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Nuclear Chemistry Division

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Annual Report FY83

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Cover—Collage showing activities in the Nuclear Chemistry Division's clean-room complex. The complex is divided into several rooms with different functions; this provides a high degree of cleanliness, contamination control, and flexibility. We use the complex for chemical separations and mass-spectrometric analyses that must be conducted in a clean environment to ensure that chemical or isotopic contamination does not compromise results.

PREFACE

Our intent in the annual reports of the Nuclear Chemistry Division is to provide a timely summary of research activities pursued by members of the Division during the preceding year. Throughout, details are kept to a minimum; readers desiring additional information are encouraged to read the referenced documents or contact the authors.

The Introduction presents an overview of the Division's scientific and technical programs. Next is a section of short articles describing recent upgrades of the Division's major facilities, followed by sections highlighting scientific and technical advances. A brief overview introduces each section. Reports on research supported by a particular program are generally grouped together in the same section. Section 7 lists the scientific, administrative, and technical staff in the Division, along with our visitors, consultants, and postdoctoral fellows. It also contains a list of recent publications and presentations.

Some contributions to the annual report are classified and only their abstracts are included in this unclassified portion of the report (UCAR-10062-83/1); the full article appears in the classified portion (UCAR-10062-83/2). We intend that these two documents provide the reader with complete information about the programs and capabilities of the Nuclear Chemistry Division.

The preparation of this report required the talents and participation of many members of the LLNL Technical Information Department. Cynthia Talaber supervised the work, and she and Jay C. Cherniak were the editors; Bill Fulmer and Brett Clark designed the report and coordinated the production of figures and layout; Marion Capobianco did the typesetting. Special mention must also be given to Nancy Shaw, who typed the original drafts of the entire report, and to the secretaries in the Division—Joyce Plis, Pamela Beringer, Faith Phillips, and Amelia Wilcox, who worked closely with the authors to put the drafts in their final form.

Gordon Lee Struble

Nuclear Chemistry Division

MASTER

UCAR--10062-83/1

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Annual Report FY83

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Introduction

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1 Introduction

The primary mission of the Nuclear Chemistry Division has been to provide diagnostic information on the performance of nuclear explosives, using radiochemical techniques, for use in defense and in peaceful applications. We do this by sampling the highly radioactive debris produced by a nuclear explosion, accurately determining radioactive species, and interpreting the analytical results in terms of device performance. Division members participate in all aspects of this effort.

Following a period of rapid development and expansion during the 1960s, weapons support to the Division was significantly reduced in the last decade. This decline has recently been reversed. However, during the period when traditional funding sources declined, scientists of the Division adapted and applied their knowledge and expertise to a variety of problems in other fields. Thus, although the Division still has the

primary mission of supporting the weapons programs, it has significantly increased its involvement in other Laboratory programs.

The transition from a division supported by major programs to one receiving support from a large number of small and diverse programs has created a number of problems in administration and communication. To minimize these problems, the Division is attempting to serve only those programs that require skills in nuclear science. However, we interpret this constraint very broadly.

The Division provides both routine services and research and development for 26 separate programs. Support from these programs includes money for staff salaries, expenses, and capital equipment and facilities. The number of Division staff used by a program provides a convenient measure of its relative size, although this measure does not reflect variations in expense

Figure 1. Nuclear Chemistry Division programmatic-support levels for FY83.

and capital equipment money and the level of engineering involvement.

Figure 1 shows the programmatic-support levels for FY83. In some cases, several small programs are grouped in a single category. In all of these programs, the Nuclear Chemistry Division attempts to maintain a balance between routine programmatic duties and sophisticated applied and fundamental research.

Department of Energy (Weapons)

- **Test Program**—The Division develops and uses radiochemical techniques to diagnose the performance of underground nuclear explosions.
- **Chemistry Program**—The Division conducts long-range research and development for the nuclear explosives programs.
- **Laser Program**—The Division performs mass-spectroscopy measurements for the laser isotope-separation efforts and provides advice on radiochemical diagnostics for the inertial confinement fusion project.
- **Other Weapons**—The Division makes routine measurements to help a number of small programs develop techniques to detect undocumented releases of radioactivity. This work also includes support for the joint United Kingdom and United States nuclear weapons test program.

Department of Energy (Nonweapons)

- **Magnetic Fusion Energy (MFE)**—The Division conducts experimental and theoretical

research on 14-MeV-neutron-induced damage in construction materials of interest to the MFE program.

- **Nuclear Waste**—The Division supports a number of small programs that address issues of fission-product containment in underground repositories and at nuclear-detonation sites.
- **Environmental Sciences**—The Division supports programs that address issues of radionuclides in the environment of the Livermore Valley in California, of Nevada and Utah, and of the South Pacific Ocean.
- **Office of Basic Energy Sciences (OBES)**—The Division conducts experiments to accurately measure the half-life of ^{187}Re so that it can be developed as an accurate cosmological chronometer.
- **Safeguards Technology Program (STP)**—Because the accountability of special nuclear materials is of great concern to the weapons and nuclear-power communities, the Division develops sophisticated instrumentation to quickly and quantitatively measure fissile materials in nuclear-material production and reprocessing facilities.

Reimbursable Programs

- **International Safeguards Project Office (ISPO)**—The Division develops sophisticated instrumentation to measure fissile materials in nuclear-material production and reprocessing facilities for the Department of State.

Facilities

2

2 Facilities

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Overview

The Nuclear Chemistry Division operates several major facilities for LLNL. Most of these are located in Livermore, but we also have permanent experimental facilities in Los Alamos, NM, and Berkeley, CA.

Most of the Division's offices and laboratories are located in Building 151 at LLNL. All routine chemistries involving moderate levels of radioactivity are performed in this building. In addition, it houses shops, counting facilities for radioanalytical chemistry (located in the basement two stories below ground level, which is well shielded against external sources of radiation), a mass-spectrometry laboratory containing six solid- and gas-source instruments, laboratories dedicated to measuring very low levels of radioactivity in environmental samples, and central processing units for an extensive unclassified timesharing computer network. The Division also occupies additional office and laboratory space in Building 281 and an adjacent trailer, intended primarily for students and visitors and for employees who work on completely unclassified projects.

In this section, we describe progress in two projects for major additions to Building 151. The first project is a major upgrade of our mass-spectrometry facility that has involved upgrading or replacing our current instruments and installing a clean room for the preparation of samples. The second project is a complete upgrade of our information-processing and communication systems. We are implementing completely new methods of data acquisition, storage, and retrieval for most of our routine analytical measurements. The plan involves centralized data storage and analysis capabilities built around VAX computers. Satellite workstations having identical hardware configurations are used to control measurements, to temporarily store unanalyzed data, and to move blocks of data to the central data bases. We report on major advances in both the hardware

and the software necessary to fully implement this system. In addition we are installing extensive electronic-office facilities. We intend to automate the transfer and retrieval of information—both scientific and administrative—through the use of personal workstations.

The Division routinely deals with large quantities of very radioactive material. Experiments or processes requiring special containment or remote handling are performed in Building 251, which contains high-level nuclear-chemistry laboratories equipped with glove boxes and hot-cell facilities. The building, which also contains special shop areas and an office complex for a small resident staff, was recently renovated extensively to meet stringent DOE safety requirements and to provide new capabilities. This year we report on the upgrade of a laboratory intended to produce sources and targets of rare-actinide isotopes and the upgrade of our plutonium-analysis facility.

The Division maintains permanent experimental facilities at the Lawrence Berkeley Laboratory (Berkeley, CA) and the Los Alamos National Laboratory (Los Alamos, NM). We configured Cave 3 at the Lawrence Berkeley Laboratory's 88-in. cyclotron to provide intense beams of heavy ions for the production and characterization of rare isotopes and for the study of heavy-ion reaction mechanisms. At the Los Alamos National Laboratory's three-stage tandem Van de Graaff laboratory, we are developing a permanent station for measuring gamma rays, electrons, and light nuclei produced during the course of nuclear reactions. Such data give valuable information on the structure of atomic nuclei. This year we report on the theoretical characterization of a superconducting solenoid at the Los Alamos facility that permits the high-resolution measurement of internal conversion electrons.

A Clean-Room Complex for Mass Spectrometry

S. Niemeyer and G. P. Russ

A new clean-room complex for mass spectrometry that provides a high degree of cleanliness and contamination control was designed and constructed this year. Preliminary tests indicate that air cleanliness in the work areas rates better than Class 10 (i.e., 10 particles per cubic foot with a diameter greater than $0.3\ \mu\text{m}$); typical laboratories rate on the order of Class 1,000,000. Construction materials were selected to provide high resistance to degradation from chemical attack. Because the complex is divided into several rooms of differing functions, and because work areas are divided into numerous well-isolated spaces, this facility has the flexibility to meet future needs as they arise.

