
 Cosmic Rays (J. H. Kays and P. J. Cressy), 68-73.
 P60q 5. Calculation of Production Rates of Specific Nuclides in
 Meteoroids by Cosmic Radiation (P. S. Goel), 74-80.
 P60r 6. Search for Extinct Natural Radioactivity of Cm²⁴⁷
 (E. R. Merz), 80-82.
 P60s 7. Chronology of Nucleosynthesis and Extinct Natural
 Radioactivity (T. P. Kohman), 82-86.
 P60t 8. Tektite Investigations (T. P. Kohman and P. S. Goel), 86-93.

NUCLEAR CHEMISTRY RESEARCH AT CARNEGIE INSTITUTE OF TECHNOLOGY,
 1960-1961. PROGRESS REPORT.

T. P. Kohman and A. A. Caretto, Jr., Supervisors.
 TID-12849 (1961 June 15).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Reaction Studies

- P61a 1. A Systematic Study of (p,xp) Reactions between 100 and
 400 MeV (D. L. Morrison), 11-27.
 P61b 2. Excitation Functions of Light Mass Nuclides from Targets
 in the Mass Region A = 25-60 (J. H. Gray), 27-32.
 P61c 3. The (p,n) Reaction at 100-400 MeV (W. J. Treytl), 32-33.
 P61d 4. (p,2 nucleon) Reaction (D. J. Reuland), 33-34.
 P61e 5. Recoil Behavior of the Cu⁶⁵(p,pn)Cu⁶⁴ Reaction
 (E. R. Merz and A. A. Caretto, Jr.), 34-37.
 P61f 6. The Al²⁷(p,3pn)Na²⁴ Monitor Reaction Excitation Function
 (D. J. Reuland), 38-39.
 P61g 7. Cross Section Measurements of Some Long-Lived Radionuclides
 from Oxygen, Silicon, and a Few First-Row Elements
 (J. P. Shedlovsky and P. S. Goel), 40-41.

B. Properties of Radionuclides

- P61h 1. The Nuclides Cr⁵⁶ and Ti⁵² (E. R. Merz), 42-44.
 P61i 2. Neutron Rich Isotopes of Hafnium (E. R. Merz), 44-45.
 P61j 3. Sb¹²⁹ Branching Ratio (D. L. Morrison), 46.

C. Nuclear Geochemistry and Cosmochemistry

- P61k 1. Cosmogenic C^{14} and Cl^{36} in Iron Meteorites and C^{14} in Stones (P. S. Goel), 47-54.
- P61l 2. Cosmic-Ray-Produced Beta- and X-Ray Emitters in Stone Meteorites (P. J. Cressy, Jr.), 54-58.
- P61m 3. Electron-Capture Nuclides Produced in Iron Meteorites by Cosmic Rays (J. H. Kays), 58-63.
- P61n 4. Cosmic-Ray-Induced Radioactivity in Meteorites. B^{10} , Al^{26} , K^{40} , and Co^{60} in Siderites (M. N. Chakrabartty), 63-68.
- P61o 5. Cosmic-Ray-Induced Radioactivity in Meteorites. B^{10} , Al^{26} and Co^{60} in Aerolites and Tektites (M. N. Chakrabartty), 68-72.
- P61p 6. Cosmic-Ray-Induced Radioactivity in Meteorites. Mn^{53} and Ni^{59} in Stones (J. H. Kays and M. N. Chakrabartty), 72-74.
- P61q 7. Production of Co, Mn, Cr, and V in Thick Iron Targets by 440-MeV Protons Simulating Meteorite Exposure to Cosmic Rays (J. P. Shedlovsky), 74-84.
- P61r 8. Effect of "Pseudo-Diffusion" in Marine Sediments on the Ionium Dating Method (T. P. Sarma), 84-88.
- P61s 9. Possible U^{235} Dating Method for Minerals (T. P. Sarma), 88-90.

D. Miscellaneous

- P61t 1. Preparation of Tellurium Targets by Electrodeposition (W. J. Treytl), 91.
- P61u 2. Separation of Iodine from Tellurium Targets (W. J. Treytl), 91-92.
- P61v 3. Graphical Method for Determination of Nonlinearity Correction of Counters (P. Zwitkowski), 93-96.
- P61w 4. Feasibility of Reactor Production of K^{40} for Tracer Use (T. P. Kohman), 96-99.

NUCLEAR CHEMISTRY RESEARCH AT CARNEGIE INSTITUTE OF TECHNOLOGY, 1961-1962. PROGRESS REPORT.

T. P. Kohman, Supervisor.

TID-16290 (1962 June 1).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Reaction Studies

- P62a 1. Production Rates of Long-Lived Radionuclides from Iron, Cobalt, and Copper with 0.4-6.2-BeV Protons (J. P. Shedlovsky and P. S. Goel), 11-12.
- P62b 2. Proton Excitation Functions for Mn^{52} , V^{48} , Cr^{51} , and Be^7 from Iron from 100 to 400 MeV (G. V. S. Rayudu), 13.
- P62c 3. Study of Induced α -Activity in the Microsecond-Millisecond Half-life Range (F. Mesojedec), 14-16.

B. Nuclear Geochemistry and Cosmochemistry

- P62d 1. Radioactive Dating of Marine Sediments (T. P. Sarma), 17-19.
- P62e 2. Pseudo-Diffusion in Ocean and Maria Sediments (T. P. Sarma), 20-30.
- P62f 3. Cosmogenic C^{14} in Meteorites (P. S. Goel), 31-38.
- P62g 4. Cosmogenic Cl^{36} in Iron Meteorites (P. S. Goel), 39-42.

- P62h 5. Cosmogenic Radionuclides in Iron Meteorites (J. H. Kaya), 43-46
 P62i 6. Cosmogenic Radionuclides in Stone Meteorites
 (P. J. Cressy), 47-48.
 P62j 7. Cosmogenic Radionuclides in the Harleton Chondrite.
 (M. M. Chakrabartty, P. J. Cressy, P. S. Goel, J. H. Kaya,
 and J. P. Shedlovsky), 49-53.
 P62k 8. Upper Limits on Al^{26} , Be^{10} , and C^{14} in Tektites
 (M. M. Chakrabartty and P. S. Goel), 54-57.
 P62l 9. Further Search for Al^{26} and Be^{10} in Tektites
 (G. V. S. Rayudu), 57-60.
 P62m 10. 3-BeV Proton Bombardment of a Simulated Iron Meteorite
 (J. P. Shedlovsky), 60-66.
 P62n 11. Simulated Cosmic-Ray Irradiation of a Thick Iron Target with
 450-MeV Protons (G. V. S. Rayudu), 67-70.

C. Miscellaneous

- P62o 1. Teflon Still for Hydrofluoric Acid Decomposition of
 Silicates (P. J. Cressy), 71-73.
 P62p 2. Search for Natural Radioactivity of Ca^{48} (M. I. Appelbaum),
 73-74.
 P62q 3. Calibration of Low-Level Solid-Sample Beta Geiger Counters
 (J. P. Shedlovsky, M. M. Chakrabartty, P. J. Cressy, and
 P. S. Goel), 74-79.
 P62r 4. Performance Characteristics of Methane-Flow Proportional
 Counters (T. P. Kohman), 80-89.

NUCLEAR CHEMISTRY RESEARCH AT CARNEGIE INSTITUTE OF TECHNOLOGY, 1962-1963. PROGRESS REPORT.

T. P. Kohman and J. P. Shedlovsky, Supervisors.
 TID-18961 (1963 June 1).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Reaction Studies

- P63a 1. Thin Target Cross Sections from Iron and Stone Meteoritic
 Elements for 130 to 400 MeV Protons (G. V. S. Rayudu), 10-14.
 P63b 2. Simulated Cosmic-Ray Irradiation of a Thick Iron Target with
 100 MeV Protons (G. V. S. Rayudu), 15-20.
 P63c 3. Simulated Cosmic-Ray Irradiation of a Thick Iron Target with
 450 MeV Protons (G. V. S. Rayudu), 20-22.
 P63d 4. Simulated Cosmic-Ray Irradiation of a Thick Iron Target with
 1 GeV Protons (J. P. Shedlovsky and G. V. S. Rayudu), 23-29.
 P63e 5. Simulated Cosmic-Ray Irradiation of a Thick Stone Target with
 440 MeV Protons (G. V. S. Rayudu), 30-32.
 P63f 6. Simulated Cosmic-Ray Irradiation of a Thick Stone Target with
 1 GeV Protons (G. V. S. Rayudu and J. P. Shedlovsky), 33-37.
 P63g 7. Simulated Cosmic-Ray Irradiation of a Thick Stone Target with
 3 GeV Protons (J. P. Shedlovsky and G. V. S. Rayudu), 38-39.
 P63h 8. Study of Induced Alpha-Activity in the Microsecond-Millisecond
 Half-Life Range (F. Mesojedec), 40-41.

B. Nuclear Geochemistry and Cosmochemistry

- P63i 1. Absolute Dating of Sediments with Natural Radioactive Nuclides (T. P. Sarma), 42-44.
- P63j 2. Effect of "Pseudo-Diffusion" in Ocean and Maria Sediments (T. P. Sarma), 45-62.
- P63k 3. Cosmogenic Radioactivities in Iron Meteorites (J. H. Kaye), 62-77.
- P63l 4. Cosmogenic Radionuclides in Stone Meteorites (P. J. Cressy, Jr.), 78-85.
- P63m 5. Cosmogenic Radionuclides in the Bondoc Achondrite and Bogou Iron (J. P. Shedlovsky), 86-87.
- P63n 6. The Half-Life of Manganese-53 (J. H. Kaye and P. J. Cressy, Jr.), 88-90.
- P63o 7. Cosmogenic Radioactivities in a Fragment of Sputnik IV (J. P. Shedlovsky and J. H. Kaye), 91-97.
- P63p 8. Search for Be¹⁰ in Tektites (G. V. S. Rayudu), 98-99.
- P63q 9. Conversion of Sealed Anticoincidence Shield Counters to Flow Counters (A. Henry and N. Viswanathan), 100-102.

C. Miscellaneous

- P63r 1. Search for Natural Radioactivity of Calcium-48 (M. R. Miller), 102.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,
 CARNEGIE INSTITUTE OF TECHNOLOGY, 1963-1964. PROGRESS REPORT.
 T. P. Kohman, Supervisor.
 TID-21007 (1964 July 31).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Reaction Studies

- P64a 1. Radionuclide Distribution in a Thick Iron Target Bombarded by 6.1-GeV Protons (G. V. S. Rayudu), 7-9.
- P64b 2. Nuclear Reactions for the Study of Solar Flare Protons (G. V. S. Rayudu), 10-13.
- P64c 3. Formation Cross Sections of Various Radionuclides from Ni, Fe, Si, Mg, O, and C for Protons of Energies between 0.5 and 3.0 GeV (G. V. S. Rayudu), 13-16.
- P64d 4. Cross Sections for Production of Li, Be, and B (A. Huff), 16-19.
- P64e 5. Interpretation of Thick-Target Bombardments (M. L. Bender and T. P. Kohman), 20-25.

B. Nuclear Geochemistry and Cosmochemistry

- P64f 1. Dating of Marine Sediments by Ionium and Protactinium Methods (T. P. Sarma), 26-27.
- P64g 2. Search for Cosmic-Ray-Induced Activities in Tektites (G. V. S. Rayudu), 28-30.
- P64h 3. Cosmogenic Radionuclides in Stone Meteorites (P. J. Cressy, Jr.), 31-32.
- P64i 4. Depth Studies of Cosmogenic Radionuclides in Iron Meteorites (R. F. Schall, Jr.), 32-34.
- P64j 5. Muon-Induced Radioactivity in Nature (N. Viswanathan), 35-37.
- P64k 6. The Possibility of Using I¹²⁹ in Geochronology (A. T. Kuo), 37-39.

C. Miscellaneous

- P64l 1. Radiochemical Aspects of Disintegration Scheme Studies (G. V. S. Rayudu), 40-41.
- P64m 2. Self-Absorption Curve for Al^{26} Positrons in Al_2O_3 (R. F. Schall, Jr.), 41-45.
- P64n 3. Mössbauer Studies of Iron in Natural Materials (S. J. Catz), 46-48.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,
CARNEGIE INSTITUTE OF TECHNOLOGY, 1964-1965. PROGRESS REPORT.
T. P. Kohman, Supervisor.
(No NYO number; TID number not found) (1965 July 31).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Geochemistry

- P65a 1. Application of Fission-Track Dating to Tektites and Volcanic Glasses (R. G. Sextro), 6-10.
- P65b 2. Muon-Induced Radioactivity in Nature (N. Viswanathan), 10-12.
- P65c 3. Dating of Deep-Sea Sediments by Radioactivity Methods (A. T. Kuo and C. F. Beck, Jr.), 12-15.

B. Nuclear Cosmochemistry

- P65d 1. Determination of Mercury, Thallium, Lead and Bismuth in Iron Meteorites by Neutron Activation Analysis (S. N. Tandon), 16-20.
- P65e 2. Search for Extinct Natural Pb^{205} Radioactivity (S. N. Tandon), 20-23.
- P65f 3. Cosmogenic Radionuclide Studies in Large Iron Meteorites (R. F. Schall, Jr.), 24-28.
- P65g 4. Interpretation of Thick-Target Bombardments and Depth Dependence of Nuclear Reactions in Iron Meteoroids (M. L. Bender and T. P. Kohman), 29-64.

C. Nuclear Reaction Studies

- P65h 1. Search for Radioactivity of Ca^{48} (F. J. Van Itallie), 65-69.
- P65i 2. Bombardment of Thick Iron Targets with 450 MeV and 3 GeV Protons (G. V. S. Rayudu), 69-70.
- P65j 3. Muon-Induced Nuclear Reaction Studies (N. Viswanathan), 71-76.

D. Techniques and Instrumentation

- P65k 1. Iron Meteorite Digestion Apparatus (R. F. Schall, Jr.), 77-80.
- P65l 2. Radiochemical and Radiometric Procedures for Iron Meteorites (R. F. Schall, Jr.), 80-81.
- P65m 3. Carbon and Argon Extraction System for Silicates and Water-Soluble Materials (N. Viswanathan), 81-84.
- P65n 4. Small-Volume Gas Proportional Counters (R. F. Schall, Jr. and N. Viswanathan), 85-86.
- P65o 5. Mössbauer Spectrometry (M. J. Toia, S. J. Catz, and R. R. Edwards), 87-90.
- P65p 6. Counting Systems (M. J. Toia), 90-97.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,
 CARNEGIE INSTITUTE OF TECHNOLOGY, 1965-1966. PROGRESS REPORT.
 T. P. Kohman, Supervisor
 NYO-844-67 (1966 June 30).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Geochemistry

- P66a 1. Terrestrial Occurrence and Distribution of I^{129}
 (R. R. Edwards), 7-12.
- P66b 2. Mössbauer Spectrometry in Geochemistry
 (J. F. Lefelhocz), 13-17.
- P66c 3. Radiochemical Techniques for Uranium, Protactinium, and
 Thorium in Ocean Sediments (A. T. Kuo), 18-20.

B. Nuclear Cosmochemistry

- P66d 1. Cosmogenic I^{129} in Stone Meteorites
 (R. R. Edwards), 21-22
- P66e 2. Iron Meteorite Studies (R. F. Schall, Jr.), 23-25.
- P66f 3. Determination of Mercury, Thallium, and Bismuth in
 the Metal and the Troilite Phases of Iron Meteorites
 by Slow Neutron Activation Analysis (S. N. Tandon), 26-31.
- P66g 4. Determination of Lead in Iron Meteorites by Fast-Neutron
 Activation Analysis (S. N. Tandon), 32-37.

C. Nuclear Reaction Studies

- P66h 1. Search for Radioactivity of Ca^{48} (F. J. Van Itallie), 38-40.
- P66i 2. Muon-Induced Nuclear Reaction Studies (M. Viswanathan), 40-45.
- P66j 3. Bombardment of Thick Iron Targets with 100- and 450 MeV
 Protons (G. V. S. Rayudu), 45-47.

D. Techniques and Instrumentation

- P66k 1. Iron Meteorite Digestion Apparatus (R. F. Schall, Jr.), 48-50
- P66l 2. Mössbauer Instrumentation (M. J. Toia), 50-54.
- P66m 3. Small Volume X-Ray Counter (R. F. Schall, Jr.), 55-60.
- P66n 4. Counting Systems (M. J. Toia), 61-62.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,
 CARNEGIE INSTITUTE OF TECHNOLOGY, 1966-1967. PROGRESS REPORT.
 T. P. Kohman, Supervisor
 NYO-844-71 (1967 June 30).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Geochemistry

- P67a 1. Mössbauer Spectroscopy of Iron in Coal
 (J. F. Lefelhocz), 8-17.
- P67b 2. Terrestrial Occurrence and Distribution of I^{129}
 (R. R. Edwards and P. Rey), 18-29.
- P67c 3. Negative-Muon-Induced Nuclear Reaction Rates in Natural
 Materials (E. A. Pérez), 30-34.

B. Nuclear Cosmochemistry

- P67d 1. Mercury, Thallium, and Bismuth in Metal and Troilite Phases of Iron Meteorites by Neutron Activation Analysis (S. W. Tandon), 35-37.
- P67e 2. The Determination of Mercury, Thallium, and Bismuth in Iron Meteorites by Neutron Activation (J. T. Tanner), 38-44.
- P67f 3. Isotopic Composition of Thallium in Meteorites (R. G. Ostic), 45-48.
- P67g 4. Plans for Radiogenic Isotope Studies of Lunar Materials (R. G. Ostic and T. P. Kohman), 49-50.
- P67h 5. Expectation Levels of ^{129}I in Meteorites (T. P. Kohman), 50-62.

C. Nuclear Reaction Studies

- P67i 1. Search for Radioactivity of Ca^{48} (F. J. Van Itallie, R. D. Allerton, and M. R. Rao), 63-64.

D. Techniques and Instrumentation

- P67j 1. Counting Systems (M. J. Toia), 65-72.
- P67k 2. Cylindrical Anticoincidence Guard Counter (J. M. Huey, Jr.) 73-74.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,
 CARNEGIE-MELLON UNIVERSITY, 1967-1968. PROGRESS REPORT
 T. P. Kohman, Principal Investigator
 NYO-844-75 (1968 July 31).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Geochemistry

- P68a 1. Cadmium Abundances in Tektites by Neutron Activation (J. T. Tanner), 7-9.
- P68b 2. Mössbauer Spectroscopy of Iron in Coal (E. A. Pász), 10-20.
- P68c 3. Alpha Spectra of Uranium in Tektites (V. Tsoml), 21-30.

B. Nuclear Cosmochemistry

- P68d 1. Interpretation of Meteoritic Thallium Isotope Ratios (R. G. Ostic and T. P. Kohman), 31-37.
- P68e 2. Mercury and Bismuth Abundances in Iron Meteorites by Neutron Activation (J. T. Tanner), 37-41.
- P68f 3. Gamma-Gamma Coincidence Spectrometer for Measurement of Low-Level Gamma Emitters (J. R. Noyce), 42-53.
- P68g 4. Determination of Thallium and Lead in Meteorites (J. M. Huey), 54-65.

C. Nuclear Reaction Studies

- P68h 1. Search for Natural Radioactivity of Ca^{48} (E. Y. Fang), 66-72.

D. Techniques and Instrumentation

- P68i 1. Counting Systems (M. J. Toia), 73-80.

P68j PART III. CORRECTIONS TO 1966-1967 PROGRESS REPORT

- II. A. 2, 81.
- II. A. 3, 82.
- II. B. 3, 83.
- II. C. 1, 83.
- II. D. 1, 83.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH.

CARNEGIE-MELLON UNIVERSITY, 1968-1969. PROGRESS REPORT

T. P. Kohman, Principal Investigator.

NYO-844-76 (1969 June 30).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Geochemistry

- P69a 1. Mössbauer Spectroscopy of Natural Materials (E. A. Paez), 7-18.
- P69b 2. Alpha Spectroscopy of Uranium in Tektites
(T. J. Thomas), 19-33.
- P69c 3. Geochronological Potential of the K^{40} - Ca^{40} System
(T. P. Kohman), 34-48.

B. Nuclear Cosmochemistry

- P69d 1. Isotopic Composition of Lead and Thallium in Meteorites
(J. M. Huey), 49-54.
- P69e 2. Investigation of the Sm^{147} - Nd^{143} and Sm^{146} - Nd^{142} Systems
as Dating Methods (R. Y. Saleh), 55-69.
- P69f 3. Possible Additional Cases of Extinct Natural Radioactivity
(T. P. Kohman), 70-83.

C. Applied Nuclear Chemistry

- P69g 1. Concentrations of Gold, Indium, and Silver in Semi-
conductor Silicon (J. T. Tanner and P. C. Smith), 84-85.
- P69h 2. Segregation of Antimony in Nickel-Chromium Alloy Steel
During Temper Embrittlement (J. T. Tanner and
C. L. Smith), 85-86.

D. Techniques and Instrumentation

- P69i 1. Instrumentation (M. J. Toia), 87-92.
- P69j 2. Fast Gamma-Gamma Coincidence Spectrometry for
Activation Analysis (M. W. Haremic), 93-97.
- P69k 3. Gamma-Gamma Coincidence Spectrometer for Measurement
of Low-Level Gamma Emitters (H. J. Konish, Jr.), 98-104.
- P69l 4. Least-squares Fitting of Data with Large Errors
(T. P. Kohman), 105-107.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,
 CARNEGIE-MELLON UNIVERSITY, 1969-1970. PROGRESS REPORT.
 T. P. Kohman, Principal Investigator.
 NYO-844-82 (1970 June 30).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Geochemistry

- P70a 1. Investigation of the $\text{Sm}^{146}\text{-Nd}^{142}$ and $\text{Sm}^{147}\text{-Nd}^{143}$ Systems as Dating Methods (R. Y. Saleh), 6-13.
- P70b 2. Tables of Uranium-Thorium-Lead Decay-Growth Functions (T. P. Kohman), 14-17.
- P70c 3. Search for Long-Lived Transuranium Alpha Emitters and Other Anomalous Alpha Emitters in Natural Materials (N. Landes), 18-30.
- P70d 4. Mössbauer Spectroscopy of Coal-related Minerals (J. D. Ulmer), 31-36.