The imminent occupation by the Mass Spectrometry Group of a clean-room complex marks a major milestone in the renovation and upgrading of our facilities. The primary goal of this complex is to provide a chemistry and mass-spectrometry laboratory that is well isolated from isotopically anomalous materials handled within the Nuclear Chemistry Division. This facility will enable us to embark on a basic research program to measure isotopic effects in terrestrial and extraterrestrial samples; an auxiliary function will be to provide ultraclean reagents and labware to other Division members when such needs are imperative. In addition, it represents the establishment of a technology for contamination control, which may well prove beneficial for the needs of other Division personnel, especially with regard to avoiding potential cross-contamination between isotopically dissimilar samples.

The clean-room area is divided into several rooms, each with a different function, in order to enhance flexibility. The layout is shown in Fig. 2:

- The **gown room** provides a progressively cleaner entryway as one enters the complex. Gowning procedures include exchanging street shoes for permanent clean-room shoes and donning a hood, coveralls, and shoe covers that enclose the ends of the coveralls. This uniform (except for the shoes) is changed twice a week and laundered before re-use.

- The **wash room** is designated for cleaning all supplies and equipment before entry to the clean room; it also provides a place to transfer ultraclean reagents to outside the complex. Foot traffic will not be permitted through this room.

- The **instrument room** houses a new VG354 mass spectrometer, a filament bake-out device, and mobile furniture (not shown on the diagram). This room is designed to accommodate another instrument of comparable size; the removable partition gives a full 5-ft passageway. The temperature of the area occupied by the spectrometer is controlled to within $\pm 1/2\ ^\circ\text{F}$ to enhance the ability to perform ultrahigh-precision isotopic measurements.

- The **chemistry laboratory** is where most of the operations pertaining to chemical separations and sample loading will take place. The east end will house an ion-exchange unit for producing clean water and sub-boiling quartz stills for making ultraclean water and acids. The small adjoining room contains a desk at which documentation tasks will be handled. Ultraclean work areas are available both with and without exhaust out of the complex: (1) The fume hoods have a vertical laminar flow of air down onto the work surface; the air is then exhausted through holes in the work surface. A small amount of room air is drawn into the front face, but it penetrates the work area only a couple of inches before it is drawn through the work

surface. Hence, the hoods exhaust all noxious fumes generated in the hood, but without contaminating the hood interior with room air. (2) An island work counter with a plexiglass shield hanging from the ceiling provides exceptionally good protection from contamination from the rest of the clean room. Air flows down from ceiling filter modules onto the work counter and then out into the room. The maintenance of a good laminar flow allows people to work at 4-ft intervals without cross-contamination. This feature, together with the number of fume hoods, permits several

researchers to concurrently carry on independent projects without compromising each other's work.

- The **powder room** is for storing and handling rock samples; it will include microscopes for visual inspection and equipment for mineral separations. The solid samples are then dissolved in preparation for chemical separation.

- The **acid room** is primarily for "dirtier" operations that pose potential contamination and/or corrosion problems in the chemistry lab. For example, labware will be cleaned here

Figure 2. Layout of the clean-room complex for mass spectrometry. The two doors that are shown closed are for emergency use only. Crosses on cabinetry units indicate fume hoods. All air-supply and exhaust ducts run above the upper structural ceiling; the indicated "air supply" is where make-up air is brought up from the basement to above the clean room. Monitoring and control equipment, including electrical breaker boxes, is located outside the complex in the hallway just outside the gown and wash rooms.

because of the large volumes of acid involved. Because of the possibility of higher acid-fume concentrations, air flow in this room is "once-through" rather than recirculated.

The basic design of the airflow pattern is a modified vertical laminar flow. Conditioned make-up air is dumped into an air plenum formed by a new upper structural ceiling and a lower T-bar ceiling. Each HEPA (high-efficiency particulate air) filter is clamped into a module upon which a blower is mounted; this ceiling module sits on the T-bar ceiling so the blower withdraws air from the plenum and drives it through the HEPA filter into the room below. Air is exhausted from the room through fume hoods, door grills, and return-air grills in the side walls. The return-air grills contain prefilters to remove the bulk of the particulates, and the air is then drawn up through the sidewall and back into the plenum. Each room has enough HEPA modules to change the air in the entire room at least twice a minute. Airflow patterns within the rooms are designed to minimize cross-contamination between work areas and to inhibit transfer of particles from the floor to work levels. The entire complex is maintained at a positive pressure with respect to ambient pressure so that any air leaks cause air to leave the complex. Furthermore, the plenum and side walls are at a negative pressure with respect to the room; consequently, leaks in the lower ceiling and walls result in air moving from the room back into the plenum. Thus, all air that enters the clean room is forced to pass through a HEPA filter, except for air passing through doorways. The desired flow directions through doorways, as shown by the arrows on the diagram, are established by maintaining appropriate pressure differentials between the various rooms. Since air continually flows through door grills, turbulence upon opening a door is lessened and upstream migration of particulates is minimized.

Because of the critical importance of maintaining proper pressure differentials, the entire complex has a back-up air supply that is activated whenever the main supply loses power. A control box at the clean-room entrance contains controls and time delays to switch all air-handling equipment on and off in the appropriate sequence so that proper pressures and airflow patterns are maintained at all times. Since the necessary equipment is on the building's emergency-power system, the integrity of the clean room is preserved during power outages and routine maintenance of either air-supply system.

An important aspect of this clean-room design was to use non-particulating construction materials that are resistant to acids and chemicals. Metal fares particularly poorly in this type of laboratory since most metals readily corrode in the presence of hydrochloric and nitric acid, our most common acids. (Even stainless steel is corroded within a year of exposure to low levels of HCl fumes.) This is especially damaging because steel contains a number of elements of potential interest for mass-spectrometric analyses. Thus, we eliminated metal wherever possible, and where metal was needed, the preferred choice was anodized aluminum. Where steel was required, it was coated in some manner. A few examples of our approach to corrosion prevention are: steel wall studs are plastic-coated; seismic restraints on lights and HEPA modules are polypropylene rope rather than the customary steel hanger wire; the galvanized hanger wires for the T-bar are encapsulated in shrink tubing; the T-bar is anodized aluminum; the usual light-diffuser assemblies are replaced by gasketed polypropylene sheets bolted directly to the T-bar; all utility conduits (except for single gas and oxygen lines) are PVC; all metal fasteners in the furniture are covered or coated; valves for water, nitrogen, and vacuum supplies are PVC; door hinges are plastic; and sprinkler heads are protected by a vapor-deposited polyethylene material. The fume hoods are especially noteworthy, since they are constructed almost entirely of polypropylene using a seam-welding technique. Some other construction materials of importance for cleanliness and durability are: ceiling and wall panels are industrial-grade particle board coated with electrostatically deposited polyester which is then UV-cured; the floor covering is an extruded PVC that is coved for easy cleaning; doors are plastic-laminated; countertops are constructed of solid epoxy-resin cast material that is highly immune to acid leaching of trace elements; and the HEPA module units are constructed of fiberglass that is non-particulating and acid-resistant.

Since construction is not quite complete (99+ % finished), final evaluation of the clean room's performance is pending. In terms of maximizing air cleanliness (quoted as a Class number, which represents the number of particles per cubic foot with a diameter greater than $0.3\ \mu\text{m}$), the remaining important tasks are to scan over the entire face of each HEPA filter to locate and repair leaks and to give a final cleaning of the entire complex. Yet preliminary tests in the presence of ungowned personnel

(people are the primary source of dirt once a clean room is operational) indicate particle counts about an order of magnitude lower than specified. For example, the Class 100 work counters generally rate better than Class 10 (typical laboratories are on the order of Class 1,000,000). Therefore, we fully expect that this

clean-room complex will perform as designed, giving a high degree of contamination control and cleanliness in an environment that is highly resistant to chemical attack, and with a built-in flexibility to meet future needs as they arise.

This research was supported by the LLNL Nuclear Test Program.

The Information Upgrade Project

J. B. Carlson

The Nuclear Chemistry Division deals with a great deal of data, both scientific and administrative in nature. The scientific data alone accumulate at the rate of about 2×10^8 bytes per year; the accumulated data base is approximately 4×10^9 bytes. The Information Upgrade Project is a multifaceted effort to introduce state-of-the-art information technology in order to increase individual productivity. Its basic aim is to allow easy access to both scientific and administrative information in a coherent and integrated manner.

A sophisticated data-base-management system (DBMS) will be needed if Nuclear Chemistry personnel are to continue to be able to use this data base effectively. The current methods of collecting, organizing, and storing the data involve a great deal of manpower merely to move data from sources to destinations. The large variety of media (cards, paper tape, magnetic tape, etc.) and data formats makes an integrated facility almost impossible.

With current information technology, users should be able to deal directly with information, and not have to search for data or worry about media and format changes. The basic intent of the Information Upgrade Project is to develop information systems that will allow fast access to correct and up-to-date information. This means that all data must be collected and stored in a standard format in a logically centralized location. The information systems dealing with all types of information (scientific, technical, administrative, etc.) must also be integrated (along with external information systems) into one information-management system.

This ultimate goal is not currently attainable with today's technology. The various information systems that deal with administrative and scientific data (even within scientific systems) are not amenable to this integration. This is why the various phases of the Upgrade Project are proceeding down somewhat independent paths, to allow the systems to be installed and made operational within a finite time span. A time line of estimated completion dates is shown in Fig. 3. The intention is to proceed in the development

with integration in mind, and merge the systems as technology becomes available.

The Upgrade Project has five distinct areas: Counting Facilities Upgrade, Local Data Analysis, Local Data Interpretation, Office Automation, and Personal Workstations. Each of these areas deals with an information-intensive problem that can be attacked relatively independently of the other areas. They have been included in the scope of the project to assure some level of compatibility between them, a necessary prerequisite for integration into an information-management system. The first three areas deal with scientific needs and the last two deal with administrative or computational needs.

The Counting Facilities Upgrade phase deals with the data-acquisition needs of Nuclear Chemistry. The Division operates nuclear-radiation-counting and mass-spectroscopic facilities to perform analytical analysis of isotopic species for programmatic studies. While the primary responsibilities are for the Nuclear Test Program, a variety of other users such as the Environmental and Safeguards programs use the facilities. The data accumulated from measurements of samples are currently transferred using a wide variety of methods (punched cards, magnetic tapes, etc.) to the Laboratory's 7600 mainframe computers for analysis and interpretation. Much of the present facility was developed in the late 1960s and 1970s, and there has been little major improvement in the instrumentation or data paths since then. A much-improved and

integrated system to handle the counting needs of the facility is needed.

The first step in the Counting Facilities Upgrade is to provide a standardized instrumentation controller, which will allow flexibility both in equipment configurations (to allow for new analytical techniques) and in data flow paths. The latter will allow us to store collected data in a central data base for easy access by analysis-and-interpretation codes. The major goals of the Counting Facilities Upgrade are twofold: first, to replace old nuclear instrumentation with current state-of-the-art equipment; second, to provide a standard mechanism for centralized data access and control.

The Local Data Analysis phase involves the local analysis of counting data on an in-house VAX minicomputer. The counting data will be retrieved from a central data base, along with all parametric data necessary for analysis. The analysis results will also be stored in a central data base for subsequent use by the data-interpretation codes.

The Local Data Interpretation phase involves the writing of codes to allow interpretation of the analyzed counting data on the in-house VAX minicomputer system.

The current emphasis in these three phases is on the Counting Facilities Upgrade, since its functioning is a prerequisite for the other two phases.

In the Office Automation phase, we installed a

commercially available product, CEO (Computerized Electronic Office) from Data General Corporation, in February 1983 for trial use. The present level of acceptance of this product has been so high that the computer system used for the test has been severely overloaded for its primary function. To continue the development in this area, a dedicated MV/4000 minicomputer has been ordered to operate the office systems. This will allow expansion of the system from its present 20-terminal limit to approximately 40 terminals, as well as eliminating the slow response to users.

The Personal Workstation phase of the Upgrade Project involves development of local computational and display needs for users. The IBM PC personal computer has been selected as the standard personal workstation. A great deal of software to handle administrative and scientific information needs is available for these machines, which makes them valuable tools. In addition, they are currently seen as the key to the creation of the integrated information-management system. One approach to allow interaction with a multitude of incompatible information systems is to place an intelligent "gateway" between the systems and the users that will allow access to the systems transparently. The personal workstation may provide such a gateway.

This work was supported by the LLNL Nuclear Test Program.

Figure 3. Time line for the five phases of the Information Upgrade Project.

The Counting Facilities Upgrade Project

D. N. Hunt¹ and J. B. Carlson

The Nuclear Chemistry Division operates facilities to perform quantitative isotopic analysis using nuclear-radiation-counting and mass-spectroscopic techniques. We began work on the Counting Facilities Upgrade Project (a phase of the Information Upgrade Project), in an effort to modernize both the counting equipment and the information techniques used in the facilities. The end result will be to integrate the different counting systems into one unified system.

The Nuclear Chemistry Counting Facilities are charged with support of Nuclear Test Program diagnostics. In addition, the facilities are a Laboratory-wide resource, providing ongoing support for the Safeguards Program, the Environmental Program, geological research, and basic research in nuclear science. Comprising a collection of more than 100 counting stations with a variety of configurations, the facilities perform alpha, beta, gamma-ray, and x-ray counting and spectroscopy in addition to magnetic mass spectroscopy.

The Laboratory has required radiometric analysis systems since its inception. With the need to maintain a state-of-the-art facility, the Nuclear Chemistry Division has continued both to improve earlier systems and to develop new systems. The current facility, housed in Building 151, contains a variety of nuclear-radiation counting systems. Although the facility performs modern, sophisticated analyses, users of the different systems are encumbered by a variety of operating procedures, a number of manual operations, and an assortment of output media.

The Counting Facilities Upgrade Project was undertaken to upgrade the facilities' control and operation into one integrated system. The major goals include a standard interface to the scientific user with menus customized to his needs, computer control of existing equipment, and automatic data acquisition and archiving. Computational resources for data reduction will also be provided. Another major benefit of the upgrade will be the ability to integrate future developments with more ease.

The requirements for the counting facilities called for a central data base that will be accessed by the Division's data-reduction codes. Distributed instrumentation control and data collection local to the user's instrumentation were also required. Given these requirements, we developed a system consisting of a set of workstations networked to a central computer. The workstations are responsible for the acquisition of data, while the central computer will archive and reduce the data. This system, the Nuclear Chemistry Counting Facilities Network (NCCF-NET), is the end product of the Counting Facilities Upgrade Project. The network is implemented using hardware available from Digital Equipment Corporation (DEC) and DECNET, a DEC networking software package.

The workstations are based around the DEC LSI-11/23 microcomputer running the RSX11M operating system (see Fig. 4). The workstation is responsible for the user-machine interface, count scheduling and maintenance, instrumentation control, data acquisition, local data storage, and transmission of data to the central computer. The workstations attached to the NCCF-NET will be distributed throughout the building, in the vicinity of the counting stations that they control. Each workstation supports from one to ten counting stations.

The user interfaces with the workstation through a video terminal. The terminal displays a set of menus that provide for sample entry, acquisition parameter entry, count scheduling, and status display. The set of menus is

customized for the equipment that the scientist is using. In addition, as the facility users schedule their counts in a variety of ways, the format, entry, and maintenance of the schedules are tailored to the counting station configuration.

At this point, the network connection between the workstations and the central host computer is used for file transfer. Data files containing the data acquired by the instrumentation are transmitted to the host via this connection. In addition, these data files are stored locally in the workstation. The disk units on the workstation have sufficient storage to maintain the data collected for a 72-h period. This allows the workstations to operate unattended when the host computer is unavailable. This loose coupling between the host and the workstation also increases the reliability of the network as a whole—no single unit, host or workstation, can drastically affect the remainder of the network in the event of a crash.

Control of nuclear instrumentation is one of the major tasks of the workstation. This instrumentation includes scaler/timers, pulse-height analyzers, and sample changers. The workstation is responsible for setup, initiation,

and termination of counts using the schedule specified by the scientist. In addition, acquired data must be read from the instrumentation and stored on the workstation's disk unit. The workstation must be capable of performing these tasks on all instrumentation to which it is attached. The pulse-height analyzer is attached to the workstation via a serial line; the remainder of the instrumentation is attached via a CAMAC subsystem. (CAMAC is an international instrumentation-packaging standard.)

After going through a competitive bid process, the Division decided on the Nuclear Data ND66L as the standard pulse-height analyzer (PHA). This instrument met the necessary requirements and was controllable both from its keyboard and by a host computer. Any command that can be invoked from the keyboard can also be invoked by the host computer. The ND66L communicates to the workstation via a standard serial line using a link-level communication protocol (DDCMP) to assure reliability. The workstation will use a programmable serial line card with DMA capabilities (ISL-11) to connect to the PHA's

Figure 4. Workstation and attendant instrumentation in the Nuclear Chemistry Counting Facilities Network.

serial line. The ISL-11 will transfer commands from the memory of the workstation to the PHA and transfer data from the PHA to the memory of the workstation without the assistance of the workstation processor. These transfers include handling the DDCMP protocol in the ISL-11.

Application codes in the workstation will make use of a library of functional software that will provide a control path to the ND66L. Use of this library will allow the application code to perform any desired function on the PHA via standard subroutine calls. The library includes such functions as: send a command; dump a spectrum; load a spectrum.

In addition to the PHA, the Division requires a scaler/timer to perform gross counting and single-channel analysis. This scaler/timer must accumulate live times, real times, and several data channels at once. In addition, it must be able to terminate a count upon reaching either a preset live time, a preset real time, or a preset count in any of the data channels. Since the scaler/timer will be attached to the workstation via the CAMAC subsystem, it will be packaged as one or more CAMAC modules. Along with the above requirements there is a need for a rapid, accurate dead-time determination and for long count periods. There are no acceptable CAMAC modules commercially available with these requirements.