B. Nuclear Cosmochemistry

- P70e 1. Isotopic Composition of Lead and Thallium in Natural Materials (J. M. Huey), 37-45.
- P70f 2. Chronology of Galactic Heavy-element Nucleosynthesis (J. M. Huey and T. P. Kohman), 46-56.

C. Nuclear Chemistry

- P70g 1. Search for Natural Radioactivity of Calcium-48 (M. J. Yeager), 57-64.

D. Techniques and Instrumentation

- P70h 1. Instrumentation (M. Haramic), 65-68.
- P70i 2. Chemical Procedures for Purification of Uranium and the Alpha-spectroscopy of Uranium (N. Kan), 69-76.
- P70j 3. Alpha-isotope-dilution Analysis of Uranium and Thorium (B. M. P. Trevedi), 77-82.
- P70k 4. Least-squares Analysis of Experimental Alpha Spectra (B. M. P. Trevedi), 83-90.
- P70l 5. Design and Construction of a Position-sensitive Proportional Counter (J. R. Yocum, Jr. and J. F. Monahan), 91-95.

E. Miscellaneous

- P70m 1. A Universal Magnitude System for Astronomical Objects (T. P. Kohman), 96-97.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,
 CARNEGIE-MELLON UNIVERSITY, 1970-1971. PROGRESS REPORT
 T. P. Kohman, Principal Investigator; P. J. Karol, Co-Investigator.
 NYO-844-84 (1971 May 31).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Geochemistry

- P71a 1. Search for Anomalous Alpha Activity in Natural Materials (B. M. P. Trivedi), 6-11.
- P71b 2. Alpha Spectroscopy of Uranium in Tektites (N. Kan), 12-20.
- P71c 3. Neodymium-Samarium Separations for Geochronological Applications (P. J. Karol), 21-25.

B. Nuclear Cosmochemistry

- P71d 1. Isotopic Composition of Lead and Thallium in Natural Materials (J. M. Huey), 26-43.
- P71e 2. Chronology of Galactic Heavy-Element Nucleosynthesis (T. P. Kohman and J. M. Huey), 44-47.

C. Nuclear Chemistry

- P71f 1. Search for Natural Radioactivity of Calcium-48 (M. J. Yeager), 48-53.

D. Techniques and Instrumentation

- P71g 1. Instrumentation (M. Haramic), 54-56.
- P71h 2. Development of a Position-Sensitive Proportional Counter (C. Ritter), 57-59
- P71i 3. Least-Square Analysis of Experimental Alpha Spectra (B. M. P. Trivedi), 60-69.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,
 CARNEGIE-MELLON UNIVERSITY, 1971-1972. PROGRESS REPORT.
 T. P. Kohman, Principal Investigator; P. J. Karol Co-Investigator
 COO-3236-3 (1972 May 31).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Geochemistry

- P72a 1. Mössbauer Spectroscopy of Iron in Coal and Related Minerals (P. Kamarchik), 5-16.
- P72b 2. Alpha Spectroscopy of Uranium in Tektites (T. P. Kohman), 17-21.

B. Nuclear Cosmochemistry

- P72c 1. Pb²⁰⁷-Pb²⁰⁶ Isochron and Age of Chondrites (J. M. Huey), 22.
- P72d 2. Search for Extinct Natural Radioactivity of Lead-205 in Chondrites (J. M. Huey), 23.
- P72e 3. Lead-Thallium Cosmochronometry in the Early Solar System (J. M. Huey), 24-25.
- P72f 4. Lithium, Beryllium, and Boron in Cosmic Matter (M. S. Lancet), 26-33.
- P72g 5. Chronology of Galactic Heavy-Element Nucleosynthesis (T. P. Kohman and J. M. Huey), 34-43.

C. Nuclear Chemistry

- P72h 1. Lead Isotope Ratios Associated with Works of Art
(B. Kaisch and R. C. Callahan), 44-45.
- P72i 2. Radionuclide Production Rates in Argon by High-Energy
Protons (P. S. Goel), 46-50.
- P72j 3. Search for Natural Radioactivity of Calcium-48
(M. J. Yeager and T. P. Kohman), 51-56.

D. Techniques and Instrumentation

- P72k 1. Instrumentation (M. W. Harnic), 57-59.
- P72l 2. Manipulation of Alpha Spectra to Compensate for Drift
(S. B. Nachimson), 60-63.
- P72m 3. Sum-Coincidence Gamma Spectrometry (C. A. Ritter), 64-71.
- P72n 4. Julian-Day Calendar for Radioactivity Records and
Calculations (T. P. Kohman), 72-76.
- P72o 5. Procedure for Isolation of Rare-Earth Elements from
Natural Materials for Mass Spectrometry
(B. M. J. Banaal and P. J. Karol), 77-79.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,

CARNEGIE-MELLON UNIVERSITY, 1972-1973. PROGRESS REPORT.

T. P. Kohman, Principal Investigator; P. J. Karol, Co-Investigator
COO-3236-6 (1973 May 31).

PART II. RESEARCH PROGRESS REPORT AND PLANS

A. Nuclear Geochemistry

- P73a 1. Alpha Spectrometry of Uranium in Tektites
(S. B. Nachimson and T. P. Kohman), 5-18.

B. Nuclear Cosmochemistry

- P73b 1. Uranium-Thorium-Lead Dating of Meteorites (M. S. Lancet), 19-23.
- P73c 2. Search for Extinct Natural Radioactivity of Pb^{205}
(M. S. Lancet), 24,25.
- P73d 3. Search for Extinct Natural Radioactivity of ^{146}Sm .
(C.-C. Lin), 26-27.

C. Applied Nuclear Chemistry

- P73e 1. Lead-Isotope Ratios Associated with Works of Art
(B. Kaisch and R. C. Callahan), 28.
- P73f 2. Potential for Lead-Isotope Studies of Environmental
Pollution in Industrial Regions (T. P. Kohman), 29-43.

D. Techniques and Instrumentation

- P73g 1. Mass Spectrometry and Data Reduction
(M. S. Lancet and S. B. Nachimson), 44-45.
- P73h 2. Chemical Procedures for Low-Level Isotopic Analysis of Lead,
Thallium, Uranium, Thorium, and Lithium (M. S. Lancet), 46-51.
- P73i 3. Separations of Rare-Earth Elements for Geochronological
Applications (C.-C. Lin), 52-55.

NUCLEAR CHEMISTRY AND GEOCHEMISTRY RESEARCH,
 CARNEGIE-MELLON UNIVERSITY, 1973-1976. PROGRESS REPORT
 T. P. Kohman, Principal Investigator
 COO-3236-9 (1976 May 20).

PART II. RESEARCH PROGRESS REPORT

A. Nuclear Geochemistry

- P76a 1. Uranium-Thorium-Lead Geochronology of Terrestrial Rocks
 (R. A. Witter), 5-7.
- P76b 2. ^{147}Sm - ^{143}Nd Method of Geochronology (C.-C. Lin), 8-20.
- P76c 3. Tables of Uranium-Thorium-Lead Decay-Growth Functions
 (T. P. Kohman), 21-25.

B. Nuclear Cosmochemistry

- P76d 1. Uranium-Thorium-Lead Dating of Meteorites
 (M. S. Lancet and T. P. Kohman), 26-44.
- P76e 2. Niobium-92 As a Possible Extinct Natural Radionuclide
 (T. P. Kohman, M. S. Lancet, and C.-C. Lin), 45-56.
- P76f 3. Chronology of Galactic Heavy-Element Nucleosynthesis
 (T. P. Kohman), 57-67.

C. Applied Nuclear Chemistry

- P76g 1. Studies of Environmental Lead (D. L. Svec and D. L. Manis),
 68-71.
- P76h 2. Lead-Isotope Ratios As a Tracer of Environmental Lead
 Intake by Children (M. D. Rowe), 72-73.
- P76i 3. Lead-Isotope Ratios Associated with Works of Art
 (B. Keisch and R. C. Callahan), 74.

D. Instrumentation and Techniques

- P76j 1. Mass Spectrometry and Data Reduction
 (T. P. Kohman, M. S. Lancet, and C.-C. Lin), 75-80.

V. RESEARCH ACTIVITIES AND RESULTS

In Part IV all publications and reports are listed with complete titles, in chronological order within each category, so that a simple perusal of these lists gives a rather complete and accurate indication of the areas and extents of interest and activity at any time during the course of this Project. The following narrative summary, on the other hand, is organized according to the areas and objectives of the research activities themselves.

It would be impractical to discuss each topic thoroughly with a well balanced literature survey and citations of preceding, parallel, and subsequent work elsewhere, so it has been decided to omit such citations altogether. In general, the papers and reports themselves contain references to the pertinent open literature up to the time of their writing. In some cases subsequent developments are mentioned parenthetically.

It is also impractical to give many quantitative numerical results and data, but the discussions indicate the nature of such data which can be found in the various publications and reports.

A. High-Energy Nuclear Reactions

The Carnegie synchrocyclotron began producing circulating 350-MeV proton currents of $\sim 0.1 \mu\text{A}$ in 1952 and $\sim 1 \mu\text{A}$ in 1953. It was immediately used by Jones in attempts to produce certain unknown nuclides of potential usefulness (P52f). Success was achieved with ^{28}Mg , but only after it had already been discovered by others (P53d). The yield and an approximation cross section for production of ^{28}Mg from silicon at 350 MeV were determined (J53a).

Jones later measured the cross section for the production of ^{28}Mg , ^{35}S , ^{32}P , and ^{24}Na from natural chlorine with protons of energies from 45 to 433 MeV (P56e, D56b). The curve for ^{28}Mg was confirmed by Merz (P60e), following publication of a discrepant curve by another laboratory. Merz also determined the practical yields of ^{28}Mg from NaCl and KCl targets at 400 MeV (P60e).

Following the discovery of the long-lived ^{26}Al (Section B), practical thick-target yields for the production of this nuclide and ^{24}Na from magnesium bombarded with 15-MeV deuterons were determined (P56e, J57b). Rightmire also determined the thick-target ^{24}Al and ^{22}Na yields from magnesium bombarded with 20.8-MeV protons, found measurable but not useful amounts of this nuclide in heavily reactor-neutron-irradiated aluminum, and set an upper limit to the amount produced in 440-MeV-proton bombardment of KCl (P57e, D57a).

Beginning with Professor Caretto's association with this Project in 1957 there was a surge in the use of the Carnegie synchrocyclotron for the study of high-energy nuclear reactions (P58e, P59e). Caretto's chief interest was in elucidation of the mechanisms of relatively "simple" spallation reactions, in which only a small number of particles are emitted from the struck nucleus. Strohal measured the cross sections for 10 (p,2 nucleon) reactions with 250-440-MeV protons on mass 81-187 targets, and interpreted the excitation functions in terms of varying contributions of knock-on-cascade and nucleon-evaporation processes (P60a, J61d). Morrison determined the excitation functions from 100 to 440 MeV for a number of (p,xp) reactions, with $x = 2, 3, 4, 5$, for targets of mass number 30 to 187. The results indicated that both knock-on and evaporation contributed to the cross

sections, with the former predominating for large A because of Coulomb-barrier effects (P60b, P61a, D61a, J62e). (The cross sections, systematics, and theory have proven valuable in predicting the yields for practical production of useful neutron-rich nuclides by (p, xp) reactions with high-energy accelerators.) Gray measured 150-400-MeV-proton cross sections of a number of reactions, with particular interest in the effect of target mass on the yields of specific nuclides, which would be important not only for nuclear theory but also for interpretation of levels of cosmogenic radionuclides in meteorites (P60c, P61b). Reuland undertook the study of a number of (p, 2 nucleon) reactions (P61d), for which a refinement of the excitation function for the $^{27}\text{Al}(p, 3p)^{24}\text{Na}$ monitor reaction was considered necessary (P61f). Treytl made a special study of the (p, n) reaction for energies 130-423 MeV and target mass numbers 45-186. Although it was considered that nucleon evaporation should not play an important role in these cases, Monte Carlo calculations based on a simplified cascade model were inadequate to account for the observed cross sections (P61c, J66a).

Caretto's group also employed and refined the product-recoil technique for the study of "simple" reactions. Merz found that the recoil-kinetic-energy spectra for the reaction $^{65}\text{Cu}(p, pn)^{64}\text{Cu}$ at 73-400-MeV incident energies exhibited surprising amounts of structure. The conventional two-step mechanism could account for both the spectra and the recoil fractions below 200 MeV; above this energy the recoil fractions could be explained by increased importance of the knock-on mechanism, but the spectra showed discrepancies from the predictions based on that assumption (P60d, P61e, J62d). Morrison made similar studies of the $^{68}\text{Zn}(p, 2p)^{67}\text{Cu}$ reaction from 80 to 430 MeV. He found that the forward, backward, and transverse integral-range distributions could best be explained by predominance of the knock-on mechanism at all incident energies (P61a, D61a, J64b). Gray also used the recoil technique in the study of the nuclear reactions he was interested in (P61b). Reuland and Caretto, joined later by N. K. Ganguly, made two refinements in the recoil technique: (1) in 2 π geometry a number of thin recoil catchers replaced the single thick catcher; (2) a cylindrical thick catcher surrounding the target yielded, after sectioning, the angular distribution of the recoil nuclei. Both were applied to the $^{65}\text{Cu}(p, pn)^{64}\text{Cu}$ reaction. The partial-integral yield-versus-range curve from method (1) indicated that there were two range groups at 396-MeV bombarding energy but only one at 130 MeV. The short-range recoils were attributed to an inelastic (p, p') process followed by neutron evaporation, and the long-range recoils to the knock-on ejection of a neutron. Both methods suggested roughly equal contributions by each mechanism (J64c).

In connection with the studies of cosmic-ray-induced radioactivity in meteorites (Section E), laboratory experiments on production rates in target elements abundant in meteorites with artificially accelerated particles were undertaken (P61g). Shadlovsky and Goel measured cross sections for ^7Be , ^{14}C , ^{32}Si , and ^{36}Cl in iron at 0.4, 3.0, and 6.2 GeV as well as in Cu and Co at 0.44 GeV (P62a, D62a). Rayudu measured the cross sections for the production of a number of radionuclides (^7Be , ^{24}Na , ^{24}Na , ^{32}P , ^{48}V , ^{51}Cr , ^{52}Mn , ^{54}Mn , ^{56}Co) from a number of target elements (C, O, Mg, Si, Fe, Ni) at a number of proton energies from 130 to 400 MeV (P62b, P63a, J64a). For these products plus ^{44}Sc and ^{47}Sc and the same target elements, similar measurements were made for proton energies from 0.5 to 2.9 GeV (P64c, J68a). Because of their importance in atmospheric radioactivity, Goel et al. determined the cross sections for formation of a number of radionuclides with 360- and 580-MeV protons in argon (P72i, J74b).

Plans to determine by mass-spectrometric isotope-dilution analysis the yields and isotopic composition of Li, Be, and B from proton spallation of heavier elements, because of their importance in the nucleosynthesis of the light elements (P64d), were never carried out.

A program was initiated by Viswanathan for the study of radioactivity induced by cosmic-ray muons in natural materials. Of particular interest were ^{14}C and ^{26}Al , which could give information about exposure conditions in the past, and ^{37}Ar and ^{22}Na , which would give production-rate information (P64j, P65b). Plans were made to study the production of ^{22}Na , ^{37}Ar , and other short-lived nuclides by the negative-muon beam of the Carnegie synchrocyclotron (P65j), and bombardments were initiated (P66i). A gas-extraction system for ^{37}Ar and ^{14}C was built and tested (P65m), as were small-volume counters for Ar radioisotopes (P65n). Chemical procedures for separation of radiosodium from large amounts of Mg were sought (P66i). Viswanathan then elected to do his thesis work in another area, and the challenge was taken up by Pérez (P67c), but he, too, abandoned it for work requiring less arduous developmental preliminaries.

Because cosmogenic nuclides in meteorites are produced both by the primary and secondary cosmic rays, a number of "simulated-cosmic-ray-bombardment" experiments were carried out, using very thick targets exposed to proton beams from various accelerators. In most of these, thin target plates were placed at intervals between thick matrix plates of steel (representing iron meteorites) or plate glass (representing stone meteorites). Except in some of the low-energy experiments, the target plates were cut into radial zones for analysis. The measurements of a number of product radionuclides were made by radiochemical analysis, by gamma spectrometry, or both. These experiments were done by Goel, Shedlovsky, and Rayudu.

Bombardments of thick iron targets were done with the McGill University synchrocyclotron at 100 MeV (P63b, P64b, P66j, J68b), the CIT synchrocyclotron at 440 MeV (P60f, P61q, P63e, J62a Appendix I), the University of Chicago synchrocyclotron at 450 MeV (P62n, P63c, P66j), the Brookhaven Cosmotron at 1 GeV (P63d, J64d) and 3 GeV (P62m, J64d, P65i) and the Lawrence Berkeley Bevatron at 6.1 GeV (P64a). In the 3-GeV bombardment some measurements were made of ^7Be , ^{14}C , and ^{32}Si in the steel matrix plates (D62a, J64d; see P65i, J67c for corrections). Silicate-target bombardments were done at 1 GeV (P63f) and at 3 GeV (P63g). A composite target was bombarded at 100 MeV with measurements in glass, silicon, and several other materials (P64b, J68b).

Kohman and Bender reviewed all thick-target bombardment data and developed a two-component model and mathematical method for systematizing the data on measured radionuclides and estimating the parameters for other nuclides of interest in natural solid materials exposed to primary cosmic rays (P64e, P65g, J67c).

Theoretical calculations of cosmogenic production rates of specific nuclides in meteorites were made by Goel (P60q, J62a, D62a), by Kaye (D63a), and by Kohman and Bender (P65g, J67c).

B. Artificial Radionuclides

Early in the lifetime of this Project there were many empty spaces in the chart of nuclides, and it was hoped that some of these spaces might correspond to undiscovered nuclides which might be useful as radioactive tracers or which might

occur in nature. Radioisotopes of Mg, Al, Si, and Ti suitably long-lived for tracing were not known, and for Al there was not even the possibility of stable-isotope tracing. Plans were made to use the Carnegie synchrocyclotron to produce and characterize missing isotopes of these elements (P52f, P53d), and other nuclides which might have half-lives in the range for extinct natural radioactivity (P51d, P52d, P53c).

During searches by Jones for ^{25}Mg , ^{32}Si , and ^{44}Ti , these nuclides were discovered by others. We published a confirmation of ^{25}Mg and a description of its production by proton spallation of silicon (J53a). Jones determined half-life exclusion limits for ^{22}Mg , ^{26}Si , ^{30}S , ^{38}S , ^{52}Ti , ^{56}Cr , and ^{60}Fe in synchrocyclotron bombardments and for ^{60}Fe in double-neutron capture (D56b). (All of these nuclides have subsequently been discovered to lie outside of Jones' limits except for ^{38}S , which is barely inside his lower limit.)

In 1954 we became aware of experimental and theoretical indications that the known 7-second ^{26}Al was probably an isomeric excited state, and a search for a long-lived β^+ - and γ -emitting ground state in heavily deuteron-bombarded magnesium proved successful (J54f). The initial half-life estimate of $\sim 10^6$ y based on reaction-yield comparisons was uncertain by a factor of 10. Improved estimates of 1.1 ± 0.1 My (J57b) and 0.8 ± 0.1 My (P57a, D57a) were combined to yield a value of ~ 1 My, considered uncertain by a factor of 2 (J58b). A preliminary specific-activity determination using absolute activity measurements (P57a, D57a) and mass-spectrometric reverse-isotope-dilution analysis in collaboration with Professor H. Hintenberger of Mainz yielded a half-life of 0.62 ± 0.31 My (D57a, J58b), and a definitive determination by this method yielded 0.738 ± 0.029 My (J58d). The disintegration scheme was worked out by a careful series of instrumental measurements (P57a, D57a, J59a). (A recent slight improvement in the β^+ branching fraction and recalculation from our specific β^+ activity has yielded a revised half-life of 0.716 ± 0.032 My.)

When this nuclide was discovered, it was noted that hitherto aluminum had been the only element without either a radioactive or stable isotope suitable for tracing (J54f), and it was considered that tracer applications of ^{26}Al would probably be its greatest value (J58b). Although some uses as a tracer and as a constant radioactive standard have been made, its occurrence as a cosmic-ray-induced radionuclide in meteorites, first discovered in work of this Project (J58c, J58f), has become of even greater importance. (Natural ^{26}Al has now been observed also in ocean sediments from accumulated cosmic dust and in lunar surface materials, and there are even indications of excess ^{26}Mg from extinct ^{26}Al in minor components of some meteorites, possibly dust grains formed in cooling supernova shells.)

Following negative results in the search for ^{60}Fe (P52d, P53d, D56b), a positive observation of this nuclide was achieved by Roy through extraction of the $^{60\text{m}}\text{Co}$ decay product from iron separated from a copper target which had received heavy 400-MeV-proton bombardment (P55b, P56a, J57a). From comparison with the ^{59}Fe yield, the ^{60}Fe half-life was estimated as $\sim 3 \times 10^5$ y, uncertain by a factor of 3. After the ^{59}Fe in this sample had decayed away, the ^{60}Fe was confirmed by Merz through $^{60\text{m}}\text{Co}$ extraction (P59a). The low-energy-photon spectrum showed a strong continuum due to ^{55}Fe electron-capture inner bremsstrahlung, the expected 59-keV gamma from the $^{60\text{m}}\text{Co}$ isomeric transition, and a strong peak at 27 keV, which was attributed to a prompt gamma of this energy following the ^{60}Fe beta

emission. The beta spectrum was measured with an anthracene scintillator and found to have an end-point at 135 keV. A disintegration scheme was deduced involving an energy level in ^{60}Co at 86 keV. An improved half-life of $\sim 1.0 \times 10^5$ y was estimated by a comparison of the ^{60}Fe and ^{55}Fe activities (P60h; see J61a). (The 27-keV gamma has not been accepted by the Oak Ridge Nuclear Data Group because there is no other evidence for a ^{60}Co level at 86 keV. Unfortunately, the ^{60}Fe sample has been lost, so the measurements could not be repeated and extended.)