The Electronics Engineering group is designing a module that will meet the NCCF-NET requirements. This CAMAC module will include multiple general-purpose scalers. The required scaler/timer system will be configured from several of these general-purpose scaler modules. The display for the CAMAC scaler/timer system will be provided by a NIM scaler/timer module, which will be local to the counting station and run in parallel with the CAMAC scaler/timer. Control of the scaler/timer system will be from the workstation via the CAMAC subsystem. A library of functional software will be developed to provide the application code access to the scaler/timer system through standard subroutine calls. Functions supported by this library will include: set scaler/timer; start scaler/timer; stop scaler/timer; read scaler/timer.

The Division uses eight different varieties of sample changer to support the counting stations in the facility. These sample changers range from four positions to 200 positions, from a simple turntable to a carousel with eight stacks of 10 samples. The workstation must have the ability to manipulate all aspects of sample-changer

control, including positive position feedback and counting-system status monitoring. This control capability requires three major parts: an interface to the workstation, an interface to the sample changer, and functional software to drive these interfaces.

To minimize the amount of customized software and hardware needed, the interface to the workstation and the major portion of the interface to the sample changers have been standardized. A small amount of customized hardware will be necessary to adapt each of the eight varieties of changer to the standard interface. The interface to the workstation will be a serial line packaged in a commercially available CAMAC module. The interface to the sample changer will be a CAMAC microcrate. This microcrate will contain a crate controller, an input module, and an output module, all commercially available. The customizing hardware will be designed and built by the project design team.

The functional software supporting the sample-changer subsystem for the workstation will be in the form of a library of subroutines. Application codes in the workstation will have complete control of all sample-changer functions via standard subroutine calls. This library will include such functions as: move sample changer to specified position; search to a position with sufficient activity; report current sample-changer status.

The majority of the instrument control performed by the workstation is supported by the CAMAC subsystem. In addition to the crate directly connected to the workstation, the subsystem also includes the microcrates connected to the sample changers. The major benefit of the use of CAMAC in the network is that most required functions are available commercially and are compatible with the CAMAC standard. This allows not only more rapid initial development, but also facilitates future expansion. The CAMAC subsystem is also supported by a functional software library. This library allows the application codes in the workstation to control all functions available from the modules currently attached to the CAMAC subsystem. The functional software will support not only the crate attached directly to the workstation, but also modules residing in the microcrates at the sample changers.

The majority of the network has been specified, and work on all major sections of the project has begun. Upon conclusion of this

effort, scientists in the Nuclear Chemistry Division will have a modernized, automated counting facility that is customized to their needs. In addition, the network will be readily adaptable to future developments and the future

needs of the Division. We expect that the network will continue to evolve as needs change and as we accumulate experience with it.

This work was supported by the LLNL Nuclear Test Program.

1 Electronics Engineering Department, Lawrence Livermore National Laboratory

Data-Base-Management System

G. G. McMillan

Using a sophisticated new data-base-management system (DBMS), we are beginning to integrate the data (parametric, descriptive, and measured) from sources both internal and external to the Nuclear Chemistry Counting Facilities Network. We chose a DBMS based on the relational approach because it offers the greatest flexibility for our purposes. The system we chose has complete user packages, an interface that can use any of several programming languages, and potential for distributed-data-base applications.

The Nuclear Chemistry Division operates counting facilities for the acquisition of nuclear data. The large amounts of counting and parametric data generated are needed for subsequent analysis and interpretation. In order to catalog and retrieve these data, we needed a sophisticated DBMS. A DBMS allows the applications programmer great flexibility in storage and retrieval of data needed for a particular analysis procedure.

We may conveniently categorize data-base systems according to the approach they adopt for the form of the data model and accompanying data sublanguage. The three best-known approaches are:

- The network approach.
- The hierarchical approach.
- The relational approach.

In the relational approach, associations are represented in the same way as other entities, i.e., as records of relations. In the hierarchical and network approaches, certain associations are represented by means of links. For our purposes the relational approach offers the greatest flexibility.

One such relational DBMS is INGRES, developed by Relational Technology, Inc., for the VAX series computers from Digital Equipment Corporation. This DBMS is the one we have chosen for use in the Nuclear Chemistry Division for support of the Counting Facilities Upgrade Project. It is a fully relational DBMS with complete user packages and potential for distributed-data-base applications. INGRES has a

number of unique and valuable features that made us decide to use it for the Upgrade Project.

QUEL (QUery Language) is the INGRES query language, which allows one to retrieve, manage, and maintain data in an existing INGRES data base. QUEL statements are simple expressions of "what" needs to be done, rather than "how" it should be done. QUEL statements can be used directly from a user terminal or they can be embedded within programs written in high-level languages via EQUQL (Embedded QUEL).

EQUQL is provided as the programming language interface to INGRES. Because EQUQL is essentially the same in all languages, statements used in Fortran, COBOL, BASIC, PASCAL and C, all of which are supported by INGRES, are interchangeable. Also, the interface with the user is independent of the actual implementation of the interface to INGRES. Thus, changes to the underlying implementation that enhance performance or functionality do not affect previously written programs.

A number of utilities allow INGRES to be easily used for retrieval of information from the data base. Query by Forms (QBF) is a visually oriented forms-driven interface to INGRES. QBF allows users to interrogate the data base from a standardized menu.

The INGRES report-writing facilities allow users to create highly formatted listings of data from the data base. The Report Writer can be used to create organized summaries of data for inclusion in other documents and to produce

regular listings of data for management and production needs, as well as for other *ad hoc* purposes. Report by Forms (RBF) is the user interface to the INGRES Report Writer. RBF is a forms-driven visual editor.

Graph by Forms (GBF) is a new part of the total package available in INGRES. GBF provides complete graph-definition and output utilities. It is a forms-based interface used to specify the charts for an application. With GBF the user specifies how to extract out the data from the data base, alter the appearance of the graph, and store the graph in the data base.

Using these tools we have begun to develop a system to integrate the flow of parametric, descriptive, and measured information from all sources, both internal and external to the network (see Fig. 5).

Spectral data originating from a workstation is prefixed with a standard header block that contains identification information and data about how the measured data were acquired. These files are transmitted to the central host computer where the DBMS resides. An executive program will be initiated from the workstation transmitting the file to decompose the header

Figure 5. Physical and logical flow of data in the Nuclear Chemistry Counting Facilities Network (only two workstations are shown). Dashed lines indicate future development.

into the proper records for inclusion in the DBMS tables. These records will be correlated with other data associated with the experiment, as derived from users' direct input, digital balance readouts, etc.

The DBMS provides the nucleus for application codes to retrieve data in a dynamic mode to allow prompt interpretation of experiments. The new GOSPEL code, which will be used to interpret and report data for nuclear-event diagnostics, will rely heavily in its design on this capability to provide users with essentially real-time access to information about

samples. We are currently preparing detailed plans for the structure of the GOSPEL program.

Another design goal for which we intend to utilize the DBMS is the quality assurance (QA) of our instrumentation. By comparing standard spectra or similar criteria against QA samples, perhaps initiated via the DBMS, we will be able to more promptly determine and thereby correct any tendency toward problems in data acquisition.

This work was supported by the LLNL Nuclear Test Program.

The Office Automation Project

J. B. Carlson

We are examining how the administrative processes of the Division can benefit from current electronic-office technologies. An integrated electronic-office system that uses commercially available software was implemented on a trial basis in February 1983; it has been so successful that we have requested hardware for expansion.

The aim of the Office Automation Project (a phase of the Information Upgrade Project) is to apply the information technologies that are familiar to the scientific environment to administrative processes. This project deals with information management in three areas: word processing, administrative information support, and electronic office. The realization that offices are not collections of independent operations, but rather a coordinated set of operations that are logically related, spurred our decision to implement an integrated electronic-office package. The CEO (Comprehensive Electronic Office) software package was chosen because it gave us the integrated environment that was needed. The key components of CEO are: electronic mail, electronic calendaring and resource management, word processing, meeting and conference scheduling, electronic filing, and user tailoring. Implementation of this software was done on a trial basis using the existing ITSS computer systems. Five terminals and two printers were added to the system to allow the software to function.

This system proved to be enormously successful as various groups realized its potential to deal with information. The communications ability of the electronic mail has supplemented the conventional telephone to a larger extent than expected. The ability of users to compose drafts that can be electronically transferred for final typing has saved a great deal of turnaround time.

The system was accepted with such enthusiasm that the computer system (which is still being used for data-analysis operations) has been swamped. This has stunted the growth of

the project, for lack of resources. To address this issue, an MV/4000 processor has been requested to serve as a dedicated CEO machine. This 32-bit minicomputer will allow us expansion as well as much faster response to users. The current users of CEO will be transferred to the new system when it arrives and is installed, approximately in November 1983.

This annual report for 1983 was initially prepared using the CEO system. Documents were either composed directly on the system by CEO users or typed in by a secretary. Once in the system, the document was electronically mailed to the scientific editor for approval or revisions. Documents were then transferred to magnetic tape and carried to the Technical Information Department's Atex phototypesetter, where they were electronically translated into the typeset copy, edited, and put in final format. Using CEO as a central repository for all documents has tremendously streamlined the annual report process, and the electronic transfer to Atex has eliminated a large number of transcription errors.

The word processing needs of the Division include the requirement to type equations and other special constructs. Of the commercially available word processors on the market, the NBI series provides one of the best equation- and scientific-typing packages, so we leased two NBI 4000S systems for a one-year trial. In addition to excellent scientific-typing support, the NBI systems also provide comprehensive communications support that allows both document transfer and virtual terminal support to other computer systems; we need such communications abilities in order to integrate the

NBI systems into the total office system. The NBI systems have been installed and work is in progress to connect them to our CEO system.

The final area of the Office Automation Project concerns administrative-information management and decision support. We are using our CEO package and IBM PC microcomputers for managing data on (and reporting on) fiscal resources, manpower, and project status. The large amount of software available for the PC makes it a valuable administrative tool. In addition, the IBM PC will be linked to the CEO and NBI systems to allow information to be moved electronically between them.

We may integrate the functions of CEO and the IBM PC into a single package, which is made

possible by hardware recently introduced by Data General Corporation. The Model 10 provides a package that supports local CEO word processing and an 8086 coprocessor that supports an IBM PC-like environment (Fig. 6). The Model 30 supports a local multi-user CEO environment with word processing and decision support. Both systems provide a distributed-processing link to a central host for calendaring and mail functions. We will test and evaluate these systems in FY84.

This work was supported by the LLNL Nuclear Test Program.

Figure 6. The Nuclear Chemistry Division's office automation/administrative systems network.

Modernization of the Actinide-Target Facility

R. W. Lougheed, J. H. Landrum, and E. K. Hulet

We established a laboratory with new and modernized equipment for producing actinide targets. The facility includes capabilities for chemical purification and electrodeposition of actinide isotopes, and vacuum systems for sublimation of actinide compounds or *in situ* reduction of actinide oxides to the metal and their subsequent vaporization. An electron-gun vaporization system was built to overcoat targets with thin metallic protective layers; low-geometry counters for final characterization of the targets were also modernized.

The preparation of actinide targets or sources for nuclear chemistry and physics experiments presents many problems that do not occur when using stable elements. Foremost among these problems are the radioactivity and scarcity of many of the actinide isotopes. The radioactive isotopes require special facilities for purification and target preparation. The scarcity of many of the actinide isotopes requires high deposition yields and eventual recovery of the material. These criteria led us to design equipment for our new laboratory that is much smaller than that normally used to prepare targets and to develop specialized designs for recovery of undeposited material. We developed a modern integrated facility for target preparation that includes purification and recovery of the rare-actinide isotopes, and developed several techniques for the actual target preparation and characterization.

A partial list of rare-actinide isotopes from which we have made targets is ^{245}Cm , ^{248}Cm , ^{249}Bk , ^{249}Cf , ^{253}Es , ^{254}Es , and ^{257}Fm . For the isotopes where very little material is available (i.e., ^{254}Es and ^{257}Fm), or in cases where only low target densities ($<0.5 \text{ mg/cm}^2$) are needed, electrodeposition techniques are frequently used. In cases where higher target densities or better stability and adherence of the target material are needed, vacuum-deposition techniques are necessary. Target densities as high as 11 mg/cm^2 of ^{248}Cm metal have been achieved. The targets

are prepared on a small variety of metal substrates with a usual deposit diameter of 2–6 mm.

The vacuum deposition of actinide isotopes is usually performed with one of two techniques. The first method is the direct sublimation of the actinide fluorides¹ from a 4-mm vitreous (glasslike) carbon crucible. A cap containing a 1-mm orifice is inserted in the crucible after precipitation of the fluoride in the crucible. In the second method the actinide metal is vaporized from a tantalum crucible following an *in situ* reduction of the actinide oxide with thorium metal powder.² The crucible charge is prepared by heating the actinide oxalate in air to produce the oxide, which is then mixed with thorium metal and pressed into a 1/8-in.-diam pellet. In both methods the crucible is heated with a radiofrequency power supply to a temperature slightly below that at which the actinides vaporize, in order to remove more volatile impurities, and then heated to a temperature sufficient to deposit the actinides onto a heated metal substrate. Yields are usually 25–30% for a 6-mm-diam target with a crucible-to-substrate distance of 8 mm.

Targets or sources can also be prepared using a third vacuum technique, "flash" volatilization. In this method, the actinides are evaporated in air onto a tungsten filament from nitric or hydrochloric acid solution. The actinides are then vacuum-evaporated by passing a large current

pulse (~ 200 ms wide) through the filament. This technique is particularly well suited for the rapid preparation of thin sources of the less-rare-actinide isotopes. Because of adherence problems with larger amounts of material and difficulty in precise control of vaporization conditions, it is only occasionally used for target preparation.

A summary of the equipment available for target or source preparation is:

- Glove boxes and shielded hot cells for quantitative chemical purification and recovery of actinide isotopes and electrodeposition of targets.
- A nonradioactive-target vacuum system for the development of deposition techniques using stable rare-earth elements.
- A vacuum system for radioactive target preparation (see Fig. 7). This and the above system use cryopumping as opposed to oil-diffusion pumps. A shutter has been included to avoid breaking the vacuum after low-temperature heating to remove impurities. A solid-state detector has been installed to permit *in situ* alpha counting and pulse-height analysis of the targets.

- A vacuum system for "flash" evaporation of actinide isotopes using resistance-heating of a tungsten filament.

- A new vacuum system containing an electron gun for overcoating targets with thin metal layers. The amount of overcoating is determined using an oscillating quartz crystal during deposition. Substrate heating or heating of the source using radiofrequency or resistive power supplies is included in the design.

- Low-geometry counters modified to perform alpha pulse-height analysis. Counting geometry factors of 100 to greater than 10^5 are available for counting highly radioactive targets.

- A pinhole alpha-counting system or autoradiography to characterize target uniformity.

This research was supported by the LLNL Nuclear Chemistry Research Program.

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Figure 7. Apparatus for vacuum sublimation of rare-actinide isotopes. The vacuum chamber is in a ventilated enclosure that can be converted to a glove box by addition of a front window. A hot cell for chemical purification and recovery of the actinide isotopes is on the far right.

Upgrade of the Plutonium-Analysis Facility

A L Prindle, R Gunnink, W D Ruhter, and J B Niday

We moved our plutonium-scrap-analysis equipment into the hardened area of Building 251 during FY83. In conjunction with this relocation we replaced obsolete hardware and gamma-ray-analysis software. The room where the analyses take place was equipped with safeguard and security controls required by LLNL and the Department of Energy.

The Nuclear Chemistry Division is responsible for the isotopic analysis of scrap plutonium from LLNL; these analyses are used for safeguarding and accountability purposes. In the last few years systems have been developed to carry out isotopic analyses using gamma-ray spectrometry instead of mass spectrometry. This method of nondestructive analysis has merit because it removes the need to open the plutonium containers, is cost-effective, and produces results on a timely basis. In FY83 we were required to move the facility for performing these analyses into the recently completed hardened area of Building 251 to meet LLNL safety requirements.

The room where the analyses are performed was upgraded to meet LLNL and DOE safeguard and security requirements. Procedures for two-person surveillance were developed, and

appropriate alarms and locking devices were installed.

During the move to the new area much of the equipment was upgraded or replaced. The small high-resolution Ge(Li) detector that has been in use for several years was replaced with a small high resolution high purity-germanium detector. In addition, a second larger high purity-germanium detector was incorporated into the analysis system. The new counting arrangement consists of a steel shield with an opening in each side to allow the detectors to view the sample, which is located within the shield on a rotating table (see Fig. 8). The detectors are connected to a Nuclear Data 66L pulse-height analyzer through appropriate pulse electronics (see Fig. 9). The system allows acquisition of data from both detectors simultaneously through separate

Figure 8 The new gamma-ray-spectrometry system. A sample is shown on the rotating table within the shield; the two detectors view the sample through openings in the shield.

Figure 9. The pulse-electronics rack for the system, with the ND66L pulse-height analyzer and LSI-11 computer.

analog-to-digital converter modules. With this new system, counting times have been reduced to 1 h from 8–10 h. Since the new system has high-count-rate electronics, we can obtain a higher precision for a given counting interval.

The pulse-height analyzer system is equipped with a Western Dynex Series 6000 hard disk for data storage, which is coupled to a LSI-11 computer for data reduction. We replaced our old software with a new automatic analysis program called MGA (Multi-Group Analysis)

which has been described elsewhere.^{1,2} This new program allows processing of the data collected from both detectors and considers all intense peaks or group of peaks from 90 to 400 keV. The MGA program has options that allow it to consider freshly processed and aged plutonium as well as mixed oxide samples. Provisions have also been made in the program for interferences such as ^{237}Np .

This work was supported by the LLNL ABW Program.

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Computer Analysis of the LLNL Superconducting Electron Transporter

H. I. West, Jr.

We developed computer codes that accurately simulate the transmission of typical solenoidal-field electron transporters used in nuclear spectroscopy. The effects of source and detector size as well as the angular distribution of the source are included in the code.