Roy and E. Perlstein of the Physics Department obtained indications by extraction of the Co decay products that ^{61}Fe and ^{62}Fe have half-lives of 6 and 3.5 minutes, respectively (P56a). (The ^{61}Fe half-life has been confirmed elsewhere, but the ^{62}Fe half-life is unconfirmed.) Roy obtained a similar indication of a 6-minute half-life for ^{38}S by ^{38}Cl extraction (P56a), but this has subsequently been found to be in error (the currently accepted half-life being 2.84 hours).

The nuclide ^{74}Kr with half-life 20 ± 1 min was discovered and characterized by Gray, Hamster, and Caretto, who also obtained an indication of ^{75}Kr with half-life $\leq 0.8 \pm 0.4$ min (P59a, P60g, J60a). (The present accepted half-lives of ^{74}Kr and ^{75}Kr are 16 and 3.5 min, respectively.)

In 300-400-MeV-proton bombardments of Ta, W, and Lu, Merz and Caretto observed and characterized three pairs of neutron-deficient Hf-Lu isobars: ^{170}Hf (9 h) $\rightarrow$ ^{170}Lu (1.9 d); ^{169}Hf (1.5 h) $\rightarrow$ ^{169}Lu (1.5 d); ^{168}Hf (22 min) $\rightarrow$ ^{168}Lu (7.0 min), and obtained inconclusive evidence for a fourth: ^{167}Hf (~ 10 min) $\rightarrow$ ^{167}Lu (~ 4 min) (P60f, J61e). Except for ^{169}Lu a d ^{170}Lu , all of these discoveries were original. (All have been confirmed, except that the ^{169}Hf half-life is now given as 3.25 min. The chain of presumed mass number 167 has been reassigned to 166.)

Merz also made searches for a number of other nuclides, some of which might be expected to be long-lived and thus of cosmological interest. In proton-bombarded Pb and Au, a 1.4-hour Hg decaying by electron capture to a 1.7-hour Au EC and β^+ activity was observed. These were assigned to ^{190}Hg and ^{190}Au . Since other workers had assigned another Hg-Au pair to mass number 190, a definitive experiment was planned (P60i). (No one else has reported these half-lives among the many Hg and Au activities now known. They may well be isomeric states of known nuclides.) Negative results of a search for ^{206}Hg from the $^{208}\text{Pb}(p,3p)$ reaction by 4.2-min ^{206}Tl extraction yielded exclusion limits of 15 min to 100 y, but with a tentative positive indication of a half-life of ~ 140 y (P60j). (This experiment was apparently not continued, possibly because of the identification elsewhere of an 8-min ^{206}Hg . However, it is quite possible that a long-lived isomer might also exist.) A similar search for ^{182}Hf from the proton bombardment of W by extracting Ta and looking for 16-min and/or 112-day ^{182}Ta led to an exclusion range of 20 min to 2000 y (P60j). The upper limit was later raised to 10^4 y, but the search was discontinued when two other groups reported successful searches and a half-life $\sim 8 \times 10^6$ y (P61i). (A 65-min isomer of ^{182}Hf has been reported subsequently.) Evidence was found in a similar experiment with a short bombardment for the chains ^{183}Hf (1.0 ± 0.1 hr) $\rightarrow$ ^{183}Ta (5.0 da) and ^{184}Hf (2.2 ± 0.1 hr) $\rightarrow$ ^{184}Ta (8.7 hr). The Ta activities and ^{183}Hf had been known already (P61i). (^{184}Hf has been reported by others to have a half-life of 4.1 hr.) By periodic extraction of Mn from purified Cr from a proton-bombarded Co target, 2.6-hour ^{56}Mn was observed with an indication that the ^{56}Cr parent had a half-life

Of 7.2 ± 0.5 min. It was then learned that others had observed ^{56}Cr directly and given a more accurate half-life of 5.9 ± 0.10 min (P61h). From the Ti fraction from irradiated Mn it was possible to extract the 3.8-minute ^{52}V . In the Ti itself a 12-minute beta component was present in the gross beta activity and also in the negative electrons separated by a permanent magnet. In addition, the 1.43-MeV ^{52}V gamma was observed in the Ti fraction decaying with a 12-min half-life. This indicated that ^{52}Ti has a 12-min half-life and is the parent of ^{52}V (P61h). (Current compilations include a 1.7-min ^{52}Ti , apparently well characterized. Perhaps the 12-min and 1.7-min activities are isomers.)

An early objective was a search for electron-capture decay of ^{204}Tl . If it were stable against this process, and ^{204}Hg were long-lived, the assumption of geochronology that ^{204}Pb in nature is completely non-radiogenic would be questionable. A proportional-counter x-ray spectrometer, with which electron-capture x-rays and internal bremsstrahlung might be detected, was constructed by Mulholland (P51e), but the electron-capture decay of ^{204}Tl was discovered elsewhere before he was able to perform the experiment.

Morrison began an experiment to determine the β^+ branching ratio of 4.3-hour ^{129}Sb to the 34-day metastable state and the 72-min ground state of ^{129}Te (P58a, P61j). (This experiment does not seem to have been completed. The ^{129}Sb decay scheme is extremely complicated and still not completely unscrambled, but the branching ratio is now fairly accurately established.)

The half-life of ^{53}Mn , whose measurement in accelerator targets is made difficult by overwhelming amounts of ^{54}Mn , was estimated from meteorite observations to be 1.9 ± 0.5 My (P63n, D63a, P64h, D64a, J65a). (Some more recent and more direct measurements have yielded 2.9 ± 1.2 , 3.7 ± 0.4 , and 3.9 ± 0.6 My.)

Rayudu participated with S. Jha and others of the Physics Department in the determination of the decay scheme of the 1.25-min ^{82}Sr (J64f); Rayudu performed the radiochemical preparation of Sr from a proton-bombarded Rb target (P64d). He collaborated similarly in many other disintegration-scheme studies (P64d) (for which he received no recognition by co-authorship in any of Jha's publications).

Theoretical considerations published by the writer in 1943 indicated the likelihood that alpha activity should be a rather widespread phenomenon in highly neutron-deficient isotopes of medium-heavy elements (rare-earth elements to lead). One of the original objectives of this Project was the search for and study of such activities using high-energy-proton bombardments. Although this objective was confirmed from time to time (for example, P52f), other activities completely occupied available personnel until 1958-59, when Merz, Strohal, and Macfarlane initiated some radiochemical studies of heavy-element targets bombarded in the Carnegie synchrocyclotron (P59a, P60k). Although for the most part only some previously reported alpha activities were observed, there was an indication of a rare-earth alpha activity of ~30-hour half-life and ~3.3-MeV energy. Since it was observed in ^{86}Er and heavier targets but not in ^{85}Tb , it was surmised that it must be a new isotope of ^{85}Tm , ^{86}Er , or ^{86}Ho . Unfortunately, this lead was not followed up. (A 13-hour activity with 3.77-MeV alpha particles has been observed and assigned to ^{154}Dy . However, ^{154}Dy should have been produced in Merz's work by the reaction $^{159}\text{Tb}(p,6n)$. Moreover, there are other puzzling features about the 13-hour activity and its assignment.)

In 1961-62 Mesojedec began the design and construction of a system for measuring half-lives and energies of very short-lived alpha emitters (microsecond-millisecond range) by pulsed bombardments with the Carnegie synchrocyclotron (P62c, P63h). The solid-state alpha detector was to be in the cyclotron chamber near the target but not in the beam, and the associated electronics would measure both the energy and the time delay of each alpha particle detected. Although considerable progress was achieved, the system was not completed before Mesojedec left in 1964, and this approach was not pursued further here. (It has been employed quite successfully elsewhere.)

C. Primary Natural Radioactivity

1. Beta Activity

At the time of initiation of this Project, natural neodymium had been reported to emit soft β radiation, and the writer had given arguments for believing that ^{150}Nd was unstable, in which case ^{150}Pm would accompany it in nature. Mulholland used a large proportional counter to look for natural radioactivity in highly purified neodymium, but could not find any (P51b, P52a, J52a, D52a). The possible instability of ^{150}Nd was discussed further, but plans for a more intense search (P53a) were not carried out. (Present data indicate that the ^{150}Pm β -disintegration energy is probably slightly greater than the ^{150}Nd - ^{150}Sm mass difference, so ^{150}Nd is probably stable.)

The odd-odd nuclide ^{138}La had been found as a natural isotope of lanthanum, and γ - and x-radiation had been found about the time we undertook work on it (P51b, P52a). Mulholland confirmed the x-radiation and discovered β^- radiation (P52a, D52a, J52d). Selig made a more accurate measurement of the specific K x-ray intensity and gave an upper limit to the specific L x-ray intensity (J54d, D54a).

There were (and still are) cogent reasons for believing that ^{48}Ca is unstable relative to β^- decay to ^{48}Sc . Although the method of active-daughter extraction would be the most sensitive way of observing this transition (see Subsection 2), it seemed possible that the decay could go through an unknown isomeric ^{48}Sc , bypassing the ground state. Accordingly, a direct search for β radiation from Ca was made by Selig, but with negative results, practically eliminating the possibility of a low-spin ^{48}Sc isomer (P53a, J54d, D54a). (Excited states of ^{48}Sc accessible from ^{48}Ca are now known, but their spins are close to that of the ground state.)

A similar case is ^{96}Zr , which seemed (and still seems) unstable relative to ^{96}Nb . Selig made a direct search for the resulting β radiations, but with negative results (P53a, D54a). Several kilograms of reactor-grade zirconium metal were placed around a NaI(Tl) scintillator; some gamma radiation was observed, but of undetermined origin (Simanton, P56b). The zirconium was sent to Kaye (one of our Ph.D. alumni) at Batelle Pacific Northwest Laboratories in 1969, who measured it in a low-level γ - γ -coincidence counter recording also in a singles mode. Preliminary results indicated no prominent γ radiation, but were inconclusive about weak radiation (private communication).

Another even-even nuclide considered as possibly unstable was ^{100}Mo ; if it decayed to ^{100}Tc , it might explain the observations of Tc in some stars. But

Selig could find no β activity in purified molybdenum (P53a, J54d, D54a). (^{100}Mo is now known to be stable.)

Selig looked for β particles and x-radiation from the odd-odd nuclide ^{50}V without success, obtaining longer limits for the EC and β^- half-lives than those of prior investigators (P53a, J54d, D54a). (Both of these transitions have been observed subsequently, with even longer half-lives.)

An attempt to detect the expected β^- radiation of ^{113}Cd was also unsuccessful (P53a, J54d, D54a). (The transition has subsequently been found with a half-life only slightly greater than Selig's lower limit for the energy available.)

At that time the relative masses of ^{123}Sb and ^{123}Te were unknown, and Selig examined Sb for β^+ activity and Te for L x-rays. In each case only lower half-life limits could be set (P53a, J54d, D54a). (^{123}Te has subsequently been found to undergo electron capture to ^{123}Sb .) The Te measurement also set a limit to possible ^{130}Te β^- decay via ^{130}I (which is now known to be energetically impossible).

The writer (J54c) discussed the phenomenon of "bound-beta decay" in connection with an apparent discrepancy between ^{87}Rb - ^{87}Sr ages and those based on U and Th alpha decay. He suggested that the half-life of ^{87}Rb might be considerably shorter than the then-current value of 62 Gy based on β^- -particle counting, and that transitions in which the β^- entered an unoccupied atomic orbital instead of the continuum might account for the discrepancy. (The discrepancy has been eliminated to the accuracy of each type of measurement, a few %, and there is now no reason for believing that bound-beta decay is quantitatively important in this nuclide. But read on.)

Some measurements were made of the β^- radiation of ^{187}Re , which was believed to have a maximum energy of ~1 keV. The radiation was confirmed, with an indication that the maximum energy is ~800 eV. At amplitudes corresponding to very low energies in the proportional counter operated at very high gain, the pulse-height distribution showed a strong peak at ~25 eV and successively smaller peaks at several integral multiples of this energy. We interpreted the first peak as due to single electrons which entered the gas with insufficient energy or inclination to ionize the gas, and the other peaks to events in which the primary electron was accompanied by 1, 2, 3, ... secondary electrons. Because of the phenomenon of diffusion of electrons in nonconducting media (the source was Re_2S_7), all that could be said about the half-life was that it was $> 3 \times 10^9$ y (P55a, P56b, P56f, P57b; Simanton, Rightmire, and Kohman, incomplete manuscript). (Subsequent workers have published β^- end-points up to 2.6 keV, which is accepted. However, there is still a considerable disagreement between the half-life values calculated from β -particle counting and from radiogenic ^{187}Os in Re-containing minerals of known age. Bound-beta decay could be an important phenomenon in this nuclide.)

Many investigators, including Mulholland (D52a, J52d) had observed that commercial aluminum exhibits radioactivity. When the probable existence of a long-lived ^{26}Al became known, it was thought possible that it could be long-lived enough for a residual from nucleosynthesis to be extant. Rightmire obtained some 99.99% pure aluminum sheet and measured it in the large low-background proportional counter. The activity was only ~10% of that observed

by Mulholland. Since it was soon found that the half-life of ^{26}Al was only $\sim 10^6$ y, the activity was assumed to be due to residual impurities (P55a, D57a).

2. Search for Certain Natural Beta Activities by Active-Daughter Extraction

The writer had proposed in 1948 that certain naturally occurring and apparently stable even-even nuclides near the limits of beta stability might actually be energetically unstable but with very long half-lives. In such a case, the immediate odd-odd decay product would be an energetic and short-lived β emitter. Chemical extraction of the daughter element from a large quantity of the parent element and measurement in a low-level radiation detector would then provide an extremely sensitive method of detection of the parent activity. The writer had previously applied this technique to Ca^{48} via Sc extraction, with negative results, setting a half-life limit of $\geq 5 \times 10^{15}$ y. A more intense search was planned (P51c).

Jones used 100 pounds of calcium nitrate (9 kg Ca), and found no detectable 44-h ^{48}Sc activity, from which the ^{48}Ca half-life is $\geq 2 \times 10^{16}$ y (J52c, P52b). In a smaller-scale experiment (0.5 kg Ca) with faster chemistry, he found no evidence for a shorter-lived ^{48}Sc isomer (56b).

Appelbaum noted nuclear-physical evidence that ^{48}Ca was unstable by 268 ± 17 keV, that the ^{48}Sc ground state has spin-parity of $6+$, and that an excited state at 112 ± 5 keV offered the possibility of a less-than-sixth-forbidden β -transition from ^{48}Ca . He carried out ^{48}Sc tracer experiments on the separation of Sc from saturated $\text{Ca}(\text{NO}_3)_2$ solutions, and found that $\text{Sn}(\text{OH})_4$ was a more suitable carrier than $\text{Fe}(\text{OH})_3$ when the precipitate was to be mounted directly for counting (P62p).

Miller developed chemical methods of purifying Sc coprecipitated from $\text{Ca}(\text{NO}_3)_2$ solutions by $\text{Fe}(\text{OH})_3$ (P63r).

Van Itallie discussed new nuclear data: the ^{48}Sc ground state has spin-parity $6+$ and lies 289 ± 12 keV below ^{48}Ca ; there are two accessible excited states of ^{48}Sc at 131 ± 2 and 253 ± 2 keV excitation, with spin-parities $5+$ and $4+$, respectively. The ^{48}Ca transitions to each should be fourth-forbidden, with estimated half-lives $\sim 10^{18}$ y and $\sim 10^{20}$ y, respectively, and all should populate the 44-h ground state (P65h, P66h).

Van Itallie, Allerton, and Rama Rao added a chemical step to eliminate ^{90}Y which could be present from contaminating fission-product ^{90}Sr . With their procedure and a Sharp Low-Beta counter they observed no ^{48}Sc and gave the half-life limit as $\geq 2.5 \times 10^{18}$ y (P67i). This was later corrected to $\geq 2 \times 10^{17}$ y (P68j).

Pang used a Pu-Be neutron source to induce ^{47}Ca and ^{49}Ca activities in the solution; these decay to ^{47}Sc and ^{49}Sc in the same chemical form as any ^{48}Sc produced from the ^{48}Ca , and are good internal tracers. She found that in the previous procedure Sc was not coprecipitated well with $\text{Fe}(\text{OH})_3$, but that this became nearly quantitative at pH ~ 8.5 (P68h).

The most intensive study and improvement of the procedures was made by Yeager, who found many unexpected complications. He finally developed what seemed to be a satisfactory procedure for use with 100 kg Ca. However, in application, a small amount of activity behaving like ^{210}Pb , generating ^{210}Bi was observed; from the

absence of discernable ^{44}h decay, a limit of $\geq 5 \times 10^{17}$ y was set for the ^{48}C half-life (P70b, P71f). The procedure was further refined to eliminate Pb and to speed up the separation. But three final applications yielded quite high activities which grew for several days and then decayed with apparent half-lives of about a year. It was suggested they were ^{210}Pb - ^{210}Bi plus ^{46}Sc from contamination or cosmic-ray effects. It was concluded that β detection was not specific or sensitive enough for this purpose (P72j).

Parallel work elsewhere was noted in which β - γ -coincidence detection was used, yielding a half-life limit of $\geq 1.3 \times 10^{19}$ y (1967) and $\geq 9 \times 10^{20}$ y (1969, unpublished). Yeager suggested that we use γ - γ -coincidence counting (P71f) in the sum-coincidence mode, which should be remarkably sensitive and specific for ^{48}Sc ; this would also allow simpler and faster chemistry (P72j). For this and other applications, Ritter set up our low-level γ - γ -coincidence system having two 3-in X 3-in NaI(Tl) crystals and a plastic-scintillator anticoincidence guard (Section M) in a sum-coincidence mode, and partially tested it with ^{22}Na sources (P72m). The Ca solution, handling equipment, and detector system are available for continuation of this work.

Of a number of other even-even nuclides near the limits of beta stability whose actual stability had been suspected (P52b, P53a), only ^{96}Zr seems to be definitely unstable. The situation is surprisingly similar to that of ^{48}Ca : the ^{96}Nb ground state has spin-parity $6+$ and lies 180 ± 6 keV below ^{96}Zr ; there are two accessible excited states of ^{96}Nb at 43 and 142 keV, with spin-parities $5+$ and $4+$, respectively; the ^{96}Nb transitions to each should be fourth-forbidden. A third state at 180 keV and spin-parity $3+$ might also be accessible, the transition being second-forbidden. All transitions should populate the 23-hour ground state. A direct-counting search for the ^{96}Zr - ^{96}Nb γ activity is described above. If this or the ^{48}Ca experiment yields positive results, this case should then be examined by the active-daughter technique.

3. Alpha Activity

A major initial objective of the Project was the search for additional cases of natural alpha activity in medium-heavy elements. Initial work was done with nuclear emulsions. Selig and Waldron observed a slight indication of ~ 3 -MeV α activity in ^{204}Pb (P51a, P52c, P53b), but a limit of $\geq 3 \times 10^{16}$ y for the half-life was reported (J54d). (Others later reported 2.6-MeV α -particles from ^{204}Pb with a half-life of 1.4×10^{17} y. However, nuclear masses now indicate that only 1.94 MeV is available for the ^{204}Pb - ^{200}Hg transition, so the α -activity should be completely unobservable.)

The proportional-counter measurement of neodymium made in search of β activity by Mulholland also set a low limit to possible alpha activity of the element (J52a, D52a). Using nuclear emulsions, Waldron and Schultz did observe α -particles from neodymium, which were attributed to ^{144}Nd (P53b, J54a). The assignment was confirmed with enriched ^{144}Nd (P55a). Macfarlane confirmed the activity using a large internal-sample ionization chamber and obtained a more precise energy and half-life (P58b, P59b, D59a, J61c).

Macfarlane also obtained a more precise α energy and half-life for the well known ^{147}Sm (P59b, D59a, J61c). He was unable to detect any α activity in ^{148}Sm and ^{149}Sm using enriched isotopes of each. (He and a student have subsequently found and characterized the ^{148}Sm activity.) Absence of ^{146}Sm

in natural samarium was also confirmed. (Its half-life is now definitely known to be too short.)

Preliminary measurements of cerium seemed to confirm an alpha activity which had been reported by others (P58b), but later measurements set a much lower limit (P59b, D59a, J61c). (^{142}Ce , the isotope in question, is now known to have too little energy for observable activity.)

The alpha activity of platinum, known since 1921 but forgotten until it was again "discovered" in 1954, was confirmed, the assignment to ^{190}Pt established by use of the enriched isotope, and more precise energy and half-life values determined (P57b, P58b, P59b, D59a, J61c). A search for lower-energy alphas from ^{192}Pt using a larger counter gave negative results (P58b).

Alpha activity was found in enriched ^{174}Hf (P58b, D59a, P59b), but was also found by others before our publication of more accurate energy and half-life (J61c).

Experiments with nuclear emulsions did not reveal α activity in gadolinium (P53b, P55a). With the proportional counter, however, a definite activity was found in ^{152}Gd (P58b, P59b, D59a, J61c). The specific results were in disagreement with a parallel claim published in 1959.

No activity could be found in ^{180}W , in conflict with a claim in the literature. Likewise, a search for α particles from ^{196}Hg yielded a negative result (P58b, P59b, D59a, J61c).

4. Anomalous Alpha Activity

Observations of radichalos in minerals have revealed several types which cannot be explained on the usual basis of incorporation of known long-lived α -emitting radionuclides in minute accessory-mineral inclusions in major rock-mineral crystals. The anomalous types, studied recently particularly by R. V. Gentry, include giant halos (with radii corresponding to ranges of 9-15-MeV α -particles), polonium halos (containing only spheres of radii corresponding to ^{210}Po , ^{214}Po , and ^{218}Po , the latter two with their α -emitting descendants), dwarf halos (corresponding to 2-3-MeV alphas), and "type Y" halos (corresponding to a single α energy of 6.55 MeV). Among the many reports in the literature of unidentified α -activity is a 4.4-MeV peak in the α spectrum of thorite and huttonite isolated from Conway, New Hampshire, granite.

If the effects are due to natural α emitters, the latter must either be primary natural radionuclides or must have non- α emitting parents or ancestors which are primary natural radionuclides, because the rocks involved are not of extreme ages. All known macroscopically occurring elements of $Z = 1-83$, 90, and 92 have been well examined for α -activity, and the near-transuranium region $Z = 93-100$ has been thoroughly explored, so the unknown long-lived nuclides, if they exist, must almost certainly belong to other elements. There are two main possibilities: (1) the transbismuth region ($Z = 84-89$, 91), and (2) the "superheavy-element" region ($Z \geq 100$). Since the $Z = 84-91$ region has also been well explored, possibility (1) requires the existence of long-lived isomers of known nuclides. These might be shape or high-spin isomers. A remote possibility is (3) unknown long-lived isomers of macroscopic elements, especially ^{93}Bi and ^{92}Pb . In any case, the necessary ad-hoc nuclear properties are rather unusual.

Landes initiated experiments to try to find anomalous α emitters, particularly the 4.4-MeV and high-energy nuclides. Conway granite was ground and the high-density non-magnetic minerals concentrated. Alpha spectra were taken on samples prepared by acid dissolution and evaporation on Pt at both low and high temperatures, and on samples of the material volatilized in air in a fused-silica tube in a furnace, condensed in a cold extension, dissolved in acid, and evaporated on Pt. The latter technique should concentrate Pb, Bi, Po, At, Fr, Ra, and some superheavy elements. In all spectra obtained, only the expected known members of the ^{238}U and ^{232}Th series were seen, the 4.4-MeV and >9 -MeV regions showing nothing above backgrounds. The volatility samples were dominated by ^{210}Po at 600°C ; Ra and Bi were partly volatilized at 700°C (P70c). Landes' work was terminated by her graduation.

Trivedi continued the volatilization experiments. He used large pulverized whole-rock samples of Conway granite and evacuated the volatilization-condensation system. One sample was heated 4 days at 200°C and 2 days each at 400°C , 600°C , and 800°C . The first two samples showed strong ^{210}Po activity only. The third showed in addition small peaks at 3.9 and 5.0 MeV but of dubious statistical significance. The fourth showed ^{224}Ra and ^{226}Ra and their descendants plus excess ^{210}Po . Nothing was seen above 6 MeV (possibly because of system non-linearity?) (P71a). A second experiment was only partially completed before Trivedi's departure.

D. Extinct Natural Radioactivity

A theme throughout this Project has been the search for cases of extinct natural radioactivity, and the exploitation of the cosmological implications of those cases which have been discovered during this period. Two early reviews, written before any case was known, discussed possible manifestations and potential significance of the phenomenon (R54a, R56a).

Jones, who had previously indulged in radioactivity lifetime statistics, made a statistical survey of lifetime data for beta-labile nuclides near stability. He considered only closed classes of nuclides adjacent to or within the stability region, based on the even or odd numbers of protons and neutrons. In the half-life region of principal interest for extinct natural radioactivity, 10^{15} - 10^{16} s (~ 30 - 300 My), no beta-unstable nuclides were then known. He predicted that of the order of two were yet to be found (P51g, P53c, D56b). (Only one has been found: ^{92}Nb ; see below. However two alpha emitters are known in this region: ^{244}Pu , ^{146}Sm . The region of interest has been extended to shorter lifetimes, where there are several β - and α -labile nuclides. But not all nuclides with the proper half-life can produce identifiable natural effects (R56a)).

Even now only two cases of extinct natural radioactivity have been established with certainty: ^{129}I and ^{244}Pu . There is controversial evidence for a super-heavy nuclide generating a unique fission component of meteoritic xenon, and an indication that some meteoritic solids, probably formed far from the Solar System, bear evidence of extinct ^{26}Al .

In Section B are described attempts to produce and characterize a number of nuclides which might have half-lives in the ranges from $\sim 10^7$ to $\sim 3 \times 10^8$ years. None were found. However, as just mentioned, ^{26}Al , which was found in the course of our work, might be in this category, even though its half-life is less than 10^6 y.

In 1950 the chronology of the Earth, the Solar System, and the Universe was poorly known. It was felt that, in addition to meteorites, terrestrial minerals might be hosts for detectable amounts of the decay products of extinct radio-nuclides with half-lives somewhat too short to have survived from their formation to the present.

Initial efforts focussed on the unknown nuclides ^{60}Fe , ^{202}Pb , and ^{205}Pb (P51d). Geologically old minerals of iron and lead were collected (P52d). Nickel was extracted from some old iron minerals and mineral phases of iron meteorites by Donaruma and Lykos (P52d, P53c, J54e) and examined mass-spectrometrically by R. J. Hayden and M. G. Inghram at the Argonne National Laboratory, but no ^{60}Ni excesses were found (Private communications; see R56a). ^{60}Fe was used as an example of the major importance as a source of heat early in the history of the Earth of a nuclide of appreciable primordial abundance and disintegration energy and half-life ~ 100 My (R54a, R56a). After its half-life was found to be $\sim 1-3 \times 10^5$ y, calculations showed that Fe^{60} must have rivalled ^{26}Al as the major source of radiogenic heat in Solar-System matter from ~ 1 to several My after the termination of nucleosynthesis, but was probably of no importance after solid bodies were formed (J61a).

Plans to isolate mercury from old lead minerals (P52d, J54e) were dropped after it was found that ^{202}Pb has a half-life of $\sim 3 \times 10^5$ y. (Recent observations of variable $^{202}\text{Hg}/^{196}\text{Hg}$ in some fractions of some meteorites might conceivably be due to extinct ^{202}Pb , since the deviant ratios are always higher than the normal ones.)

Efforts were made to detect enhanced ^{137}Ba abundances in barium in old rare-earth minerals due to ^{137}La , at a time when only a minimum half-life for the latter nuclide was known. Saito determined Ba spectrochemically and isolated samples for mass-spectrometric analysis, which were made by Hayden and Inghram. No abnormalities were seen (P53c, J54e; see R56a). (The half-life was subsequently determined to be too short. It is now given as ~ 60 ky.)

A major effort was made to find enhanced ^{205}Tl abundances attributable to extinct ^{205}Pb activity, particularly after indications that its half-life must be quite long (1954; see R56a). Work on extraction of thallium from terrestrial lead minerals (P52d, J54e, P55d) succeeded when El-Badry isolated the traces of thallium from samples of Ivigtut and another terrestrial galena (P56d) and from several meteoritic phases (P57d). Chemical procedures (P58d) were described in detail by El-Badry (S60a). A suitable spectrochemical method for the determination of very small amounts of thallium was developed by E. S. Hodge and W. K. Baer of the Mellon Institute Spectroscopy Laboratories, and applied to aliquots of El-Badry's isolates (S60b). El-Badry and Kohman discussed the cosmochemistry of thallium and the possibility of observing radiogenic ^{205}Tl (S60c; see J61a). Aliquots of the isolated thallium samples were held for mass-spectrometric examination, but difficulty was experienced in getting them run elsewhere (P59c). After the installation and tune-up of our own mass spectrometer, these were the first samples analyzed by Ostic. All samples were identical in $^{205}\text{Tl}/^{203}\text{Tl}$ to terrestrial reagent thallium, with about half the uncertainty of a similar series of measurements made elsewhere in the meantime. Literature values of the lead contents of the meteorites were used, and an "isochron" method using ^{204}Pb as a reference nuclide was used to interpret the results in terms of the chronology of nucleosynthesis and meteorite

formation. The desirability of determining both thallium and ^{204}Pb in the same samples in future work was indicated (P67f, P68d, J69a).

In the meantime Tandon undertook the determination of thallium and lead in meteorites by activation analysis (P65d, P65e). A few successful Tl determinations in metal and troilite from iron meteorites were made (P66f, P67d). He made calculations on the possibility of measuring ^{204}Pb by fast-neutron activation to 62-hour ^{203}Pb , particularly with the University of Pittsburgh cyclotron (P66g), but this was not tested. Tanner also undertook activation analysis of thallium in iron meteorites (P67e), but obtained no results on this element.

Ostic and Kohman discussed plans for thallium-isotope studies of lunar materials (P67g) (some of which were later carried out with support from NASA).

Huey made more extensive calculations on fast-neutron-activation analysis of lead and extended them to thallium. He concluded that mass-spectrometric isotope-dilution analysis was the most feasible method for both Pb and Tl (P68g). He developed chemical and mass-spectrometric procedures for both elements, initially involving volatilization from a crushed sample in a stream of oxygen (P69d). He later found that volatilization in vacuo was more effective, developed a differential-heating technique, and further refined the procedures (P70e). Final procedures were developed and blanks were greatly reduced after the new clean room in the Mellon Institute Building was put into operation, and successful analyses for both elements in several meteorites were obtained (P71d). Huey finally determined the contents and isotopic composition of lead in 16 chondrites and one achondrite (D72a, P72c, J73a) and thallium in most of them (D72a, P72d, J72a). All thallium samples, including those from meteorites with favorable Pb/Tl ratios, were identical to terrestrial thallium. The $^{205}\text{Pb}/^{204}\text{Pb}$ ratio at the time of Pb-Tl fractionation in the formation of chondrites was established as ≤ 0.00009 , lower than the $^{129}\text{I}/^{127}\text{I}$ ratio of the time of their initial xenon retention. The interval between the termination of s-process nucleosynthesis and Pb-Tl fractionation was deduced to be at least 60 My and possibly exceeding 120 My, depending on the assumptions made (J72a).

Unfortunately, throughout the course of our interest in ^{205}Pb , estimates of its half-life have steadily declined, from ~50 My in 1956 to ~14 My in 1971 (J72a). This considerably reduces the prospects for manifestations of ^{205}Pb as an extinct natural radionuclide. It was indicated that iron meteorites might offer the best possibility for detection of $^{205}\text{Tl}/^{203}\text{Tl}$ variations due to ^{205}Pb (J72a).

Lancet discussed possibilities for ^{205}Pb effects in various types of meteorites and plans for a more intensive search (P73c). He developed chemical procedures for thallium and lead in which the initial separation is made by wet-chemical methods instead of volatilization (P73h) and tested them thoroughly for lead (P76d).

Considerable work has also been done in connection with ^{146}Sm , whose half-life was given in 1953 as ~50 My, and which undergoes alpha decay to ^{142}Nd . It was at first felt that natural chemical fractionations of these two rare-earth elements would be insufficient to produce detectable ^{142}Nd variations (J54b, R56a, J61a). As information on rare-earth fractionations in meteorites became

available, the precision of mass spectrometry was increased, and a half-life of 103 ± 5 My was announced in 1966, this view changed (see P67g). Selah summarized information on Sm/Nd ratios in terrestrial rocks and rare-earth minerals and in meteorites (P69e) and began the development of suitable chemical and mass-spectrometric techniques (P69e, P70a). The chemical techniques were improved further by Karol and Bansal (P71c, P72o). Lin took up this work in 1972-1973 (P73d) and studied the chemical separation of Nd and Sm (P73i) from a solution from which some other elements had been removed first (P73h). Complete chemical and mass-spectrometric procedures for these two elements alone, starting with silicate samples, have been developed by Lin, with present interest mainly the ^{147}Sm - ^{143}Nd geochronology, but these techniques could also be used for the ^{146}Sm - ^{142}Nd system (P76b).

In 1960 Merz noted Russian reports of enhanced ^{235}U in some minerals, attributed to an extant transuranium ancestor, possibly ^{247}Cm , which was known to be long-lived. He attempted verification by extraction of uranium from two rare-earth minerals ~1 Gy old. The isotopic composition was measured by H. D. Cook at the Westinghouse Bettis Atomic Power Laboratory, who found the $^{235}\text{U}/^{238}\text{U}$ ratio to be normal to <1% (P60r; see J61a). Sarma proposed a further study, using α -particle spectrometry for determining the $^{235}\text{U}/^{238}\text{U}$ ratio (P61s). In 1963 the ^{247}Cm half-life was found to be 16 My, so this nuclide must be extinct. Since the half-life is very close to that of ^{129}I , effects in nature might nevertheless be observable. The writer considered this in detail, suggested that neodymium could be used as an indicator of the geochemical distribution of curium, and proposed a modified isochron equation (P69f). This is still regarded as a prospective case of extinct natural radioactivity.

In 1954-55 ^{182}Hf , which should generate stable ^{182}W , was suggested as a possible extinct natural radionuclide (P55d), and it was pointed out that there should be terrestrial minerals with favorable Hf/W ratios (R56a). Merz reconsidered this in 1958-59 (P59c). After the half-life was found to be ~9 My, the consideration was restricted to meteorites, and it was pointed out that stone meteorites should have favorable Hf/W ratios for enhancement of ^{182}W (J61a). An isochron equation and hypothetical meteorite isochrons were presented, and the necessary conditions for observation were discussed; it was pointed out that Hf-W fractionations may have been established before Xe-isotope retention began, a favorable factor in all cases involving non-volatile decay products (P69f).

It has recently (1974) been found that the long-known short-lived ^{92}Nb is a metastable state, and that the ground-state has a long half-life of ~170 My. This makes it an extremely interesting possibility for detection through ^{92}Zr enhancements. Although Nb/Zr ratios are generally unfavorable in natural materials, certain meteoritic and lunar minerals, especially rutiles, have surprisingly high Nb contents and/or Nb/Zr ratios. If the uncertain half-life should turn out to be somewhat longer than the value mentioned above, there is even the possibility of terrestrial manifestations. This has been discussed thoroughly in the last Progress Report (P76e).

Following our discovery of the long-lived ^{26}Al , a suggestion was made that this nuclide might have been important for heating and melting the parent bodies of meteorites. The writer presented quantitative arguments which made this seem unlikely (R56a). After the half-life was more carefully determined, quantitative calculations of the heat generation by all known nuclides with half-lives $\geq 10^5$ y

were made on the basis of various nucleosynthesis-chronology assumptions. In all models considered, ^{26}Al was indeed the dominant contributor from ~ 1 to several My after the termination of nucleosynthesis. Only, however, under "mixed" nucleosynthesis (continuous nucleosynthesis plus a single-event contribution by the last contributing supernova) could ^{26}Al have been important, and only then if solid bodies were formed within ~ 10 - 15 My after the last supernova's contribution (J61a). (It has been proposed that ^{26}Al produced by the "x process" in the solar nebula might have played such a role, but this has not been universally accepted.)

It has generally been considered that ^{26}Al was too short-lived for generation of observable ^{26}Mg anomalies. In 1969 there was a report of such observations in meteorites, attributed to protosolar x-process ^{26}Al . The writer presented an isochron equation expressing the situation in which both galactic and protosolar ^{26}Al synthesis may have occurred, and presented hypothetical isochrons for the cases where each dominated. It was suggested that even if the ^{26}Mg excesses were confirmed, they could be due to the action of high-energy particles on meteoritic solids either before or after incorporation in their parent bodies (P69f). (The ^{26}Mg anomalies mentioned have been disproven, but new Mg-isotope anomalies have been reported in high-temperature aggregates in the Allende carbonaceous chondrite. ^{26}Mg excesses may be due to extinct ^{26}Al , possibly in pre-solar solids formed near a site of nucleosynthesis; but there also seem to be some relative deficiencies of ^{26}Mg , and high energy irradiations are considered an alternate possibility.)

^{107}Pd , whose half-life has long been known as ~ 7 My, has occasionally been claimed to have produced ^{107}Ag anomalies. This was discussed (J61a), and ^{107}Pd was briefly considered as a prospect for lunar manifestations (P67g). (The Pd abundances and Pd/Ag ratios are probably too low and the time relationships unfavorable.) ^{53}Mn was mentioned as another possibility (J61a). Many other possibilities were mentioned in an early review (J56a), before their discovery or half-life determinations. (In nearly all cases the half-lives are now known to be too short.)

The writer discussed the possible value of extinct natural radioactivity in terrestrial geochronology (J54b, R54a, R56a). It is now evident that for a nuclide to be important for this purpose its half-life would have to be at least ~ 200 My, so that technically it would be not quite extinct (see P76a).

The first case of extinct natural radioactivity discovered, ^{129}I , was utilized by the writer in a discussion of the chronology of nucleosynthesis mentioned above in connection with radiogenic heat in Solar-System matter (P60s, J61a, J61b).

Following the discovery of the second case, ^{244}Pu , Huey and the writer used information on these two extinct nuclides and certain primary natural radio-nuclides to evaluate the parameters in several models for nucleosynthesis chronology (P70f, P71e, P72g, P76f). This is discussed in Section G.

E. Cosmic-Ray-Induced Radioactivity in Meteorites

In 1954-55 work was begun on a search for, measurement of, and interpretation of radioactivity induced in meteorites by cosmic-ray bombardment (P55c). Previously cosmogenic ^4He and ^3He had been observed, and at about that time cosmogenic isotopes of neon and argon were beginning to be observed. During the course of our work ^3He was found elsewhere.

Ehmann began by building small low-background beta counters (P55c) and developing chemical procedures for Be, Al, and some other elements (P56g). Identification of ^{10}Be and ^{26}Al in meteorites (P56c) was followed by observation of ^{60}Co (P57c). Possible applications to the study of meteorites, tektites, and impact craters were discussed (P57f). These studies constituted Ehmann's dissertation (D57b). A summary was presented at a conference in 1957 (J58c), and details of the chemical and instrumental procedures (J58e) and the results and their significance (J58f) were published. Ehmann believed he had also detected ^{10}Be and ^{26}Al in tektites (P57c, D57b, J58c, J58f); this is discussed below.

Improvement and extension of the chemical techniques were made by Nakagawa (P58c, P59d).

Goel undertook the detection and study of ^{14}C in meteorites (P59d, P60n). Chemical procedures were developed for extracting the carbon from silicates and metal as CO_2 , which was purified and measured in small proportional counters in the low-background assembly which had been used for terrestrial ^{14}C . Altogether, ^{14}C was measured or limits were determined in 9 stone falls, 8 stone finds, 1 stony-iron find, 2 iron falls, and 5 iron finds, including 3 associated with large craters. In the stone falls the ^{14}C was strongly correlated with the content of oxygen as the principal target element. Some stone finds had lower ^{14}C contents than the falls, and terrestrial ages or limits could be deduced; one was $\geq 21,000$ y old. Similar ages or limits could be deduced from $^{14}\text{C}/^{10}\text{Be}$ ratios for iron finds and the craters associated with some of them (P61k, J62b, J62c, P62f, D62a, J63a).

Goel also made extensive measurements of ^{36}Cl in 22 specimens of 10 iron meteorites (P60n, P61k, P62g, D62a). Information on the effect of depth in iron meteorites was obtained, and comparison with the depth effects for noble gases indicated that space erosion is significant. The cosmic-ray flux seemed to have been more intense in the remote past (J63b). (This evidence is not now regarded as convincing.) In addition to production by spallation of iron and nickel, appreciable ^{36}Cl is produced by neutron capture in meteorites containing chlorine (J64e).

Silicon was isolated by Goel from several iron meteorites and ^{32}Si looked for by ^{32}P extraction; the results were all negative (P60n, D62a). His theoretical calculations of production rates of radionuclides in meteorites are discussed below.

The number of cosmogenic radionuclides measured was considerably expanded by Kaye and Cressy, who were joined later by Chakrabartty, Shedlovsky (who had been engaged in similar work elsewhere), and Rayudu. Cressy constructed somewhat larger low-level beta counters for samples of low specific activity (P59d),

and with Kaye modified these and the large internal-sample counter for X-ray measurements (P60p). Kaye concentrated on iron meteorites (P61m, P61p, P62h, P63k, D63a) and Cressy on stones (P61l, P62i, P63l, P64h, D64a).

Kaye measured ^{10}Be , ^{26}Al , ^{53}Mn , ^{59}Ni , and ^{60}Co in 17 specimens of 9 irons. By intercomparisons of his results, those of Goel and others on other radionuclides, and thick-target bombardment data, he deduced the production rates as functions of depth. Neutron-capture formation of ^{36}Cl , ^{55}Fe , and ^{59}Ni was found to occur mainly in the resonance region. From his, Goel's, and Chakrabartty's results and some from the literature, he deduced terrestrial ages, upper limits, or lower limits for 10 iron meteorites, including 4 associated with craters. The iron find Idar had no ^{36}Cl (Goel) and low ^{10}Be , from which a terrestrial age of ~ 3 My was calculated. From the ^{53}Mn activity an estimate of its half-life was made. His ^{10}Be , ^{26}Al , and ^{53}Mn results were tabulated by Kohman and Bender (J67c). He also measured ^{53}Mn and ^{59}Ni in several stones with Chakrabartty (P61p, D63a).

Cressy measured ^{10}Be , ^{26}Al , ^{53}Mn , and ^{59}Ni in 13 stone meteorites and ^{36}Cl and ^{44}Ti in some of them. Most of them had been analyzed for ^{14}C by Goel. Intercomparisons gave information about depth effects on production rates. The data were used to improve the ^{14}C terrestrial ages of two meteorites. A relatively short cosmic-ray-exposure age (~ 3 My) of the hypersthene chondrite Kunashak was confirmed via ^{10}Be unsaturation; this and the ^{53}Mn unsaturation were used to estimate the half-life of the latter.

The combination of Kaye's and Cressy's observations on ^{53}Mn to yield the best half-life estimate (P63n, J65a) have been discussed in Section B.

Chakrabartty made important contributions to chemical procedures. He measured ^{10}Be , ^{26}Al , ^{40}K , and ^{60}Co in irons (P61n) and ^{10}Be , ^{26}Al , ^{60}Co (P61o), ^{53}Mn , and ^{59}Ni in stones, the latter two with Kaye (P61p). His results on ^{10}Be and ^{26}Al in irons were included in tabulations by Kaye (D63a) and Kohman and Bender (J67c). The ^{53}Mn and ^{59}Ni data were tabulated by Kaye (D63a). His work on tektites is discussed below.