We developed a computer model of the transmission of the superconducting electron transporter used by the Nuclear Spectroscopy Group at the Los Alamos National Laboratory (Los Alamos, NM). (The transporter is similar to that of Klank and Ristenen¹ and Backe *et al.*²) Figure 10 shows the transporter configuration used in early work. The electrons are detected by a high-resolution Si(Li) detector, so that the only need for the transporter is to separate the source and detector by various baffles and apertures in order to reduce detector background. The transporter is usually used in a stepped-in-B mode, in which the energy region of interest is scanned in B . The operation of the transporter can be expressed by

$$\frac{n(E)}{\Delta E} = \frac{N(E)}{\Delta B/\Delta t} \int T(E,B) dB \quad (1)$$

where $N(E)/(\Delta B/\Delta t)$ is an input function composed of source strength and spectrometer scan rate (No./keV·G in calculation, and No./[(keV·s)(G/s)] in an experiment); $T(E,B)$ is

the transmission (No. units); and $n(E)/\Delta E$ is the response (No./keV). For a given energy, the integral can be written as $\bar{T}B\rho(1/\rho_1 - 1/\rho_2) \equiv \bar{T}B\rho/\bar{\rho}$, where $\bar{T}$ is the average transmission, $B\rho = 1704.0\sqrt{(E/511.0)(E/511.0 + 2)} \text{ G}\cdot\text{cm}$, and ρ_1 and ρ_2 are the minimum and maximum radii of curvature of electrons accepted in the pass band.

The transmission of the transporter can be expressed by the equation

$$dT = \frac{1}{4\pi} \frac{dN}{dt} (\theta_0, \phi_0) \sin \theta_0 d\theta_0 d\phi_0 \quad (2)$$

where θ_0 and ϕ_0 are the usual polar angles. One can evaluate dT by stepping through all values of θ_0 in, say, 0.05° steps, testing the electron trajectory at each baffle, and summing the values of dT in the pass band. The use of Monte Carlo techniques is especially valuable in designing the transport code because of the ease in handling the details of the source and detector.

Figure 11 shows the transmission of the spectrometer for 197.8-keV conversion electrons in the decay of ^{152}Eu . The bottom curve shows

Figure 10. Early configuration of the three-loop baffle electron transporter used by the Nuclear Spectroscopy Group.

the experimental results. The middle curve shows the modeled results, which were obtained by stepping B from 540 through 820 G in 5-G steps. For each step, E was varied randomly over 197.7 to 197.9 keV. Also, for each step 20,000 electrons were started off at the source, uniformly distributed over a diameter of 6 mm. The detector was 10 mm in diameter and 5 mm thick. The results must be corrected for the detector response to make comparison with theory; the detection efficiency is reduced because of the back-scattering of electrons out of the detector and the penetration of electrons through the detector, especially near the edges. At high energies, bremsstrahlung loss is also important.

The upper curve in Fig. 11 shows the experimental data after detector-response correction by means of the SANDYL³ three-dimensional electron-photon transport code available at LLNL. To use the code in the way that best simulates the experiment, we used the code option that allows the individual position and direction cosines of each electron in a group (we used 2000 to 4000 in each group) to be input to the code.

Figure 12 shows a simulation of an experiment in which a wide energy range is scanned. Here B was varied over 300 to 5700 G in 50-G steps. For each step, 50,000 electrons were randomly selected in the energy range 100 to 3000 keV. The transmission calculation was obtained with bins 50 keV wide. The scattered data points are a direct result of the calculation; the solid curve is based on the use of all data points and was obtained by first calculating the average transmission and then using the indicated equation. For the 10-mm detector, $\bar{T}$ was 0.98%. For a 16-mm detector (about the optimum size) $\bar{T}$ was 1.70%. The results vary in energy only through their $B\rho$ dependence, and hence the ordinate for the case of the 16-mm detector is 1.73 times larger.

The results from the SANDYL calculation of the detector response have been multiplied into the computed transmission (again the factor of 1.73 applies). Measured transmissions, normalized at low energy, are shown for comparison. To get good comparison between experiment and calculation at the higher energies, we had to reduce the thickness of the detector in the calculation from the nominal

Figure 11. Comparison of measured and calculated transmission of the spectrometer in Fig. 10 for 197.8-keV conversion electrons in the decay of ^{152}Eu . The middle curve was calculated using the transport code. The bottom curve shows the measurements, and the top curve was obtained from calculated detector-response corrections applied to the measurements.

value of 5 mm to 4 mm. Why this was necessary is not well understood.

We find the ability to calculate the characteristics of the spectrometer to a reasonable degree of accuracy to be quite valuable in spectroscopy work. We are able to calculate changes in baffle-detector configurations easily using our transport codes. Detector-response calculations are not

completely satisfactory, however, but work well for $E \lesssim 2$ MeV.

I gratefully acknowledge E. Henry and W. Stöfl for data they obtained during the development and evaluation of the spectrometer, and L. G. Mann and them for many helpful discussions. This work was supported by the LLNL Nuclear Chemistry Research Program.

Figure 12. Energy response of the transporter. The upper curve was calculated using the transport code. The four bottom curves were obtained from detector-response corrections to the transport-code results and are shown in comparison with data. The 16-mm results should be increased by 1.73.

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Nuclear Explosives Diagnostics

3

3 Nuclear Explosives Diagnostics

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Overview

Nuclear explosives diagnostics is the main effort of the Nuclear Chemistry Division. The Division's missions in the LLNL Test Program are to determine with radiochemical diagnostic methods the fission and thermonuclear yields of the nuclear devices that are tested at the Nevada Test Site, and to determine other details of device performance such as yields of individual parts, burn asymmetries in fissile and thermonuclear fuels, and fast neutron outputs. We conduct research and development in methods to obtain new or more accurate diagnostic information that is desired by the design physicists.

As has been the case in recent years, the majority of our Test Program research and development this past year was directed toward improving our capability to determine the thermonuclear yields of nuclear devices. This effort was in both gas diagnostics and threshold detectors.

In gas diagnostics, we continue to focus on thermonuclear yield, using measurements of ^4He . We tested a new design for a gas-sampling hose that is much stronger than our previous design and incorporates an electrical conductor for downhole pressure transducers and a cliper to measure the length of the hose as a function of time. It is our intention to continue sampling after ground subsidence. Such sampling was impossible with our previous system because the hose invariably broke at subsidence and contact was lost with the device gases. The initial test

showed that the new design is superior, and it will be used routinely. We also measured the radiogenic helium content of the rock near the working point of devices, by comparing laboratory measurements of drill-core samples taken before detonation and measurements of post-shot residual ^4He in devices that produce very little of the isotope through fusion reactions. Results are still imprecise but they are consistent with our previous assumptions about the radiogenic helium content in our gas samples. We have also demonstrated that isotopic analysis of the xenon fraction of our gas samples can provide accurate measures of the fission yield that are sensitive to the fissioning species and to the neutron energy.

This year, we continued a program of calculations and laboratory measurements of neutron cross sections. We use these data to improve interpretation of diagnostic information and to test the adequacy of explosion code models of nuclear device performance.

Finally, we report work on a gamma-ray spectrometer and a 14-MeV neutron generator contained in a package designed to determine the gross composition of geologic material as a function of depth in the emplacement holes drilled for nuclear explosive tests. This information will be very valuable in ensuring the complete containment of radioactivity from future nuclear detonations.

Initial Field Tests of a New Gas-Sampling Conduit

C. F. Smith

A new gas-sampling conduit was field-tested for the first time on the Laban Event on August 3, 1983. The new design incorporates the electrical conductor for the downhole pressure transducers and cliper within the hose-support cable instead of external to it, and has steel cable armor and strength members surrounding the hose. It obtained high-quality gas samples, functioned in the presence of high downhole pressure, returned reliable cliper data, and survived longer than the old system. Emplacement time and cost were significantly less than for the old system.

Development of prompt gas-sampling techniques in the past several years has evolved a reasonably reliable early-time sampling system based on the cable-supported-hose concept. However, the system only rarely communicates with device gas after chimney subsidence. This feature imposes an unpredictable time limitation on the collection of samples. Design efforts were therefore begun to provide a stronger, more flexible and sleeker conduit with the potential to survive subsidence in one or more possible emplacement configurations. Design criteria included significant improvement in strength and flexibility, minimal (or no) degradation in

presubsidence sampling capability, protection of the hose and electrical cabling, and reduction in overall emplacement cost as compared with the cable-supported hose system.

Design details of the new conduit are depicted in Fig. 13. At the core of the assembly is a 14-conductor electrical cable; 12 wires service the four downhole pressure transducers and the other two function as a cliper to continuously record the electrical length of the cable. Pull tests have indicated that the electrical length of this cable is indicative of the physical length of the conduit when failure occurs. Surrounding the electrical cable and inside the polyurethane hose

Figure 13. Design of new conduit for prompt gas-sampling. The external armor gives the conduit greater tensile strength and protects the electrical components.

are three counter-laid layers of twisted wire strands. These support the hose against crushing. The middle layer is missing alternate strands; gas flow is principally through the voids between these strands. Outside the hose is the external armor, consisting of 20 1/4-in. wire ropes. These ropes are constrained by a loose-mesh steel braid and the entire external structure is gas-blocked to prevent uncontrolled transport of cavity gases. The external armor is also the principal strength member of the system. Pull tests have indicated that the new system's breaking strength is approximately 200,000 lb. The burst pressure of the hose has been determined to be 2700 psi. The outside diameter is 2 in.—the same as the old system's.

The old system had an *internal* wire rope to support the hose and external (separate) clipper and electrical components. Tensile loads had to transmit through the hose to the internal cable. Hose damage occurred at approximately 18,000 lb at the hose couplings. These couplings occurred at 20- to 80-ft intervals along the length of the conduit to allow field assembly and facilitate handling; they also provided movement-resistance points along the hose exterior. The external placement of hose, clipper, electrical cable, and pressure transducers made these constituents vulnerable to damage from relative ground motion. Common failures included loss of electrical signals, capricious clipper information, and opening to air at significant distances from the cavity.

Both systems incorporate two vacuum-demand valves to control downhole gas. These valves require that a partial vacuum be established in the hose above in order that gas can pass through them from below. The valves incorporated in the new system provide separate internal terminations for the external armor, the hose, and the internal support strands. Electric cables are gas-blocked and two pressure transducers are contained within the housing, which is also the tension member. These valves are the only interruptions in the otherwise continuous conduit.

Both new and old conduit designs were fielded at Laban. The systems were emplaced to 937 ft and were configured identically. A vacuum tank was used to initiate gas flow in the new hose and the pumping trailer was used to evacuate the old hose. Either system could be diverted to the sampling pumps for collection of gas to be analyzed at LLNL. Pumping began at +3 min, but by +5 min unusually high downhole pressure (>500 psi) forced the old

system's vacuum-demand valves closed. The new system also showed high pressure (372 psi max) but not enough to prevent gas flow. Radioactive gas was obtained at +14 min in the new system, and was diverted into the pumping skid for collection of the first sample. Six samples were taken between +24 min and +93 min: samples 1, 2, 4, and 6 from the new hose, samples 3 and 5 from the old hose after the downhole pressure ebbed sufficiently to permit gas flow (flow at ~+25 min, radioactivity at ~+1 h). Sampling was terminated when both systems showed air in the hoses. Samples 1-5 are of diagnostic quality. Sample 6, taken from the new hose after subsidence, was mostly air. Samples 2, 4, and 5 are noticeably better than 1 and 3, as expected because of their later sampling times. Ingrowth from precursor decay was incomplete in all samples (more so in the old hose samples 3 and 5), indicating premature separation of the gas and suggestive of postdetonation inlet-elevation differences between the systems. Further detailed analyses of these samples and of the field analyses will be required to detect any subtle differences in the sampling performance of the two systems. At this stage in the data processing none have been identified.

Both systems appear (from clipper measurements) to have shortened at the initial ground shock (new hose by ~92 ft, old hose by ~221 ft), and both exhibited restricted flow as compared to predetonation performance. The downhole electric cable serving the old hose was lost during subsurface subsidence at +75 min and the hose opened to air above the upper vacuum-demand valve at +80 min during surface subsidence. The new hose withstood subsurface subsidence but did lose communication with cavity gas at +82 min.

Subsidence at Laban occurred in two distinct stages. Subsurface subsidence at +75 min disrupted our clipper to 244 ft below ground and at +80 min a sudden precipitous drop of the ground surface occurred. The surface drop was unusually extreme and caused stretching and/or breakage of all downhole cables. Both surface hoses connecting to the sampling systems were stretched tight to the crater edge. The electrical cable to the armored hose was pulled back to the trailers and found to be broken at the transition socket to the internal cable. The old hose electrical cable had broken off and disappeared into the crater. Safety considerations prevented re-entry to the crater for further investigation. On the basis of the timing of

events, it is likely that the old hose failed during surface subsidence and that the new hose may have survived. If the 5/8-in.-diam surface hose was stretched to failure or broken off at the transition coupling as was the electrical connector, we conjecture that the armored portion of the system may still be intact. We hope that in time we may re-enter the crater, reconnect to the armored hose, and test this possibility.

Although we have not been able to evaluate its postsubsidence integrity, the new armored hose demonstrated its utility as a prompt gas-sampling conduit. We intend to proceed with the new design as the principal conduit for prompt sampling, and to repeat the "standard" emplacement configuration (straight, stemming supported) on the Branco Event in September.

This work was supported by the LLNL Nuclear Test Program.

Radiogenic ^4He Released in Underground Nuclear Tests

D. A. Leich and C. F. Smith

Thermonuclear-yield determinations based on measurements of device-produced ^4He require correction for ^4He released from the surrounding rock. This correction can be made if the ^4He content of the rock is known and if the amount of rock that is degassed can be determined. We made helium measurements on core samples from the Nevada Test Site (NTS) and, to estimate the amount of rock degassed, compared them with analyses of gas samples from three NTS events that had been corrected for air and device ^4He . The results are statistically consistent with the nominal 2 ± 1 kt rock/kt yield assumed previously, but suggest that this value may be low.

The LLNL Nuclear Test Program's gas-sampling project focuses mainly on efforts to measure the ^4He produced by thermonuclear explosives. (This measurement is the least ambiguous determination of TN yield available in an underground test.) To determine device-produced ^4He , it is necessary to correct the measured total for helium contributed by air dilution of the samples and for radiogenic ^4He released from the surrounding rock. The air contribution is obtained from post-shot measurement of neon or argon where the pre-shot helium/neon or helium/argon ratio in the pore air is well known. The rock contribution can be estimated from pre-shot measurements of radiogenic ^4He concentrations in the rock and an estimate of the quantity of rock degassed based on the explosive energy of the test.

In making pre-shot rock ^4He measurements, we are attempting to characterize a spherical volume with samples taken along one diameter. It is therefore necessary to exercise great care in sample selection. A suitable suite of samples includes cores from each of the geologic formations present near the working point and represents the range of grain size, porosity, and lithology that is observed *in situ*. The region of interest extends from the working point to one-half the expected cavity radius in both directions, and samples are usually obtained from at least nine depths distributed throughout this interval.

Each sample of about 200 g of core is ground, sieved (-100 mesh), dried, blended, and split.

Helium analyses are performed by vacuum-melting 200-mg splits of each core sample in an on-line molybdenum radiofrequency-induction crucible and analyzing them in our Nuclide static noble-gas mass spectrometer. Splits are carefully weighed, wrapped in aluminum foil, and loaded into a 10-sample turret inside the extraction-system vacuum enclosure. The extraction system is then evacuated and pumped for several days with gentle heat ($\sim 100^\circ\text{C}$) applied to the turret chamber. When a suitable vacuum ($\sim 10^{-8}$ torr) is obtained, extraction blanks are run until an acceptably low blank level is reached. The analysis of an individual sample is initiated by rotating the turret to position that sample above the crucible, which has been preheated to about 600°C . The extraction volume is then sealed and the sample is dropped into the crucible by opening a sliding trap door in the bottom of the sample wheel, allowing the sample to fall to the bottom of the crucible. The rf power is then increased to raise the temperature to a maximum of 1650°C , where it is held for 20 min and then turned off, allowing the crucible to cool.

Extracted noble gases are admitted directly into the Gas Analysis Sample Purifier (GASP) where they are purified first by exposure to hot titanium-zirconium (800°C) and copper-copper oxide (450°C) while the extraction is in progress,

and then by exposure to a 450°C zirconium-aluminum alloy getter. Heavy noble gases (argon, krypton, xenon) are adsorbed onto an activated-charcoal trap at liquid-nitrogen temperature, and the remaining gases, primarily helium and neon, are admitted into the mass spectrometer. Complete analyses (for all five gases) are usually performed on one sample from each emplacement hole, requiring sequential desorption and admission into the mass spectrometer of the heavier noble gases. For the remaining samples, only helium is measured and the remaining gases are discarded. While the mass spectrometer used for these

analyses is normally used for isotope dilution mass spectrometry (IDMS) of noble gases, these analyses are not done by IDMS, but by the less precise method of peak-height comparison. For each sample, the ^4He ion beam current is measured, blank-corrected by subtracting the ^4He ion beam current observed in the most recent blank run, and then compared with the blank-corrected ^4He ion beam current observed in analyses of our air standard containing a known amount of ^4He (0.26 nl per sample). Variations in sensitivity determined by repeated analyses of the air standard are normally at the $\pm 10\%$ level for helium and are the limiting

Table 1. Measurements of ^4He extracted from NTS core samples.