During the course of this work samples of three freshly fallen meteorites were obtained, and measurements were made of several short-lived radionuclides in addition to the long-lived ones: the chondrites Harleton (P62j, D63a, D64a, J67a) and Peace River (D64a, J67a) and the iron Bogou (P63n).

Also, about this time, the Sputnik 4 satellite re-entered the Earth's atmosphere after 2.3 years in orbit, and a recovered steel fragment was obtained. Shedlovsky and Kaye measured a number of cosmogenic radionuclides in it. Low-energy solar-flare particles were deduced to have been important in their production. Some apparently non-cosmogenic ^{60}Co was also found (P63o, J63c).

During this period an unusual mesosiderite, Bondoc, was found, and a very low ^{26}Al content was measured elsewhere. Cressy and Shedlovsky made measurements of several radionuclides in the silicate and metal phases. The low ^{26}Al was confirmed, and activity comparisons allowed them to decide that the low activity levels were due to heavy shielding from cosmic rays rather than a great terrestrial age or short time as a small body in space (P63m, J65c).

Schell undertook to make extensive measurements of the ^{14}C and ^{39}Ar contents at various known positions inside of large iron meteorites (P64i). He tabulated all of the known large (> 1000 kg) meteorites of the World, large (> 200 kg) irons located in North America (P65f), and large meteorites which had been cut (P66e). An acid-digestion apparatus for dissolving meteoritic iron and a gas-handling system for sweeping out the CO_2 and Ar, with a metered amount of enriched ^{36}Ar as a tracer and contamination indicator, were built and partly tested (P65k, P65l, P66k). This project was never completed.

In Ehmann's initial work on ^{10}Be and ^{26}Al in meteorites, he made similar measurements on tektites, to try to settle the question of their terrestrial versus meteoritic origin. For an australite composite sample, ^{10}Be activity in the range of that observed for chondrites, and ^{26}Al at a somewhat lower level, were observed. For a sample of Libyan Desert silica-glass, somewhat lower ^{10}Be and considerably lower ^{26}Al were observed. Since these values were 3-5 times their statistical counting uncertainties, and they corresponded to a moderate terrestrial age for australites and a considerable age for Libyan glass, they were accepted as evidence for an extraterrestrial origin. Negative ^{26}Al and inclusive ^{10}Be activities were obtained for a moldavite composite and a bediasite specimen (P67c, P57f, D57b, J58f; see M58a). It appeared to the writer that only an extra-Solar-System origin could account for all of the known facts about tektites including their indicated long exposure to cosmic radiation (J58a). There were doubts, however, among others as well as ourselves, and additional measurements were undertaken (P60n, P60o). Chakrabarty was unable to verify the occurrence of ^{10}Be or ^{26}Al in any tektites. Some of Ehmann's Be and Al samples counted were re-cycled further, and the activities dropped or disappeared. Evidently Ehmann's samples had not been purified adequately, and should have been regarded as giving upper limits (P61o, P62k). Goel found null levels of ^{14}C in the tektites he examined (P62k, D62a). Rayudu made more extensive measurements on ^{10}Be and ^{26}Al and again obtained only null results (P62l, P63p, P64g). The data were summarized and presented by Rayudu at a conference in 1963, with the conclusion that these and other negative results on cosmogenic nuclides made an extraterrestrial origin quite unlikely (J63d).

Along with the observations and measurements of cosmogenic radionuclides in meteorites, calculations of production rates in space as a function of size and composition of parent body, position of sample within it, and cosmic-ray flux and spectrum were made. The measurement of certain high-energy-proton cross sections and of the distribution of products in thick-target bombardments, discussed in Section A, were an integral part of this program, and the resulting data were utilized in many of the calculations.

Goel was a veteran of such activity when he resumed it here (P59d). A five-step method was used. The first was to calculate, on the basis of cosmic-ray and bombardment data (including a 440-MeV-proton thick-iron-target bombardment here) the distribution of spallations ("stars") in iron along the beam direction. The second was to calculate by integration the distribution in spherical objects in an isotropic flux. A method of transforming the iron data to silicates was devised. The third was to calculate the fractional yield per "star" of various elements and sum over target elements. The fourth was to calculate the distribution among isotopes of each element of interest. The fifth was to sum over direct production of nuclides of interest and their short-lived beta-labile precursors.

Comparisons of calculated production-rate ratios with observed activity or abundance ratios showed order-of-magnitude to excellent agreement for irons, but some serious discrepancies for stones (P60q, J62a). Alternate models were considered, and depth dependences were calculated on the basis of more recent data, including some of his own data particularly on ^{36}Cl (D62a).

Kaye continued the calculations of position effects in iron meteorites, using especially the thick-target data of Shadlovsky and Rayudu (D63a). Similar considerations for stone meteorites were presented by Cressy (D64a).

The approach of Bender and the writer was based very heavily on the thick-iron-target data. Our method of representing the bombardment data by a number of physically significant parameters (P64e) has been mentioned in Section A. We devised methods for estimating the nuclide parameters for nuclides not measured but of interest, and of integrating the production rates for various depths in spherical targets or semi-infinite slabs in an isotropic primary flux. With the help of Sarker, the actual cosmic-ray flux-spectrum was represented by four groups of protons of energies corresponding to the thick-target bombardments (0.45, 1.0, 3.0, and 6.1 GeV). Families of curves for production rates within spherical meteoroids were presented for ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , and ^{53}Mn . In two-nuclide (^{10}Be - ^{26}Al , ^{36}Cl - ^{26}Al , ^{53}Mn - ^{26}Al) correlation plots, most iron-meteorite measurements fell within or near the calculated fields. There were, however some specific cases of disagreement, partly attributable to experimental errors and partly to over-simplifications of the model. Applicability of the approach to thick-silicate-target data, stone-meteorite data, and the lunar surface was discussed, as were Homework Assignments for improving the situation experimentally and theoretically (P65g, J67c).

The special case of ^{129}I in stone meteorites was considered by Kuo (P64k) and Edwards (P66d), under the assumption that barium was the most important target element, and by the writer (P67h), who considered all possible target elements. It was found that tellurium (suggested elsewhere) was the most important target element in all classes of chondrites and achondrites considered. All concluded that the production rates are so low that the accumulated ^{129}I would be extremely difficult to detect by neutron activation to ^{130}I in any case.

F. Radioactive Dating

A pervasive theme of this Project has been the application of radioactive dating methods to various areas of interest, terrestrial and cosmic. One aspect of this work has been to seek the nuclear bases for additional possibilities and, when found, to explore their applicability.

A critical review of the various methods known in 1954 was included in a general review written with Saito on Radioactivity in Geology and Cosmology (R54a). The possibilities and potentialities of extinct natural radioactivity were mentioned there and discussed in detail in a review specifically on that subject (R56a).

Much of our interest has been in meteorites, which have turned out to be extremely rich in information about themselves and the Universe as a whole, much of it involving radioactivity, nuclear reactions, isotopes, and nuclear properties. When the prospect of obtaining lunar samples arose, we were naturally interested

in becoming involved in nuclear studies of them (P67g). (The actual work on lunar materials was carried out in a separate project under a contract with NASA.)

1. Methods Based on Primary Natural Radioactivity

A variant of the ^{238}U - ^{206}Pb method, believed then to be original, was proposed in 1952. The specific ^{210}Pb activity of the lead from a uranium-containing mineral would be measured and would yield the U/Pb ratio without chemical analyses. The $^{206}\text{Pb}/^{238}\text{U}$ ratio would then be determined by mass-spectrometric analysis of the lead (P52a). Development of this method was undertaken by Saito, and considerable progress was made. But it was found that the method had been suggested in the literature in 1951, that its feasibility had already been demonstrated, and that a number of other laboratories were working on it (P53e). Since we did not have ready access to a mass spectrometer, work on the method was not continued after Saito's departure.

After the acquisition of our mass spectrometer, Ostic and Huey made plans for applications to meteorites and lunar samples. After Ostic's departure, Huey, who had concluded that mass spectrometry was definitely preferable to neutron-activation methods (P68g), developed techniques for the chemical separation of lead from silicate materials and its mass spectrometry. Initial separation by volatilization from a fused-silica boat in a fused-silica tube in a stream of oxygen (P69d, P70e) was replaced by volatilization in vacuo (P71d). Separation from thallium (also of interest; see Section D) and other elements was by ion-exchange columns. Lead blanks were reduced to ~10 ng following the use of the new dust-free laboratory beginning in 1970 (P71d, D72a, P72c). Pb contents were determined by isotope dilution, adding ^{208}Pb (and ^{203}Tl for thallium; see above) to the acid solution of volatiles. U and Th were not analyzed in this work, and the $^{207}\text{Pb}/^{206}\text{Pb}$ ratio was used as the chief indicator of age.

Sixteen chondrites and one achondrite were analyzed. All points on a $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ plot fell quite close to a straight line. From the slope of the best chondrite ^{207}Pb - ^{206}Pb isochron, the mean formation age (using the latest ^{235}U and ^{238}U half-lives) was 4.505 ± 0.008 Gy. The single achondrite, Norton County, lay significantly above the chondrite isochron and seemed to be ~50 My older (D72a, J73a). Using the mean of 3 slightly radiogenic meteorite leads, corrected for radiogenic ^{207}Pb and ^{206}Pb via literature values of their U contents, as initial $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$, single-stage model $^{207}\text{Pb}/^{206}\text{Pb}$ ages were calculated for the moderately and highly radiogenic meteorites. Distinctly different model ages were found, with a range of ~50 My about the mean (J73a).

It was recognized that uranium and thorium contents of the specimens would be desirable for interpretation of the lead-isotope information. Trivadi investigated the possibility of alpha-radiometric isotope-dilution analysis using ^{232}U and ^{228}Th as spikes for application to meteorites and lunar samples. The chemical procedure of Sarma (D64b; see below) for these two elements was adapted and partly tested (P70j). (Calculations showed that this method would not have the sensitivity and accuracy of mass-spectrometric isotope-dilution analysis using ^{235}U and ^{230}Th as spikes, and the development of the radiometric method was discontinued.)

This work was taken up Lancet (P73b), who modified the chemical procedures and extended them to include the isotope-dilution determination of U and Th as well as Pb. Samples were dissolved in toto using HF and other acids (P73h).

A distillation procedure for preparing extremely pure HF was developed in conjunction with Huey (P73h, J74a). Procedure blanks were reduced to ~3 ng for Pb and were ~0.02 ng for U and ~2 ng for Th. Several of Huey's volatilization residues were analyzed, and found to contain very little residual lead; but indications were that Huey's collection was not always complete. Two new samples of Huey's pulverized chondrites were analyzed. One (Queen's Mercy) was one for which Huey had regarded the mass analysis as substandard. With this corrected datum, the chondrite model ages now showed a trend of decreasing age with increasing metamorphic grade. This could be explained if post-accretional heating of the parent bodies produced not only the metamorphism but also some loss of volatiles including Pb, thus partially re-setting the U,Th-Pb clocks. It was noted, however, that this trend was not shown by similar data from two other laboratories - which did not agree with each other, either (P76d).

Lancet also analyzed a troilite nodule from an iron meteorite (Staunton), which nodule had been in a museum for some time, for U, Th, Pb, and Pb isotopes. The Pb was rather radiogenic, and moderate amounts of U and Th were present. A separate sample was ground and leached twice with acid. The first leachate was more radiogenic, the second leachate was only moderately radiogenic, and the residue was only slightly more radiogenic than the least radiogenic ("primordial") lead known. Evidently most of the radiogenic isotopes and some ^{204}Pb were due to common-lead contamination. This points the way to possible improvement in our knowledge of primordial lead (P76d).

Witter undertook to adopt the U,Th-Pb procedures developed for meteorites to terrestrial rocks, but did not obtain any results on natural materials before his graduation (P76a).

The writer prepared in 1970 a set of six computer-generated and -printed tables giving approximately 15,000 values of 15 uranium-thorium-lead decay-growth functions useful in geochronology calculations. These differed from previously published tables in that, in addition to functions pertaining to single-stage intervals ending at present, functions for multi-stage intervals between two past times were included. They were based on nuclear constants which had been in general use in geochronology for the preceding decade (P70b). They were reproduced as a special report (S70a) and given limited distribution. After new and improved ^{235}U and ^{238}U disintegration constants became available in 1971, the computer run was repeated in 1972 with the latest values of these and the ^{232}Th disintegration constant. In 1976 the tables were again computed using a slightly different value of the "natural" $^{238}\text{U}/^{235}\text{U}$ atomic ratio which had come into vogue. However, the widespread availability of scientific hand calculators greatly reduces the need for such tables of precomputed values, although when available they are useful (P76c). There are no plans for multiple-copy reproduction of the revised tables at present.

It was mentioned in Section B that there were difficulties with the physical determination of the ^{87}Rb half-life, important for ^{87}Rb - ^{87}Sr geochronology. Although bound-beta decay (J54c) no longer seems a significant factor, the softness of the β -particle spectrum limits the accuracy of specific-activity half-life measurement. The value currently used in geochronology, 50 ± 2 Gy, is based on measurements of the radiogenic $^{87}\text{Sr}/^{87}\text{Rb}$ ratio in terrestrial minerals whose ages were determined by U-Pb dating. Thus the ^{87}Rb half-life depends on

the ^{238}U and ^{235}U half-lives. Huey proposed that the ^{87}Rb half-life might be determined more accurately by using meteorites instead of terrestrial rocks, with the new U half-lives. He made illustrative calculations using his chondrite ^{207}Pb - ^{206}Pb data and literature data on Rb, Sr, and Sr isotopes in chondrites. The use of meteorite data resulted in greater internal precision, and the new U data lowered the ^{87}Rb half-life by about 1% (D72a). Because of many uncertainties in the calculation and the need for a dedicated effort with all precautions at the state-of-the-art, this result was not published.

Sextro developed the techniques of etching glass surfaces in HF to develop fission-fragment tracks and of identifying and counting them with a microscope. The objectives were to check the reported fission-track ages of tektites and impact glasses, to check the apparent agreement between certain tektites and geographically close impact glasses, to extend these measurements, and to try to apply the method to fragments of wind-borne volcanic-glass fragments found in deep-sea sediments, thus yielding a new dating method for them. Natural track densities were measured in several samples of indochinites, bediasites, and Libyan Desert silica-glass. Plans for the necessary neutron irradiations of the glasses were formulated (P65a). However no neutron irradiations were made, so no ages could be calculated.

In parallel with the effort to find evidence of extinct natural radioactivity of ^{146}Sm via ^{142}Nd anomalies (see Section D) a search was planned to detect ^{143}Nd variations attributable to ^{147}Sm α decay and to develop a ^{147}Sm - ^{143}Nd dating method. Chemical work was begun by Saleh in 1968 under the lunar-samples program, and the writer and Ostic discussed potentialities and calculated hypothetical isochrons in a report to NASA. Saleh continued this work under this Project with a focus on terrestrial rocks and meteorites (P69e). Gamma-emitting tracers ^{145}Sm , ^{145}Pm , and ^{147}Nd were used to develop the rare-earth-element separations. Mass spectrometry of Nd and Sm was undertaken, and ^{150}Nd and ^{150}Sm spikes for isotope dilution were calibrated (P70a). He assembled all published information on Sm/Nd ratios in terrestrial igneous rocks, sedimentary rocks, and minerals and in meteorites and displayed them in histograms. The Sm-Nd fractionations were found adequate for terrestrial dating, especially of rare-earth-rich minerals, and even of meteorites (P69e).

Further developments of the rare-earth-element separation techniques, including scaling down to small sizes and employing previously developed high-pressure ion-exchange chromatography, were made by Karol (P71c). Bessel and Karol developed a complete procedure, starting with HF-HClO₄ attack of solid silicates and ending with a clean-up of the separate elements for mass spectrometry (P72o). Some Nd mass spectra were run but not reported.

Lin took up the work with initial interest in both the ^{146}Sm - ^{142}Nd and ^{147}Sm - ^{143}Nd systems in meteorites (P73d). The chemical procedures were modified to use low-pressure ion-exchange separations, and were initially designed to start with a REE-containing solution from which Tl, Pb, U, Th, and Li had been removed (P73i; see P73h). The work is continuing, with interest mainly in ^{147}Sm - ^{143}Nd dating of both meteorites and terrestrial rare-earth minerals and comparison with U,Th-Pb ages. Considerable improvement in mass-spectrometric precision has been achieved, though it is not yet at the state-of-the-art (P76b).

In previous work the writer had deduced that ^{48}Ca was probably β -unstable and would decay through ^{48}Sc to stable ^{48}Ti , and he had established by the active-daughter-extraction method that the half-life was $\geq 5 \times 10^{15}$ y. If close to that limit, the possibility of a ^{48}Ca - ^{48}Ti dating method existed. The finding by Jones (Section B) that the half-life for this transition is $\geq 2 \times 10^{16}$ y (J52c), coupled with Selig's exclusion of alternative decay paths (D54a), indicated that ^{48}Ca possesses no geochronological value.

The occurrence of electron capture in the rare ^{138}La isotope (J52d, D54a) suggested the possibility of a ^{138}La - ^{138}Ba dating method (P52e). Saito isolated Ba from an ~1-Gy-old allanite with a favorable La/Ba ratio. The Ba, examined mass-spectrometrically by R. J. Hayden and M. G. Inghram at Argonne, did not differ from normal barium (P53e), indicating probable lack of usefulness of ^{138}La for geological purposes (R54a).

The possibility of a ^{40}K - ^{40}Ca dating method based on the dominant β^- branching mode of ^{40}K had been suggested and investigated exploringly several times. The ^{40}Ca variations are expected and were found to be small because of the ubiquity of Ca and the high isotopic content of ^{40}Ca in natural Ca. The writer, nonetheless, made an exhaustive survey of the geochronological potential of this dating system. Theoretical isochrons were calculated and plotted for moderate and large K/Ca ratios and for ages from 0.5 to 5.0 Gy. A survey was made of reported $\text{K}_2\text{O}/\text{CaO}$ mass ratios for nominally high-K minerals tabulated in the 5-volume "Rock-Forming Minerals" by Deer, Howie, and Zussman, and those were bin-tabulated according to K/Ca range. Evaporites, biogenic residues, "common" calcium, meteoritic minerals, tektites, and lunar-surface materials were also discussed. It was concluded that with modern mass-spectrometric techniques and choice of minerals, ^{40}K - ^{40}Ca dating should be feasible, particularly for Precambrian igneous and metamorphic minerals, possibly for meteorites, and probably for lunar materials. The availability and greater sensitivity of the ^{40}K - ^{40}Ar and ^{87}Rb - ^{87}Sr methods, which would be applicable to the same and more materials, greatly reduces the incentive for plunging into this method, but a preliminary experimental exploration of the applicability and limitations of this method of studying the fractionation history of these two major elements might be worthwhile (P69c).

2. Methods Based on Secondary Natural Radioactivity.

The ionium (^{230}Th) method of dating ocean sediments is based on the fact that thorium is less soluble in sea water than uranium, and thus ^{230}Th deposits in sediments in amounts greater than the amount which would be in secular equilibrium with the ^{238}U simultaneously sedimenting. Ideally, the excess decays exponentially with time as the layer gets covered deeper and deeper.

A radiochemical modification, conceived in 1955 (P55d), was developed by Almodovar beginning in 1956-57 (P57f). It is called the "thorium isotopes method" because all measurements are made on isotopes of Th. The $^{230}\text{Th}/^{232}\text{Th}$ activity ratio is determined by alpha-particle spectrometry. The $^{234}\text{Th}/^{228}\text{Th}$ activity ratio, which is equal to the $^{238}\text{U}/^{232}\text{Th}$ activity ratio, is determined from the time-dependence of the gross beta activity of the sample. The ratio M_5/M_{30} of the beta activity at 5 and 30 days after separation was calibrated to yield the $^{234}\text{Th}/^{228}\text{Th}$ ratio. The difference, $I = ^{230}\text{Th}/^{232}\text{Th} - ^{234}\text{Th}/^{228}\text{Th}$, is the excess of ionium over that in secular equilibrium with the uranium on a

thorium-normalized basis. It is related to the age T of the sediment sample by $I = I_0 \exp(-\lambda_{230}T)$, where I_0 is the effective value of I at the time of deposition. Effective chemical procedures, beginning with acid leaching of the sediments, were developed. Low-level beta counters were used to measure the beta activity while alpha-ionization spectrometry was performed on a smaller aliquot of the sample. The method was applied to 9 sections of a Pacific Ocean deep-sea core with excellent results. Approximate ages of sections showing excess ionium ranged up to ~640 ky. The occurrence of "pseudo-diffusion" in the surface layers, due to bottom currents and animal life, was noted. The method appeared capable of an ultimate range of ~800 ky (P58f, P59f, D60a, P60m). The work was done with the partial support of the Petroleum Research Fund administered by the American Chemical Society. A short version presented by the writer at a meeting of the Division of Petroleum of the ACS was published (J60b). Details of the radiochemical procedure were published by Almodovar (J65b).

Sarma undertook further work on sediment dating by radioactivity disequilibrium, also with the supplementary support of the ACS Petroleum Research Fund. He considered the ^{235}U - ^{231}Pa parent-daughter pair in addition to ^{238}U - ^{230}Th . He noted that ^{234}U is an intermediate in the latter transformation, and that recent work had shown that the $^{234}\text{U}/^{238}\text{U}$ activity ratio departs from unity in many natural systems. Therefore the ^{230}Th excess should be compared to ^{234}U instead of ^{238}U , as in Almodovar's method. He also wished to put all activities on an absolute rather than relative basis, and to make all measurements by alpha spectrometry, which is capable of greater precision than beta counting. The chemical procedures would have to isolate uranium and protactinium in addition to thorium, and he planned to use solid-state alpha detectors and electronics (P62d). After successful preliminary tests of the procedures, which involved total dissolution of the samples and incorporated ^{234}Th , ^{233}Pa , and ^{232}U tracers for yield determinations (P63i), he applied them to 36 sections of 4 deep-sea cores (2 Atlantic, 1 Pacific, 1 Indian Ocean). The complexities made it evident that complete sample dissolution was indeed necessary. Because of variability of ^{232}Th contents, mass normalization was found to be more meaningful than ^{232}Th normalization. The excess ^{230}Th specific activity was the most useful index of ages and sedimentation rates; in two cores reliable ages were yielded up to about 500 ky, and in favorable cases dating might be possible to ~700 ky. Ages from the excess ^{231}Pa activity and the excess $^{231}\text{Pa}/\text{excess } ^{230}\text{Th}$ ratio were less precise, limited to ~200 ky, and more affected by "pseudo-diffusion"; their greatest use was in indicating conditions of deposition. One core, from a high-deposition-rate region of the North Atlantic, showed deficits of ^{230}Th and ^{231}Pa , and a sedimentation model was developed to account for this. One core dated what had been regarded by some as the Pliocene-Pleistocene boundary at 525 ± 20 ky (D64b, P64f). (This core was later omitted from those believed to contain the Pliocene-Pleistocene boundary. The apparent "boundary" at 165 cm may instead have been a discontinuity due to slumping ~525 ky ago. This was discussed by Kuo and Beck (P65c)).

Sarma, a competent mathematician, even earlier examined the effect of "pseudo-diffusion" on the ionium method and, using a simplified model, interpreted Almodovar's core data quantitatively (P61r). The mathematical development was extended to other models, and some numerical computations were made. It was shown how "pseudo-diffusion" would affect not only ^{230}Th and ^{231}Pa but also ^{14}C and other radionuclides, and ages and sedimentation rates derived from them (P62e, P63j, D64b). This part of his work was presented at a conference in Bombay and published separately (J66b).

In Sarma's work just described, the $^{234}\text{U}/^{238}\text{U}$ ratio was measured in most of the sediment samples. In no case did the ratio deviate from unity by more than 8%, with standard deviations of 5-19% (D64b). (In work elsewhere, and later work by Sarma, statistically significant non-equilibrium ratios were observed.) Measurements of the $^{234}\text{U}/^{238}\text{U}$ ratio in tektites are discussed below (Section H), because the focus there is simply the presence or absence of deviations from equilibrium. If this should be found, then age estimates could be made from it.

Kuo and Beck undertook further improvements in the chemical procedures for Th, Pa, and U, and planned attempts to extend the Ic method further than Sarma's limit (P65c). Considerable progress was made by Kuo (P66c) before she decided to change her field of research.

^{129}I is a fission product, and the chief occurrence of this nuclide in the Earth after the primordial Solar-System endowment became extinct and before the atomic era began was probably as a spontaneous-fission product of ^{238}U . ^{129}I had been observed in uranium minerals. Sarma described a possible ^{129}I dating method for marine sediments. Because of the high solubility of I, freshly deposited sediments should be virtually free of ^{129}I . As a sediment layer became buried and I escape was inhibited, ^{129}I should grow into secular equilibrium with the ^{238}U present. It might be detectable by neutron-activation analysis via ^{130}I (D64b). But quantitative calculations by Kuo indicated that the equilibrium level of ^{129}I in ocean sediments would be too low for detection without enormous effort (P64k). Edwards, who had thought about such matters before, gave reasons for believing that ^{129}I produced by cosmic-ray spallation of xenon in the atmosphere might accumulate in the oceans at levels two or more powers of ten higher than that from spontaneous fission (P66a). He and Ray later calculated that ^{129}I leaving the lithosphere via rock weathering and volcanic processes would amount to about half of that produced in the atmosphere by cosmic rays plus that entering it in extraterrestrial matter. All should enter the oceans and most of it should decay there. However, the levels of ^{129}I in sediments were not discussed other than to suggest that the $^{129}\text{I}/^{127}\text{I}$ ratio in fresh sediments should be the same as that in the marine environment (P67b).

3. Methods Based on Induced Natural Radioactivity

At the beginning of this Project the radiocarbon dating method was rather new and techniques were rather crude. We felt that a number of improvements were possible, and proposed a cooperative program of radiocarbon dating with archeologists at the Carnegie Institute of Pittsburgh Museum. We planned to count the ^{14}C in the form of a gas, possibly CO_2 , in a large proportional counter with several features for background reduction and monitoring (P53e).

The work was undertaken by Diethorn, who constructed a system utilizing methane under several atmospheres pressure (P55d, P56c). The sample was burned to CO_2 , then reduced to C with Mg at 600°C . The C was heated with Al to produce AlC , which was hydrolyzed to generate CH_4 . After several weeks for Rn decay, the CH_4 was counted at 1-5 atm in a counter of 1-liter volume. Background reduction was achieved by massive iron and mercury shielding and anticoincidence counters. At 5 atm modern C gave a counting rate of ~ 31 cpm over a background of ~ 10 cpm. However, the backgrounds were about 2 or 3 times those expected on the basis of ^{14}C gas counters developed in the meantime elsewhere. Moreover, variations were greater than those expected from random-count statistics.

and were anti-correlated with atmospheric pressure. This and an increase following a solar flare indicated a rather large residual cosmic-ray background component (D56a).

Preliminary natural ^{14}C measurements were made on 5 contemporary wood samples and 8 old specimens of geological or archeological interest. Because of the non-statistical background variations, actual uncertainties were greater than those calculated (D56a). (Because of the preliminary nature of these measurements, they were not published.)

Experiments were performed on the effect on gas gain of CH_4 pressure, wire diameter, and applied voltage. The results were correlated by a new theory of proportional-counter behavior (see Section M). It was predicted that the counter should be usable up to 20 atm, at which the required voltage would be ~ 12 kV (for the same gain, 4000, as with 5 atm at 4.9 kV). It was suggested that background variations could be corrected for by monitoring the local cosmic-ray flux and the atmospheric pressure. If operation at 20 atm proved feasible, if the background could be reduced considerably, and if background variations could be compensated for, high-pressure proportional counting of CH_4 might be the most sensitive and practical method of natural-radiocarbon measurements (D56a). Experiments to find a way of minimizing errors due to background variations (P56f) were dropped when Diethorn completed his dissertation work.

It was realized that the method used for CH_4 production was rather complicated and would be difficult to scale up to ≥ 6 l-atm. The reduction of CO_2 to CH_4 by circulating it with H_2 over a catalyst, developed elsewhere in the meantime, would also be difficult to scale up. We wished to develop a 3-liter counter operating at 20 atm, and proposed catalytic reduction in a bomb. Preliminary experiments by Henriksen indicated that this works well (P57h). An electronic system was developed by Mesojedec for use with a counter of the size and pressure mentioned. Several auxiliary coincidence and anticoincidence channels would provide cosmic-ray-background information. A 15-kV power supply was acquired (P58f; 1959-1960 Progress Report, I.C.2.e). (Because of limitations of funding and personnel, and competitive interests including ^{14}C in meteorites, these plans for terrestrial radiocarbon measurements were not carried out.)

The dating aspects of cosmic-ray-induced radioactivity in meteorites have been discussed in Section E, and therefore will not be mentioned further here.

Kuo and Beck planned simultaneous or complementary dating of ocean-core sections by several different methods. One of these was the $^{26}\text{Al}/^{10}\text{Be}$ method, based on the expectation that both of these nuclides would be produced in the atmosphere by cosmic rays, carried into the oceans, and rapidly precipitated; this had been demonstrated elsewhere for ^{10}Be . ^{10}Be would in effect serve to normalize the decaying ^{26}Al . The method should extend further into the past than the I_0 method. Chemical procedures for separation and purification of ^{10}Be and ^{26}Al were partially developed (P65c).

4. Methods Based on Extinct Natural Radioactivity.

The search for new cases of extinct natural radioactivity and the implications for the chronology of the Solar System of the low limit established for the $^{205}\text{Pb}/^{204}\text{Pb}$ level in meteorites are discussed in Section D.

Possible application of extinct natural radioactivity in terrestrial geochronology is also discussed in Section D.

The implications of ^{129}I and ^{244}Pu observations for the chronology of nucleosynthesis are discussed in Section G.

G. Chronology of Nucleosynthesis

Following the announcement early in 1960 of extinct ^{129}I manifestations in meteorites, the writer undertook calculations of the time-dependence of nucleosynthesis, using information on the present relative abundances of ^{238}U , ^{235}U , and ^{232}Th as well as of ^{127}I and the ^{129}I abundance indicated in the early Solar System by the radiogenic ^{129}Xe excess in meteorites. The latter implied an interval of ~ 0.2 Gy from the termination of nucleosynthesis to the retention of Xe in meteorites. Addition of this to the age of meteorites, ~ 4.5 Gy, gave ~ 4.7 Gy as the age of the Solar System, considered to have begun at the termination of the nucleosynthesis of its elements. The heavy-nuclide abundances, with estimates of their relative production rates by the r-process, then indicated by a graphical method that the duration of continuous nucleosynthesis, whose rate was assumed to follow a smooth exponential decrease, was ~ 6 Gy, with a time constant of ~ 0.10 Gy $^{-1}$. These results were similar to those published in the meantime by Fowler and Hoyle.

It had been argued by Urey, Anders, and others that a strong but short-lived source of heat in addition to that resulting from the now-extant radionuclides was necessary to account for the properties of the meteorites. Having a model for nucleosynthesis, it was possible to calculate the relative amounts of all radionuclides in the Solar System at all times after the termination of the production process, and to estimate the radiogenic heat in meteorite- or planet-like matter. All known nuclides of half-life from 10^5 y to 10^{11} y were considered and most of them included in the calculations. Two sets of calculations were made, one for continuous nucleosynthesis ending 4.7 Gy ago, and one for a single nucleosynthetic event occurring long enough ago (~ 6.5 Gy) to yield the present heavy-nuclide abundances. In the continuous case now-extinct nuclides exceeded non-extant nuclides in heat generation for ~ 6 My, and in the single-event case for ~ 65 My. But in neither case was any of their heat available at the time of formation of meteorites.

If it was assumed, however, that the bulk of the stable and long-lived nuclides were produced by prolonged continuous nucleosyntheses, and that just prior to or during the condensation of the protoplanets a single local supernova erupted and contributed several percent of the mass of the Solar System with single-event proportions of the now-extinct nuclides, the supplementary heat might be available. This was referred to as "mixed" nucleosynthesis, and curves were presented for the total specific heat production in chondrite-like matter for last-event contributions of 1, 10^{-1} , 10^{-2} , 10^{-3} , 10^{-4} , 10^{-5} , and 0. The "mixed" mechanism, with $\sim 10^{-2}$ - 10^{-1} terminal contribution, makes it easier to account for ^{129}I effects.

Some possibilities for a more realistic model of nucleosynthesis, in which the removal of heavy atoms from the interstellar medium by processes other than radioactive decay is incorporated, were also discussed. For a constant loss probability the differential equation reduces to the same form as that already used, with a change in the meaning of one parameter. The differential equation for the case where the loss probability varies like the production rates, both being proportional to star-formation rates, was set up but not solved.

This work was presented at a Session of the American Chemical Society honoring Charles D. Coryell in 1960 April (M61a), summarized at a topical conference in 1960 June (J61b) and in our Progress Report of that year (P60a),

and published in full with the other papers of the ACS session (J61a).

The subsequent establishment of the second case of extinct natural radioactivity, ^{244}Pu , made possible further deductions about nucleosynthesis chronology. Since ^{244}Pu , ^{238}U , ^{235}U , ^{232}Th , ^{129}I and ^{127}I are all r-products, they form a homogeneous group which actually delineates the time-behavior of the r-process only, and other considerations are necessary to relate that to general nucleosynthesis chronology. The additional short-lived member of this group in particular permits more critical evaluation of the concept of a terminal "spike" to nucleosynthesis (the "mixed" model).

Huey with the writer considered a number of models for the time-dependence of nucleosynthesis which had been proposed in the past, and classified them according to the number of astronomical parameters. The two most complete models were the ones considered by the writer in 1960-1961 (see above) (model 5a) and one proposed by Hohenberg in 1969 (model 5b), the number indicating the number of parameters. The parameters of model 5a were the duration of the exponentially-decreasing continuous phase (Δ), the time-constant for the decrease (λ_d), the fraction of r-products added in the terminal spike (ϕ), the free-decay interval before xenon retention in meteorites (Ξ), and the age of meteorites (M). Model 5b envisioned constant continuous nucleosynthesis (no λ_d) but instead a large initial spike corresponding to rapid nucleosynthesis in massive stars during the collapse of the galaxy to a disk (fraction α). The evaluation of 5 parameters requires 5 input quantities. One, M , can be evaluated separately from independent radiometric dating of meteorites by several methods. The other four inputs were "resultants" of abundance (N) ratios and production-rate (ρ) ratios for selected nuclide pairs:

$$\begin{aligned}x_U &= (N_{235}/N_{238})_o / (\rho_{235}/\rho_{238}) \\x_{Th} &= (N_{232}/N_{238})_o / (\rho_{232}/\rho_{238}) \\x_I &= (N_{129}/N_{127})_m / (\rho_{129}/\rho_{127}) \\x_{Pu} &= (N_{244})_m / (N_{238})_o / (\rho_{244}/\rho_{238})\end{aligned}$$

Here o indicates the present time, and m indicates the time of meteorite-formation as xenon-retaining solids.

For each x an equation was derived in terms of all six astronomical parameters and the pertinent nuclear parameters (decay constants) (inadvertently omitted from P70f but included in P71e, where the general model was designated model 6). For each model 5a and 5b (derived from model 6 by omitting one parameter) the four simultaneous equations were solved for the four remaining parameters by the Newton-Raphson method using a computer. Solutions were obtained not only for the best estimates ("nominal" values) of the resultants and M , but also for sets in which each was varied systematically through its physically interesting or possible range. The "nominal" values of the parameters seemed reasonable in both models (although we felt that model 5b was intrinsically less realistic). However, in both models each output value was quite sensitive to at least one input value, so the results were not well determined. In each model $\phi \sim 3\%$, but zero was not excluded.

After the first set of calculations (P70f) was concluded we received a pre-print from Fowler describing almost identical calculations for our model 5a,

with somewhat different input and output values ($\phi \sim 9\%$).

More extensive calculations followed, with improved computer programs and revised input values. Among the latter revisions was a reduction in the P_{235}/P_{238} ratio to allow for some self-destruction of ^{235}U and precursors by chain reactions in expanding supernova shells, proposed elsewhere in the meantime. We also made calculations for a model 5c, in which there is no terminal spike, and for the complete model 6 with the initial-spike fraction, α , arbitrarily set at 25%. In model 5c a large negative λ resulted, the unreasonableness of which seemed to virtually prove the existence of the terminal spike (ϕ now $\sim 5\%$ in models 5a, 5b, and 6) (P71e).

During the next year the input values were further reviewed and revised, and the calculations were repeated. The chief change in output was that the terminal-spike contribution ϕ was $\sim 3\%$ instead of $\sim 5\%$. The "nominal" solutions for models 5a, 5b, 5c, and 6a (special case) were illustrated graphically (P72g). The model-5a parameters were given in a paper on the search for ^{205}Tl anomalies from extinct natural radioactivity of ^{205}Pb . The values of Δ , λ_p , ϕ , and M were used to calculate the $^{205}\text{Pb}/^{204}\text{Pb}$ ratio at the termination of Solar-System nucleosynthesis, assuming that they would be applicable to s-process as well as r-process nucleosynthesis. Comparison with the experimental limit then allowed deductions about Ξ for the ^{205}Pb - ^{205}Tl case. For the same ϕ , $\Xi \geq 120$ My. This should be shorter than the value for xenon retention, ~ 160 My, because Tl-Pb fractionations should predate gas retention. If it were assumed that $\phi = 0$ for the s-process, the limit to Ξ would then be only ≥ 60 My (J72a).

In the 1971-1972 Report we also extended the r-process general model to include the time-dependence of hydrogen in the interstellar medium and in stars in the Galaxy. We followed to some extent the treatment of Fowler, which had been published in 1972. The resulting model has 8 astronomical parameters, and accordingly was designated model 8a. Five (α , Δ , ϕ , Ξ , M) are the same as in model 6a. The sixth model-6a parameter (λ_p) became a function of the other three model-8a parameters, which are related to star formation and intergalactic-hydrogen infall. The model yielded an equation for the time-dependence of the Fe/H ratio in the interstellar medium relative to the value in the Sun, which was presumably the interstellar value at the origin of the Solar System as defined above. Main-sequence stars, including the Sun, and even some giant stars are believed to have so little radial mixing that their atmospheres retain their initial composition. Like the Sun, each star samples and records the composition of the interstellar medium at the time of its formation. Isolated stars cannot usually be dated, but star clusters can. Following Arp, Sandage, and others, data on 11 open (galactic) clusters and 7 globular clusters were compiled and plotted in the plane of heavy-element content relative to Sun versus age. The points showed the well-known trend of rapid heavy-element enrichment. Because of scatter, from observational and theoretical uncertainties as well as undoubtedly real spatial inhomogeneities in the interstellar medium, it was not possible to delineate the mean trend very precisely. Thus the additional parameters could not be evaluated uniquely. Instead, the functional relationship was calculated for many combinations of parameter values for insight as to their individual influence. A number of parameter-value sets gave curves fitting the points quite well. Rough ranges of possible values were indicated. The most important conclusions were: (1) an initial "spike" is not

necessary; (2) infall of intergalactic hydrogen is not necessary; (3) it is necessary to include the removal of already-synthesized heavy atoms in later-generation star formation at the same specific rate as hydrogen removal; (4) the star-formation rate depends on the mass of the interstellar medium to a rather high power (~ 3) (P72g).

This work was presented at a Symposium on Cosmochemistry in 1972 August and an abstract has been published (J73b).

In our final Progress Report (P76f), subsequent developments were noted, and estimates of M and the four resultants up-dated. Pending a complete new set of computer computations, the variational calculations of the preceding set (P72g) were used to up-date the nominal values of the various astronomical parameters of the several models. Recent information on r -process production rates will be taken into consideration in the final computations.

It was also noted (P76f) that there have recently become available rather good estimates of the ages of individual stars or groups of non-cluster stars, with matching heavy-element-abundance data. Combination of these data with those on clusters should delineate the time-dependence of the heavy-element content of the interstellar medium considerably better than was possible previously.

It is hoped that the entire collection and assessment of data, repetition and extension of computations, and preparation for publication can be done during the coming summer.

H. Tektite Studies

As long as tektites have been known they have been associated with meteorites. For some time after their recognition they were regarded as glassy shower meteorites. At the time of our initial research interest in meteorites, this hypothesis had begun to be doubted, the chief alternative being that they were terrestrial silicious matter which had been melted and scattered by impact with a large cosmic body, asteroidal or cometary - i.e., impactites. Rather close proximity of some large impact craters to some tektite strewn-fields strengthened this hypothesis. A third hypothesis - actually a variant of the first - was in the formative stages: tektites are lunar impactites, gravitationally focussed to rather small strewn-fields during their passage from the Moon to the Earth.

As already mentioned in Section E, after we had demonstrated the presence of cosmic-ray-induced radioactivity in meteorites, we sought to test the extra-terrestrial origin of tektites by seeking the same cosmogenic nuclides in them. Unfortunately, as explained there, the initial measurements of Ehmann and the writer gave evidence for both of these radionuclides in several tektites, and this was announced as evidence of a meteoritic origin (D57b, M58a, J58c, J58f). As what seemed to be the only possible explanation of all of the known or supposed facts, an origin from out of the Solar System was suggested. A lunar origin seemed impossible because of the necessary depths of most of the melted matter. Given a translunar origin, the sizes of the strewn-fields are too large for fragmentation of a single body. But a cluster of objects, no matter how compact, would be tidally dispensed in a solar orbit. An origin outside of the Solar System would allow the cluster to remain intact until it was dispersed shortly before arrival on Earth, possibly by her own gravitational field (J58a).

While on a sabbatical leave at the Max-Planck-Institut für Chemie, Mainz, Germany, 1957-58, the writer isolated potassium from a number of tektites, and the isotopic composition was determined mass-spectrometrically by Professor R. Hintenberger, to test the possible extra-Solar-System origin (J58a). All K samples had normal Solar-System isotopic composition (P59g).

As detailed in Section E, measurements for ^{14}C in tektites by Goel and more careful searches for ^{10}Be and ^{46}Al in tektites by Chakrabartty and Rayudu failed to confirm that tektites had been exposed to cosmic rays. Other investigators arrived at the same conclusion through searches for cosmogenic noble gases as well as for radionuclides. Nevertheless, our erroneous claim undoubtedly had a stimulating effect on tektite thinking and research.

In 1959 June, Goel and the writer participated in a field trip in Texas conducted by V. E. Barnes and including among the others alumnus Ehmann. The purpose was to study the occurrence of bediasites and to collect specimens for research and exchange. 57 tektites (totalling 328 g) were found, including 3 (5.5 g) by Goel and 8 (92.1 g) by the writer. Some new areas of occurrence were identified, and non-occurrence was established in many areas. Some interesting modes of occurrence were observed (P59g, P60t). (Some specimens have been used for the stated purposes.)

Experiments undertaken by Sextro (P65a) but never completed, to apply the fission-track dating technique to tektites, are described in Section F. Neutron activation analysis of sodium in some tektites by Tanner (P68a) is mentioned in Section I.

The chief remaining alternative to a terrestrial-impact origin of tektites is that of lunar impact, which has continued to be advocated by a minority. (A 1976 book by O'Keefe is said to affirm that a terrestrial origin is impossible, and that tektites are samples of the Moon.) Of the possible terrestrial origins there are two major variants. The majority view is that the tektite matter was deep-seated and melted in the course of excavation of a large crater by the impact of a rigid dense body of meteoritic composition and asteroidal origin. The other view, suggested by Urey in 1957, is that the impacting body was a comet, whose disruption began in the atmosphere and was complete after only shallow penetration of the Earth's surface, so that tektites were formed from near-surface material. Independently, Schwarcz in 1962 noted similarities in chemical composition between most tektites and some soils, and suggested that tektites were derived from soils.

Tsomi noted the well-established fact that the $^{234}\text{U}/^{238}\text{U}$ ratio is altered in weathering of rocks and subsequent processes. As a result of the α -recoil displacement in its formation from ^{238}U , ^{234}U atoms are more readily leached from the solids by water. Residual materials, especially soils, have variable $^{234}\text{U}/^{238}\text{U}$ activity ratios, usually less than unity and down to ~0.6 (but occasionally greater than unity as a result of redeposition from ground water). If tektites contain an appreciable fraction of soil-derived matter, their initial $^{234}\text{U}/^{238}\text{U}$ ratio should be less than unity. The deficit should decay exponentially with the ^{234}U half-life, 247 ky, and should disappear as secular equilibrium is restored in somewhat over 1 My. In the Australasian tektites, dated by various methods at ~700 ky, ~14% of any initial deviation from equilibrium would still be present. Initial ratios of 0.65 and 0.85 would now

be 0.95 and 0.98, respectively. She undertook to determine as accurately as possible by α -particle spectroscopy the $^{234}\text{U}/^{238}\text{U}$ activity ratio in uranium chemically isolated from representative tektites from various localities, as a means of testing the soils hypothesis (P68c). The program has involved the efforts of several persons in the chemistry, instrumentation, and data analysis through 1973, and is not yet complete.

The chemical procedure developed by Sarma (D64b) for uranium from ocean sediments was adopted and modified by Tsomi (P68c), Thomas (P69b) and Kan (P70i, P71b). In Kan's final work she prepared 4 uranium samples from "young" tektites (philippinites, thailandite, australite) and 2 from "old" ones (bediasite, moldavite) (P72b).

Counting was done with surface-barrier detectors, initially with an active area of 200 mm^2 . Amplified pulses were recorded with a 256-channel analyzer (P68c). These were replaced with new detectors of the same size for improved resolution (P69b, P69i), and later by 450 mm^2 detectors for higher counting yields; only the one with the better resolution was ultimately used (P70h, P70i). Counting yields of ~25% were obtained by putting the planchet as close as possible to the detector, at the expense of poorer resolution (P71b). Final counting of all of Kan's samples was done by the writer under uniform conditions, in many periods of one to several days, and only visually acceptable spectra were retained for use (P72b, P73a).

Because of bias and gain drifts, the several spectra for a given sample can not be simply added. Thomas shifted separate spectra by integral numbers of channels and added (P69b), but this is unsatisfactory for varying gain. Trivedi developed a method of combining spectra of variable bias and gain, and computerized it (P71i). This was improved and incorporated into a more general program for data manipulation by Nachimson. That program also provided for subtraction of cumulative raw spectra when necessary, and for plotting each differential spectrum for visual examination and acceptance or rejection. While combining the accepted spectra in the manner indicated, it also creates a "completeness spectrum" indicating the part of the total spectrum which is complete and useable (P72i, P73a). All 6 of Kan's samples have now been processed by this program by Nachimson, yielding at least two combined spectra for each sample, with total acceptable counting times of 40 to 88 days for each sample (P73a).

Because of the complexity of the α -spectra of ^{238}U , ^{235}U , and ^{234}U (and ^{232}U and/or ^{236}U if present), analysis in terms of the activity of each nuclide in the experimental spectrum is not simple. Thomas constructed a semi-empirical function to represent the instrumental spectrum for a single alpha energy, and wrote a computer program to synthesize a complex spectrum. He chose values of the parameters which gave the best eyeball fit in successive trials, and numerically integrated the contributions from each isotope (P69b). Trivedi extended the analytical function and developed a computerized least-squares program for determining the parameters (P70k, P71i). Nachimson incorporated these ideas into an improved program, which could accommodate an arbitrary fitting function of an arbitrary number of parameters. It also produced a coarse line-printer plot of the experimental spectrum and the best-fitting analytical synthesis. This enables a visual evaluation of the fit and guides further modification of the function (P73a). The program also computes the reduced chi-squared, which will be the final criterion of goodness of fit.

Several functions have been tried (P69b, P70k, P71i, P73a), the latest and most successful of which had 8 parameters. Various weighting functions have also been tried. But a satisfactory fit has not yet been achieved.

What remains to be done is (1) experimentation with fitting functions and weighting factors until a suitable combination is found; (2) finding the most suitable way of extracting the true $^{234}\text{U}/^{238}\text{U}$ activity ratio from a spectrum and its fitting parameters (discussed in P73a); (3) analysis of the 13 combined spectra from the 6 tektites; (4) comparison of the ratios for the 4 "young" tektites with those for the 2 "old" tektites; and (5) consideration of the bearing of the results on the soils hypothesis of tektite origin. The writer intends to complete this project as time becomes available.

1. Neutron-Activation Analysis

In 1956 and again in 1962 it had been demonstrated that the fission product ^{129}I can be detected with enormous sensitivity by neutron-activation analysis via ^{130}I , and in the latter year Edwards had described many possible uses as a tracer in nature and the laboratory. He pointed out that it should be produced by cosmic rays in the atmosphere and in meteorites, and that this should lead to several dating possibilities. Nuclear explosives testing should add ^{129}I to the surface layers of the Earth in much greater quantities, but still requiring the sensitive activation-analysis technique for its detection and measurement.

Kuo reviewed the nuclear basis and some instrumental aspects of neutron-activation measurement of ^{129}I , in connection with her calculations of the expectation levels of ^{129}I in ocean sediments and stone meteorites (P64k).

Edwards discussed the sensitivity of the method in terms of ^{129}I and $^{129}\text{I}/^{127}\text{I}$ ratio; if the latter is too low, triple neutron capture in ^{127}I interferes. He quoted some results of measurements with which he had been associated previously. He described plans for measurements of biological and mineral specimens, iodine extractions already completed, and plans for exploratory γ - γ -coincidence counting of activated samples (P66a). Tests of pre-irradiation separation of iodine from chondritic meteorites were being made (P66d).

Pre-irradiation iodine separation procedures, preparation of samples for reactor irradiation, and post-irradiation chemistry were described by Edwards and Ray. β - γ -coincidence counting was deemed necessary for maximum ^{129}I sensitivity in small (< 100 mg) samples. But for gram-size samples self-absorption of β -particles would seriously limit sensitivity, suggesting γ - γ -coincidence counting as more favorable. Using disintegration schemes for ^{126}I , ^{128}I and ^{130}I , γ -scintillator single-channel-analyzer energy settings were chosen to favor detecting ^{130}I coincidences and discriminate against the two interfering isotopes, produced from ^{127}I . Several multigram I_2 samples were irradiated at the Argonne CP-3 reactor, and measurements made with the γ - γ -coincidence counter having 3-inch NaI(Tl) scintillators. After the decay of 25-min ^{128}I , ^{126}I could be observed in all samples and ^{130}I in most; calculated $^{129}\text{I}/^{127}\text{I}$ ratios ranges from $< 2 \times 10^{-2}$ to $\sim 10^{-11}$. A long I_2 irradiation was also made to measure the triple-neutron-capture interference (P67b; see P68j for corrections).

A β - γ -coincidence system was assembled using a 4 π flow β -counter and two 4-inch NaI(Tl) γ -scintillators in a thick iron shield. This and its energy settings were discussed by Edwards and Rey (P67b). Its construction and performance were described in detail by Toia (P67j). (As explained in Subsection II. C.3, this system was disassembled and removed before it was actually used.)

The writer's estimations of ^{129}I production rates from cosmic rays in meteorites were mentioned in Section E. In all cases, with saturation irradiation (≥ 50 My exposure), $^{129}\text{I}/^{127}\text{I}$ ratios were favorable. The ^{129}I mass fractions, on the other hand, were so low that perhaps ~100-gram samples would have to be sacrificed, and excellent instrumentation and techniques would have to be employed. Any attempts to detect meteoritic ^{129}I should be deferred until sufficient experience has been obtained with terrestrial materials (P67h).

Tandon initiated a program to determine Hg, Tl, Pb, and Bi in various phases of iron meteorites by activation analysis. The motivation was to study element fractionations in meteorite parent bodies. Chemical procedures were developed using ^{59}Fe , ^{203}Hg , ^{204}Tl , ^{210}Pb , and ^{210}Bi tracers. It was planned to measure ^{204}Tl and ^{210}Bi in a low-level β counter and ^{203}Hg and ^{203}Pb (produced by fast neutrons) by γ spectrometry. Several metal and troilite specimens and monitors were irradiated in the Argonne CP-5 reactor (P65d).

That irradiation and a similar one at the Wright-Patterson Air Force Base reactor indicated that the "effective thermal neutron cross section" of ^{204}Pb for the fast-neutron production of ^{203}Pb in the available favorable positions was in each case too small for use at meteoritic lead levels. Calculations for cyclotron fast neutrons indicated similar specific activity expectations. An experiment at the University of Pittsburgh 15-MeV cyclotron was planned (P66g). (There is no record that this irradiation was performed.) Huey considered both fast- and thermal-neutron irradiation of Pb and Tl, partly because preliminary determinations of ^{204}Pb and Tl in meteorites would allow selection of the more favorable specimens for mass-spectrometric search for ^{205}Pb - ^{205}Tl effects (Section D). For ^{204}Pb , fast-neutron activation inadequacy was confirmed, and for Tl it is much less sensitive than thermal-neutron activation. Thermal-neutron activation of ^{208}Pb would yield stronger activities, but the 3.3-hour half-life of ^{209}Pb would require working at a reactor site (P68g). For these reasons, plus the failure of activation methods to provide the isotopic composition of the Pb, activation analysis of that element was dropped from consideration.

Tandon refined chemical procedures for Hg, Tl, and Bi, and carried out irradiations with the CP-5 reactor. Preliminary Hg results and upper limits for Tl and Bi were obtained on metal samples from 4 iron meteorites (P66f). The Hg chemistry was improved, and it was found that because the long ^{204}Tl half-life precludes decay-curve identification, recycling to constant specific activity in the carrier was necessary. Final values for all three elements in several metal samples from the 4 irons and 2 troilite samples from Canyon Diablo were tabulated (P67d). All 3 elements were greatly enriched in the troilite as compared to the metal.

The troilite used by Tandon was from the same powder prepared and used by El-Badry et al. (S60a,b,c). Tandon's Tl contents found, 65 and 74 ppb, were considerably higher than that found by El-Badry et al., 1.5 ± 0.3 ppb. Tandon's Tl contents for Canyon Diablo metal, 5.8 and 7.8 ppb, were also higher than that

of El-Badry et al., 0.25 ± 0.05 ppb. This would greatly reduce the two corresponding $^{204}\text{Pb}/^{203}\text{Tl}$ ratios calculated and plotted by Ostic et al. (P68d, J69a).

Tanner continued the activation analysis of Hg, Tl, and Bi in iron meteorites. Chemical procedures were modified. It was found very difficult to discriminate between ^{204}Tl and contamination β activity (P67e), and this element was dropped from subsequent work (P68e). For ^{210}Bi measurement, β counting of its ^{210}Po daughter was substituted and found more specific and sensitive. γ -spectrometry of ^{203}Hg was quite specific for Hg analysis.

Tanner determined Hg and Bi in 30 metal samples from 19 iron meteorites of several structural classes, 3 troilites from different irons, and schreibersite from Canyon Diablo. No statistically significant variations between structural classes were apparent. Likewise, there were no evident correlations between Hg and Bi, or between either and Ni, Ga, or Ge. Contrary to Tandon's finding, Hg was not notably enriched in troilite. Bi was distinctly enriched, but much less than indicated by Tandon. Both elements seemed to be somewhat depleted in schreibersite relative to metal.

Tanner also undertook the determination of cadmium in tektites via neutron activation to 43-day $^{115\text{m}}\text{Cd}$, to determine whether it, like the other volatile elements Zn and Cu, was depleted relative to the presumed parent rocks. A chemical procedure was developed, and 4 samples of indochinites were analyzed for Cd using γ counting in a 4-inch well NaI(Tl) scintillator. A borrowed 30-cm³ Ge(Li) detector was found to give improved but still unsatisfactory $^{115\text{m}}\text{Cd}$ isolation. ^{124}Sb gammas were also observed, and it was planned to combine Sb analyses with future Cd analyses (P68a).

γ - γ -coincidence counting was suggested by Tanner (P68a) as an improvement for assay of $^{115\text{m}}\text{Cd}$ and other cascade- γ emitters. A fast γ - γ -coincidence system was designed (P68i), built (P68i, P69j), and partially tested (P69j). Tanner was also influential in our acquisition of a 30-cm³ Ge(Li) detector and spectrometer system including thick lead-brick shield and 512-channel analyzer (P69i).

Tanner cooperated with others in the University in activation-analysis work, using our facilities. With P. C. Smith he determined Au, In, and Ag in doped semiconductor silicon by instrumental (non-destructive) γ -ray spectrometry using the Ge(Li) detector (P69g). Parallel Hall-effect measurements were made to determine the effects of concentrations of these trace elements on conductivity.

With C. L. Smith, Tanner determined Sb as well as Cr and Fe in low-alloy steels, in which segregation of Sb and other impurities into grain boundaries was suspected of contributing to temper embrittlement. Fractured surfaces were acid-leached, and the 3 elements in the dried residue were γ -instrumentally activation-analyzed (P69h).

J. Mössbauer Spectroscopy of Natural Materials

Not long after its discovery, the Mössbauer effect assumed greater importance as a probe of the physical and chemical structure of solids than as an indicator of nuclear structure and properties. ^{57}Fe immediately dominated the field because of the simplicity, ease, and convenience of measurement of the effect with ^{57}Co sources, and of the importance of iron in physical, chemical, mineralogical, biological, and industrial systems.

Catz proposed to use the ^{57}Fe Mössbauer effect to study the chemical form of iron in tektites. All occurrences of these objects contain large amounts of both ferrous and ferric iron. In 1964 it was suggested from the magnetic properties that submicroscopic nickel-iron metallic spherules are present in some or most tektites. It was thought that Mössbauer spectroscopy might identify this iron if present. Preliminary experiments using borrowed and leased equipment were done on thallandites in the form of pressed powders and ground wafers. The abundant ($\sim 4\%$) Fe^{II} doublet was clearly identified, and evidence of a weaker doublet of Fe^{III} ($\sim 0.2\%$) was seen. Sensitivity was found to be inadequate to see the 6-peaked spectrum of Fe^0 (predicted $\sim 0.004\%$) (P64n).

With the thick low-Fe absorbers, degraded 123-keV gammas gave a high background, weakening the 14-keV-gamma absorption. Use of coincidence counting, so that only those 14-keV gammas were counted which were accompanied by a 123-keV gamma seen in a separate detector, was considered. The long coincidence resolving times required by the appreciable 14-keV-state lifetime ruled that out (P64n).

In 1965 Toia, Catz, and Edwards reviewed the rather large amount of ^{57}Fe Mössbauer work which had by then been done on minerals and meteorites. A broad program of studies of natural iron-containing materials was planned, with possible extension to minerals of tin and some other elements amenable to the technique (P65o).

The previously leased constant-velocity device had been purchased, and an automatic control and data-recording system was being built. The partially completed system and its functioning were described the following year, with further improvements being made, by Toia (P66l). A logical diagram of the final completed system was presented in the 1967-1968 Report (P68i).

In 1966 Lefelhocz described experimental techniques and data analysis, and up-dated the summary of previous work on iron in natural materials. At the suggestion of R. A. Friedel of the U. S. Bureau of Mines, Mössbauer investigations of the properties of iron in coal were planned (P66b).

The work on coal was quite successful. It had been inferred that some coals contain "organically bound" iron (OBI), and information on it was especially sought. Nine vitrain or whole-coal samples of rank from lignite (72% C) to anthracite (93% C) were investigated at room temperature and at liquid- N_2 temperature. Five, known from analyses to contain sulfide iron, yielded the narrow symmetric two-line spectrum of pyrite or marcasite. Five whose analytical data indicated OBI exhibited a different wide symmetric doublet similar to many Fe^{II} compounds. Some samples showed both components, and some showed no discernable resonance absorption. The non-pyrite-iron (NPI) spectrum was different from any known iron Mössbauer spectrum. Plots of quadrupole splitting versus isomer shift were made, with all known two-line spectra for synthetic iron compounds and natural iron-bearing solids represented by one or two points. Although there were several other points in the neighborhood of the NPI point, each of the others was coupled with another point (two forms of iron), whereas the NPI point occurs alone in coal. There was poor correlation with the analytically determined OBI. The unique iron was deduced to be Fe^{II} in a high-spin configuration, in six-fold coordination with octahedral symmetry, and apparently in a rather well-defined state. Comparisons with known compounds suggested binding to heterocyclic nitrogen aromatic groups or in a clay-like silicate mineral or gel (P67a, J67b).

Páez took up the work on coal in 1967-68, making measurements of additional coals and of samples after thermal and chemical treatments. With L. F. Vassamillet at Mellon Institute, electron-microprobe studies of NIP-bearing coal were made. Iron was found to be correlated spatially with Si, Al, and Mg but not with K, Na, or Ca. Two ferromagnesian-aluminosilicate minerals were suggested, staurolite and chlorite, with the former giving poorer stoichiometric agreement and having the wrong Mössbauer spectrum. Hence Mössbauer spectra were run on separate samples of chlorite-group minerals: chlorite itself, prochlorite (which was among the minerals previously reported in coal), and pennine. All three minerals showed two strong peaks at the same velocities as the NPI, but the absorption in the peak near zero velocity was greater than in that at high velocity. That could be because of (1) orientation effects (chlorites are phyllosilicates), (2) distribution of Fe among the two available Mg+Fe sites, and/or (3) presence of some Fe^{III}. The spectra all showed a weak Fe^{III} peak at intermediate velocity, which should be accompanied by another near zero velocity. Although OBI was not eliminated, it seemed most likely that NPI was Fe^{II} free of Fe^{III} in one site only of a chlorite-like mineral (P68b).

Páez also examined three high-iron lignites. One showed no resonant absorption, one showed ill-defined resonant absorption, and one showed a narrow low-velocity doublet similar to Fe^{III} in octahedral 6-coordination or square-planar 4-coordination. Altogether, the results suggested reduction of Fe^{III} to Fe^{II} in the metamorphism of lignite to coal (P68b).

In 1968 the constant-velocity system was retired because of inherent limitations and frequent servicing requirements (P69i). Using apparatus of the Physics Department, Páez began the study of relaxation effects in iron compounds over a range of magnetic fields to 50 kG and at temperatures down to 1.5°K, under other supervision. He noted that chlorite minerals are paramagnetic, and that relaxation effects on the Mössbauer spectra of Fe^{II} ions in these minerals and in the coal matrix might yield interesting information about both. The major part of his 1968-1969 report concerned theoretical calculations of Mössbauer spectra and relaxation effects for spin-1 Fe^{II} in paramagnetic materials. It was planned to use the results to interpret further studies of iron in coal and minerals at ambient and low temperatures with and without magnetic fields (P69a). (However, Páez changed his dissertation topic and did no more work on natural materials.)

In 1968 a constant-acceleration Mössbauer spectrometer (International Chemical and Nuclear Corporation NS-1) with electronics including a 512-channel multiscaler (Nuclear Data 1100) had been acquired. However, numerous electronic difficulties arose, most importantly unequal dwell-times in the multiscaler channels. (The dwell-times, indicated by a baseline spectrum without resonant absorber, differed in groups of $512 \div 2, 4, 8, \dots$ channels, giving a stepped baseline.) Although returned twice to the manufacturer (ND), neither it nor the supplier (ICN) would assume responsibility for supplying a properly functioning unit (P69i). Much time was lost by Páez, Toia, Ulmer (see below), Harnic (P70h), and the writer. Eventually Harnic found that the malfunctions were in the Nuclear Data dwell-time and/or reset circuit boards, and he corrected them by resoldering the connections and cleaning the boards (P72k).

In the meantime Ulmer, who took up the work in 1969-1970 (P70d), devised a procedure and computer program for taking a blank spectrum, determining baseline normalization factors, and correcting an absorption spectrum accordingly. The

procedure also eliminated the curvature of the baseline resulting from geometry effects. While not completely satisfactory, the procedure allowed him to use the new system to obtain improved spectra of some coal and lignite specimens. He was able to detect a minor component (pyrite ?) in a NPI concentrate prepared by Pérez using ultrasonic disintegration and pyridine extraction (P68b).

The most extensive measurements (12 coal samples, 15 minerals) were made by Kamarchik (P72a), using the repaired system (P72k). He applied a computerized least-squares analysis to some spectra. He found that, when an NIP-bearing coal was ashed, the residue still showed the NIP Mössbauer spectrum though somewhat weakened and broadened. A sample of chlorite similarly heated behaved the same way. This proved that NPI is not OBI but is almost certainly in a silicate mineral. The more extensive coal and mineral measurements of Kamarchik and published mineral measurements seemed to confirm that NPI is in a phyllosilicate, probably a member of the chlorite group. Since chlorite consists structurally of alternating brucite and talc-like layers, brucite and talc specimens were run. The talc resembled NPI more closely than did brucite, but neither matched as closely as some (but not all) chlorite specimens, and specimens designated as clinocllore, chloritoid, and kämmererite. Kamarchik considered the kämmererite to be the closest match, but this mineral had unequal absorption in its two peaks. No perfect match to NPI was found, and further experiments were suggested (P72a).

(Brucite, talc, and chlorite are nominally iron-free, though usually some Fe^{II} replaces Mg, as the color and name of chlorite implies. Clinocllore, prochlorite, and pennine (or penninite; kämmererite is a variety) are among the nominally iron-bearing minerals of the chlorite group. Chloritoid is not a chlorite but a "brittle mica". However museum identifications are not always trustworthy. Fe^{III} is commonly also present in phyllosilicates.)

Some additional mineral samples were prepared by P. L. Kohman, a volunteer worker during the summer of 1972, and spectra of them were taken by her and by the writer during the 1972-1973 year. However, definitive results on the identity of the NPI-bearing phase(s) have not yet been obtained. It may be a mineral distinct from any hitherto identified, and possibly more than one mineral is involved.

K. Use of our Facilities by Others

In Section I, Neutron-Activation Analysis, we have mentioned the collaborative work by Tanner and graduate students in other departments. Those were P. C. Smith of the Department of Electrical Engineering (P69g) and C. L. Smith of the Department of Metallurgy and Materials Science (P69h). In each case samples of the student's were included in reactor-irradiation packages along with Tanner's samples, and gamma spectroscopy was carried out using our Ge(Li) system.

For several years B. Keisch and R. C. Callahan of the Division of Sponsored Research of the University (now the Carnegie-Mellon Institute of Research) have been conducting studies of the isotopic composition of lead associated with works of art, using our mass spectrometer (P72h, P73e, P76i). Samples analyzed included manufactured white lead and other lead-bearing pigments, paint from paintings of known and questionable origin, and, recently, bronze alloys. Over 400 samples of lead have been analyzed. Pattern-grouping methods were developed, and some interesting relationships were found. In the nineteenth century there was a considerable diversification of sources of lead of commerce,

resulting in a greatly increased range of isotopic composition of lead in art works in the 19th and 20th centuries. In several cases the results bore significantly on the question of authenticity of paintings attributed to certain masters, or of additions to old paintings. The work has been supported by the National Gallery of Art. A comprehensive paper is in the press.

Another "fingerprinting" application of lead isotopes has been made by M. D. Rowe of the Environmental Health Program, Graduate School of Public Health, University of Pittsburgh (P76h). The objective was to discriminate between various sources of lead in young children, with particular interest in atmospheric lead, lead in food, and lead-based paints. Samples of lead chemically isolated from hair of young children, food, atmospheric particulates, and adult blood and hair were analyzed in our mass spectrometer. Hair was found to be heavily contaminated with atmospheric lead even after extensive washing, and hence is not an indicator of body lead. Although blood lead of adults is not a good indicator of their current lead exposure, preliminary results indicate that it should be in young children.

L. Chemical and Radiochemical Procedures

Chemical separations, isolations, and purifications were essential parts of many of the investigations carried out in this Project. These included the purification of ordinary elements in the search for or measurement of primary natural radioactivity (Section C); the separation and radiochemical purification of elements from bombarded targets in the study of the cross sections of nuclear reactions (Section A), in the search for and study of artificial radionuclides (Section B), in the measurement of thick-target bombardment yields and distributions (Sections A,E), and in activation analysis (Section I); the isolation and radiochemical purification of elements from meteorites and tektites for the study of cosmic ray-induced radioactivity (Sections E,H); the isolation and radiochemical purification of Th, Pa, and U from ocean sediments for ^{230}Th and ^{231}Pa dating (Section F); the isolation of U from tektites for measurement of the $^{234}\text{U}/^{238}\text{U}$ α -activity ratio (Section H); the separation and purification of Pb, U, and Th from meteoritic and terrestrial materials for mass spectrometry in applications of the U,Th-Pb dating methods and of Nd and Sm in applications of the ^{147}Sm - ^{143}Nd dating method; and similar separations of Tl, U, and Nd in the search for extinct natural radioactivity of ^{205}Pb , ^{247}Cm , and ^{146}Sm (Section D).

Many of the separations used followed standard analytical methods, were adapted from compilations of radiochemical procedures, or were taken from the literature describing similar activities. In many cases, however, modifications were necessary or new procedures were developed. Details may be found in the reports and publications cited in the sections mentioned.

Some specific chemical procedures may be mentioned. Rightmire et al. developed chemical procedures for production of carrier-free ^{26}Al and ^{22}Na from high-purity Mg targets bombarded with cyclotron deuterons (P56g,J57b). Merz developed a procedure for practical production of carrier-free ^{28}Mg from NaCl or KCl targets bombarded by high-energy protons (P60e). Treytl developed a method of preparing Te targets by electrodeposition on Ni foils (P61t) and a radiochemical procedure for separating and purifying I from these targets after bombardments (P61u). Cressy designed and built a Teflon still for the recovery of Cl and Si (as HCl and SiF_4) during the HF decomposition of stone meteorites

(P62o). Rayudu described briefly eleven radiochemical procedures he performed on proton-, neutron-, and alpha-irradiated targets for disintegration-scheme studies by Professor S. Jha's group in the Physics Department (P64k; see Section B). A number of procedures were developed for the separation and radiochemical purification of Sc from large volumes of Ca solution in the search for ^{48}Ca - ^{48}Sc natural radioactivity (Section C). A unique method was developed by Diethorn for the preparation of CH_4 from carbon in natural terrestrial materials for radiocarbon dating (Section F). Unique procedures for separating carbon from stone and iron meteorites as CO_2 and purifying it for measurement of cosmic-ray-induced ^{14}C were developed by Goel (Section E). For the activation-analysis search for and measurement of ^{129}I in natural materials, several procedures were used by Edwards and Ray for extraction of the iodine and preparation for irradiation and for radiochemical purification for counting (P66a,P67b). A method for preparation of concentrated HF of extreme purity, especially with respect to Pb, was developed by Lancet and Huey in connection with U,Th-Pb dating studies (P73h,J74a).

M. Instrumental Developments

A general discussion of the nuclear instrumentation used in the work described in this Report has been given in Part III, Sections C and D. In some cases standard commercially made instruments were used. In other cases special instruments were designed and fabricated for our needs.

This was particularly necessary in the search for and study of natural radioactivities where the absolute and/or specific activities are weak. Mulholland (J52a,J52d,D52a) constructed an internal-sample proportional counter of 1650 cm^2 area and used it in a low-background arrangement for beta and x-ray measurements in rare-earth samples. The central wire was supported by a frame cantilevered from one end so that cylindrical sample holders could be introduced from the other end. The counter was improved by Selig (D54a) and used by him for studies of a number of elements.

In his study of weak alpha activities, Macfarlane used a low-background counter of similar size (1200 cm^2) but different design and somewhat larger center wire. It was regarded as an ionization chamber, but was actually operated as a proportional counter with very low gas gain ($\sim 1.5 \times$). Because most of the primary ionization from alpha particles occurred in a region of small potential change, the resolution was good in spite of the 2π geometry (P57g,D59a,J61c).

Small low-background Geiger counters needed for cosmic-ray-induced radioactivities in meteorites were built by Elmann (P57g,D57b,J58c,J58e). Each of these gas-flow counters had a thin flat window on one side of a cylinder, and four were mounted together, windows down above sample-holding slides, in a low-background assembly. Improvements to the system were made by Almodovar (D60a), and it was also used by Cressy (D64a).

These multiple-detector assemblies each originally used about 20 large sealed anticoincidence guard Geiger counters. Because of leaks or gas degradation their performance was non-constant and their lifetimes were limited. The situation was improved on both counts by converting them to Q-gas flow Geiger counters by Henry and Viswanathan, who gave a diagram, dimensions, and performance characteristics of the adopted design (P63q).

Cressy also built a low-level beta system with four somewhat larger counters, each having a thin cylindrical wall and being cantilevered from one end so that a cylindrical sample holder could be slid over the tube from the other end (D64a).

A small internal-sample proportional counter for x-ray-emitting samples was described by Kaye (D63a) and Cressy (D64a). Kaye also modified Macfarlane's ionization chamber by reducing the wire diameter so as to obtain a high-gain proportional counter for large x-ray-emitting samples of low specific activity.

Counters for ^{14}C in terrestrial samples (Diethorn) and meteorites (Goel) have been described in Sections F and E, respectively.

For gaseous beta- and x-ray-emitting samples, especially argon isotopes, small proportional counters were designed by Schall and Viswanathan (P63n). For background reduction the counter was surrounded successively by a cylindrical mercury shield, a multiple-anode cylindrical anticoincidence Geiger counter, and a thick iron-brick shield (Schall, P66m). The guard counter was redesigned and rebuilt by Huey (P67k).

A large 4π proportional counter was designed and built by Rightmire and Simanton for study of the ^{26}Al radiations. It consisted of two identical counters on opposite sides of a multiple-thin-foil sample support, and was operated in a low-background assembly (P57g, D57a, J58b, J58d, J59a).

A low-level γ - γ coincidence-counting assembly consisting of two 3-in $\times$ 3-in NaI(Tl) scintillators within a large cylindrical plastic anticoincidence guard scintillator all inside of a massive shield was described by Toia (P65p, P66n, P67j), and its operating characteristics were explored by Noyce (P68f) and Konish (P69k). As mentioned in Subsection C.2, Ritter set this up for operation in the sum-coincidence mode and explored its operating characteristics in that mode (P72m).

A system for on-line measurements of short-lived alpha emitters produced by high-energy protons in the synchrocyclotron, designed and partially constructed by Mesojedec (P62c, P63h), has been mentioned in Section B.

Experiments and design considerations for a position-sensitive proportional counter based on a resistive center wire were described by Yocum and Monahan (P70d). These experiments were extended by Ritter (P71h), but were discontinued when it was found that the objectives had been achieved and surpassed by others.

McKelvey undertook calculations of the fraction and energy distribution of large-angle back-scattered alpha particles in low-geometry absolute alpha counting. The results indicated that the back-scattering contribution is $\sim 0.1\%$ for a gold backing (P51f). The spectrum calculations (P52g) were not completed.

Mulholland derived general formulae for the absorption and self-absorption of monoenergetic photons (x-rays) in 50% (2π) geometry and made some illustrative numerical calculations (D52a). Plans to extend the theory and calculations to the counting yields of photons in cylindrical counters (P53f) were not carried out.

Zero-thickness counting yields for a number of beta-emitting nuclides in three types of low-level beta counters were determined empirically by Shedlovsky et al. (P62q). A more detailed study of ^{26}Al counting yields for

various thicknesses of Al_2O_3 was made by Schall (P64m).

In Subsection C.1, under the discussion of ^{187}Re , it was mentioned that by use of high gas amplification and low electronic bias it was found possible to detect one-electron events in a proportional counter. This makes the sensitivity essentially equal to that of a Geiger counter (P56f).

In connection with the design of large high-pressure proportional counters for radiocarbon dating (Section F), Diethorn found that existent theories of proportional-counter gas amplification could not account for the measurements made by Diethorn and Simanton on the amplification in methane-filled counters with different center-wire diameters, gas pressures, and applied voltages. A new theory was developed by Diethorn and the writer; this yielded a formula for the amplification as a function of two physically significant parameters characteristic of the gas, the counter dimensions, the pressure, and the voltage. A linearized plot of the measurements yielded the parameters for methane. This enabled confident predictions for higher pressures and voltages (P56f, D56a). (This theory has been tested by others with other gases and found to work quite well in general.)

Before coming to Carnegie, the writer had developed a method of determining nonlinearity corrections of counters by multiple-paired-source measurements. The data were used to evaluate the first two parameters (one being the resting-detector deadtime) in a power-series representation of the relationship between the true and recorded counting rates, and least-squares analysis was employed. In the writer's course in Radiochemical Techniques in the 1960 Spring semester, it was suggested by Traytl and several other students that the evaluation of the parameters could be made quite simply by a graphical method. The method was described and illustrated by an extensive set of test data on a Geiger counter, with a discussion of its applicability to other types of counters, by Zwitkowitz (P61v). The method was further tested and found quite effective for methane-flow proportional counters and a NaI(Tl) well-type gamma counter by the writer and by several students in the Radiochemical Techniques course in the 1962 Spring semester (P62r).

The writer made and described a thorough study of the performance characteristics of commercial methane-flow proportional counters used for beta-activity measurements. It was found that simple theoretical considerations could predict the general shapes of bias and voltage curves quite well, but that actual counters deviated from ideal behavior in annoying and incompletely understood ways. Important aspects of the design of the counters and of the electronic circuitry were pointed out. For the best counter in the laboratory, the nonlinearity measurements described above were extended to over 2,000,000 counts per minute, where the nonlinearity correction was ~15%. The behavior was decidedly different from that predicted by simple "deadtime" or "sleeping-time" equations, but the empirical two-parameter equation permits very precise relative activity measurements over a very wide range of counting rates (P62r).

Mössbauer instrumentation, including a novel system incorporating a constant-velocity mechanical drive, is discussed in Section J.

General discussions of the nuclear instrumentation, with many details, were given by Toia (P65p, P66n, P67j, P68i, P69i) and Haramic (P70h, P71g, P72k).

An early attempt to rejuvenate a discarded commercial mass spectrometer was mentioned in Part III, Section E and discussed in more detail by Huff (P64d; see Section A of this Part).

The Nuclide mass spectrometer which was acquired in 1966, and which has been a mainstay of our experimental facilities since then, was described and modifications which have been made from time to time were reviewed in Part III, Section E. The modifications were described in detail by Harsmic (P70h, P71g, P72k) and others (P73g, P76j). None of the features of the instrument are novel, but the combination is probably unique.

N. Miscellaneous Activities

Here are collected a number of items of diverse nature which do not fit into the other Sections of Part V of this Report.

The writer discussed evidence indicating that the pairing energy of protons in nuclei generally exceeds that of neutrons except for light elements, several possible theoretical explanations, and the bearing of this effect on the complete beta-liability of some odd-Z elements (J52b).

Jones made a study of the systematics of spallation yields from medium-light elements ($10 < Z < 30$) with 300-400 MeV protons, and developed formulae for accommodating most of the then available data to about a factor of 3 (D56b). Predictions from these formulae were used for the calculation of half-life exclusion limits for unknown nuclides sought in bombardment experiments (Section B).

The writer studied empirically correlations between comparative half-lives and orders of forbiddenness of long-lived beta disintegrators and improved his previously published beta-stability and alpha-energy diagrams. These systematics were useful for predicting the properties of unknown radionuclides (P55f), but were not of publishable merit.

During extensive atomic-bomb testing in 1955 and 1956, the writer made numerous measurements of radioactive fall-out in Pittsburgh and of radioactivity in the city water. Several test explosions were followed by highly radioactive rain-fall and short-lived radioactivity in tap water, and long-lived radioactivity in tap water increased (P55e, P56g).

The writer prepared a compilation on Natural Radioactivity for a comprehensive Radiation Hygiene Handbook compiled and edited by H. Blatz (R59a). The first part, Natural Radionuclides, listed all known primary and secondary natural radionuclides and their properties and mentioned known induced natural radionuclides. Particular attention was given to the gamma radiation of the primary and secondary radionuclides and secular-equilibrium mixtures headed by ^{238}U , ^{235}U , ^{232}Th , and ^{226}Ra , the latter without and with 0.5-mm Pt shielding. Under Naturally Radioactive Materials were listed uranium and thorium minerals and potassium minerals with their contents of the respective elements, and the contents of these elements in rocks were discussed. (Some of the data were used (uncritically) by Blatz in a subsequent monograph under his sole authorship.)

The writer discussed the feasibility of reactor production of ^{40}K for tracer use via the reaction $^{39}\text{K}(n,\gamma)^{40}\text{K}$. Calculations, including both formation and neutron-capture destruction of all isotopes, yielded the K isotopic composition as a function of thermal-neutron fluence. The ^{40}K reaches a maximum enhancement over its natural isotopic abundance of $\sim 200\times$ at a fluence of $\sim 5 \times 10^{22} \text{ n cm}^{-2}$; an irradiation for a year in a flux of $10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ would yield an enhancement of $\sim 45\times$. It should be feasible to produce kilogram quantities of such material in production-type reactors. Using low-level counting techniques, ^{40}K should be much more suitable as a tracer for potassium than the 12.4-h ^{42}K (or the 22.2-h ^{43}K), particularly in long-time experiments, agricultural experiments, and biological experiments with large organisms (P61w). A paper prepared for publication in *Nucleonics* was rejected because no substantiating experimental data were included. The writer was unsuccessful in persuading the AEC's Isotopes Division or others to conduct an experimental test of the idea.

In connection with Sarma's mathematical formulation and calculations on the effects of "pseudo-diffusion" on radionuclide distribution in terrestrial ocean sediments (D64b; Section F), he also derived formulas for a similar situation expected in the floor of lunar maria. Here the sedimentation results from settling of dust rather than precipitation from water, the radionuclides are produced by cosmic rays rather than carried down in sediments, and the mixing is due to micrometeorite impact rather than to swimming and burrowing creatures (P62e, P63j). (In actual cores of the lunar regolith the radionuclide distributions turned out to be too irregular to be represented by a diffusion-like mixing.)

In analyzing thick-target nuclear-reaction data (Sections A,E), the writer was struck by the fact that standard input-data weighting factors for least-squares fitting to analytical functions give unreasonable results when the data have relatively large uncertainties and hence occasional large errors. It was shown that in the case where positive and negative errors of the same absolute magnitude are equally likely, more reasonable results are obtained with weighting factors equal (or proportional) to the inverse square of the calculated rather than observed values of the quantities; but this situation is unrealistic for inherently positive quantities with large errors. In such situations positive and negative errors corresponding to equal error factors should be equally likely, and the remedy is to use the inverse square of the geometric mean of the observed and calculated value of each input quantity. This procedure prevents data with large error factors from making extremely large contributions to the sum of squares of residuals and thus completely dominating the adjustment of the parameters of the function (P69k, J70a).

The great variety of magnitude scales in use in astronomy is confusing, partly because of their arbitrariness and partly because of the lack of a logical relationship between the scales for different quantum energies. The writer conceived and proposed a universal system of magnitudes based on power flux. This is in turn based on the concept of the expression of spectral (monochromatic) power fluxes as logarithmic derivatives. The system embraces spectral, integral, partial-integral, luminance, and absolute magnitudes, and its use would facilitate meaningful intercomparisons of power fluxes and magnitudes in all parts of the electromagnetic spectrum from radio to gamma radiation. The scheme was described at a meeting of the American Astronomical Society (J70b). A detailed but incomplete manuscript giving illustrative examples from various branches of astronomy was issued as a special report (S70b).

and given limited distribution. The basic mathematical formalism was presented in a Progress Report (P70m). (The normalization of the scale was given differently in S70b and P70m; that of the former is now preferred.)

The Julian-Day Calendar is a consecutive-day counting system which is used extensively in astronomical work. It is particularly useful for relating events occurring in different months and years. The writer has long felt that it could be employed profitably in radioactivity work, especially when following activity curves over long periods of time, and has so presented and recommended it in his courses. In the 1971-1972 Progress Report he described the Calendar and its origin, outlined the procedure for calculating a time in Julian Day and decimal fraction, gave several tables to facilitate the calculations, and presented several illustrative calculations (P72n). In that same Report a practical application was made: growth-decay curves of beta activity of scandium samples isolated from large quantities of calcium by Yeager and counted for about 10 months by the writer were presented with the time axis given in Julian Days (P72j; see Section C).

In connection with Huey's studies of meteorite chronology via lead-isotopic composition and his search for extinct natural radioactivity of ^{205}Pb via thallium-isotope anomalies, he determined the thallium and ^{204}Pb contents of a number of chondrites. He used the data and thermodynamic considerations to calculate accretion (condensation) temperatures from both elements for ten of the meteorites according to the two-component model of E. Anders and J. W. Larimer. In eight cases concordancy (to $\sim 15^\circ\text{C}$) was obtained, the indicated accretion temperature for the ordinary chondrites being $\sim 500^\circ\text{C}$. On a Tl versus ^{204}Pb plot, most of the points fell close to a theoretical curve which assumed 25% matrix (low-temperature component) and the remainder (mostly chondrules) a high-temperature component free of volatile elements (including Tl and Pb) (D72a, P72e). The data were privately communicated to Larimer, who published similar calculations utilizing them.

Lancet discussed the theory of nucleosynthesis of "L-elements" (Li, Be, B) and the observations of their abundances in stars and the Solar System, noting particularly reports of variability of the $^7\text{Li}/^6\text{Li}$ ratio. He initiated a study of this ratio in meteorites, including separated minerals therefrom, with particular interest in the possibility of obtaining indications of different formation histories of different meteoritic minerals (P72f). Chemical (P73h), mass -spectrometric (P72f), and isotope-dilution (P73h) procedures were developed, but time limitations prevented their application to meteorites.

The potential of lead-isotope measurements for studies of environmental pollution in industrial regions was discussed by the writer. The problem of environmental lead was reviewed briefly, and the use of lead isotopes to gain information on the sources and distribution of environmental lead was reviewed more thoroughly. Various ways of representing lead-isotope composition graphically were considered, and it was concluded that plots of $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$, as are commonly used in geochemical and geophysical work, displayed one above the other with a common $^{206}\text{Pb}/^{204}\text{Pb}$ scale, were most revealing. Such plots made of the isotopic compositions of lead from ancient and modern mining regions, the latter from measurements by Keisch and Callahan using our mass spectrometer (Section K), showed clear grouping according to source with relatively little overlap; this

provides the basis for fingerprinting applications in attempts to identify the sources of lead in works of art, environmental pollution, etc. Similar plots were made of all available data on pollutant-lead sources (gasoline, tetra-ethyl lead, coal, fly-ash from coal) and on environmental samples (aerosols, water and snow, and vegetation and soils in the vicinity of highways). A separate plot displayed together data on environmental samples and on leaded gasoline. In some cities there were striking correspondences, though this was not always the case. It was pointed out that no such studies had been conducted in heavily industrialized regions, such as Pittsburgh, where stationary sources are more important than mobile sources in general environmental pollution and would be expected to contribute importantly to lead pollution as well. It was proposed that such studies in the Pittsburgh area be undertaken, and could conceivably be important for evaluating the effectiveness of proposed lead-pollution control measures (P73f). However, the Atomic Energy Commission showed no interest in supporting us in such studies.

Nevertheless, with the help of undergraduate students working for academic credit, some small-scale exploratory experiments were undertaken (P73g). Svec collected open-air fall-out samples and extracted lead for spectrophotocchemical analysis. Menis extracted lead from leaded gasoline for similar analysis. However, both students graduated before the analytical techniques were perfected and isotopic analyses could be attempted.