Hole	Geologic medium	Depth (ft)	^4He (nl/g)
U2fd	Alluvium	1180	$0.79 \pm 10\%$
		1220	$0.94 \pm 10\%$
		1260	$0.96 \pm 10\%$
U2cp	Paintbrush tuff	1040	$0.98 \pm 10\%$
		1068	$0.43 \pm 10\%$
		1100	$0.21 \pm 10\%$
		1136	$0.76 \pm 10\%$
		1160	$0.47 \pm 10\%$
U9cr	Tunnel beds tuff	1080	$0.55 \pm 10\%$
		1100	$0.35 \pm 10\%$
		1120	$0.26 \pm 10\%$
		1150	$0.94 \pm 10\%$
Ue4ae	Alluvium	1430	$0.87 \pm 10\%$
		1455	$0.90 \pm 10\%$
		1470	$0.81 \pm 10\%$
U2fe	Mixed tuffaceous alluvium	1220	$1.17 \pm 25\%^a$
		1240	$1.49 \pm 25\%$
		1280	$1.51 \pm 25\%$
		1320	$1.05 \pm 25\%$
U9cn	Tunnel beds tuff	930	$0.45 \pm 25\%$
		950	$0.32 \pm 25\%$
		970	$1.43 \pm 25\%$
U20ag	Quartz-bearing rhyolite lava	2010	$0.65 \pm 25\%$
		2090	$1.01 \pm 25\%$
		2170	$0.63 \pm 25\%$

^a Uncertainties of 25% for this set of ten measurements are due to an unusually large sensitivity variation during the analyses.

factor in the precision of these analyses. Blank corrections are sometimes as large as 10%, but are usually only a few percent.

We measured the radiogenic ^4He concentrations in 25 samples from various depths in seven NTS emplacement holes used for recent nuclear tests. The results are given in Table 1 and compared with previous data (obtained in 1969 and 1970 by R. Crawford and P. Crawford of the LLNL Chemistry and Materials Sciences Department using a different analytical system) in Figs. 14 and 15. Our results for alluvium samples cluster about 1 nl/g and are in reasonable agreement with, albeit slightly lower than, the median of the old data which lies at 1.5 nl/g (Fig. 14). The recent results from analyses of tuff samples cluster about a 0.5-nl/g median, which is noticeably less than the alluvium measurements and in marked contrast to the older results, which have a median value of 1.5 nl/g and exhibit a wide range in their ^4He content (Fig. 15). It is not clear at this time whether the difference is due to an analytical discrepancy, or to chance in the sense that the recent tuff samples happened to belong to the low end of the distribution shown in Fig. 15 for the old data. (It should be noted, however, that seven of the 15 recently analyzed samples were from the same geologic formation as the older samples that gave the largest ^4He concentrations.) We plan to continue our analyses and to conduct studies in sampling and analytical techniques that will clarify the nature of these discrepancies between data sets.

Given that the crucial question, for test diagnostics, is the amount of ^4He contributed to the cavity gas from the rock, the laboratory measurements of $^4\text{He}/\text{g rock}$ must be coupled with an estimate of the amount of rock being degassed. Certainly a lower limit is set by the melt volume: typically, 0.7–1 kt rock/kt yield. A reasonable upper limit might be 3 kt rock/kt yield, which is the estimate used for containment

calculations of the amount of rock heated to sufficient temperature to evolve CO_2 from carbonates. The median of 2 ± 1 kt rock/kt yield appears a useful working value. Using our recent results, then, we expect 2 ± 1 litres of ^4He per kiloton device yield in the cavity gas from rock. Typically, the ^4He residual is 50–100 litres/kt thermonuclear yield so that the uncertainty is innocuous except in very high fission/fusion designs.

Figure 14. Comparison of measurements of ^4He concentrations in NTS alluvium core samples.

Figure 15. Comparison of measurements of ^4He concentrations in NTS tuff core samples.

Table 2. Calculated amounts of rock releasing ^4He in NTS tests.

Hole	Geologic medium	nl $^4\text{He}/\text{g rock}$	litres ^4He from rock kt yield	kt rock releasing ^4He kt yield
U2cp	Paintbrush tuff	0.7 ± 0.3	4 ± 6	6 ± 9
U20ag	Rhyolite lava	0.8 ± 0.3	6 ± 3	8 ± 5
U2fe	Alluvium	1.3 ± 0.2	8 ± 4^a	6 ± 3^a

^a Very preliminary result—device calculations not yet available.

Instead of attempting a rock ^4He subtraction in these high fission/fusion devices, we tried to determine the rock ^4He contribution by correcting the measured ^4He , after normal air correction, for device ^4He using calculated residuals. These experimental measurements suffer from uncertainties in the air correction, in the device-performance calculation, and in the analytical problems associated with measurement of the low concentrations involved. Results are summarized in Table 2. Taken at face value, the data suggest more rock ^4He than we would have predicted. However, the large corrections and data spread make the results quite imprecise.

Two observations are in order. First, it cannot be determined from these data that the experimental results are disparate from the 2 ± 1 kt rock/kt yield median value described previously. Second, we are not aware of a mechanism for ^4He release from the quantities of rock indicated by the data if taken at face value. If there is a discrepancy, it quite likely lies in the sampling and analytical uncertainties of both the cores and the post-shot gas. Clearly, the definitive experiment lies in obtaining good samples of rock and gas from a very low (zero) fusion event. We are searching the Test Program event designs for just such an opportunity.

This research was supported by the LLNL Nuclear Test Program.

Neutron-Fluence and Tritium Measurements for the Hotspur Program

N. A. Bonner, D. R. Nethaway, and E. Goldberg¹

The Nuclear Chemistry Division participated in a series of irradiations at the Livermore Insulated Core Transformer (ICT) facility as part of the Hotspur Program to study thermonuclear burn. The goal of these experiments is to measure accurately the production of ^4He and ^3H in bulk ^6LiD irradiated with 14-MeV neutrons. Our participation has involved making the tritium measurements and using detector foils of aluminum, niobium, and gold to characterize the 14-MeV-neutron source (position, size, and strength) and to measure the neutron fluence at the sample locations.

A series of irradiations at the Livermore ICT facility was started in late 1982 as part of the Hotspur Program to study thermonuclear burn.² The goal of these experiments is to measure accurately the production of ^4He and ^3H in bulk ^6LiD irradiated with 14-MeV neutrons. The Nuclear Chemistry Division participated in each of the irradiations by making the tritium measurements and by using detector foils to characterize the neutron source as to position, size, and strength, and to measure the neutron fluence at the sample locations. The experimental program included a number of short irradiations used to study the neutron-source and detector cross sections and two long irradiations of 48 h each using a large ^6LiD cylinder. The target arrangement with the ^6LiD cylinder in place is shown in Fig. 16.

We used detector foils of aluminum, niobium, and gold for the neutron-fluence measurements. The $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reaction cross section is the most accurately known of the threshold reactions, with an uncertainty of less than 0.5% in the 14-MeV region. Since some of the ICT irradiations were long compared with the ^{24}Na half-life, we also used the $(n,2n)$ reactions on niobium and gold, which lead to longer-lived products. The cross sections for these reactions are also well known, and were verified relative to the aluminum reaction in several of the preliminary ICT irradiations. We found that the cross sections we use for niobium and gold at

approximately 15 MeV are correct within about 2%.^{3,4} We also determined that fluence measurements based on the two proton-recoil counters at the ICT are low by 2% relative to the aluminum reaction.

By placing detector foils at several positions on the beam axis we were able to determine the neutron-source radius to be 4.2 mm, in good agreement with autoradiograph measurements. We were also able to locate the source center to within 1 mm in the x-y plane and to within 0.3 mm along the beam axis. These measurements were made by punching an array of small disks from the detector foils placed close to the neutron source, and determining the distribution of induced activity in the foils. The observed distribution was then compared to that calculated with the TART program for a particular neutron source.

One of the important nuclides to be measured in the Hotspur Program is tritium, produced principally by the reactions $^6\text{Li}(n,\alpha)^3\text{H}$ and $^7\text{Li}(n,n',\alpha)^3\text{H}$. Ideally, the targets should be LiD , since the bulk material in the experiment is LiD , but the amount of tritium in deuterium is much too large. As a result, ^6LiH was chosen as the target material. The background of tritium in these samples was a small fraction of the amount of tritium produced in the least active samples.

The samples for tritium measurements consist of 1-g pellets of ^6LiH sealed in lead. The

Figure 16. Experimental arrangement for measuring neutron fluence and tritium at the ICT facility, showing the large ${}^6\text{LiD}$ cylinder in place.

capsules are 1 in. in diam and about 0.14 in. thick. This combination of materials turned out to be very convenient from the standpoint of the chemical procedures necessary to produce hydrogen gas for counting the tritium.

The samples were put into a vacuum system, which was then pumped out. About 80 cc of mercury was introduced during the pumpout. When all of the air had been removed, the sample, now in contact with mercury, was heated. The mercury dissolved the lead container, thus exposing the LiH to mercury. At elevated temperature mercury reacts with LiH to form hydrogen gas and lithium amalgam. The amount of hydrogen formed was measured to be

sure the reaction had gone to completion. A portion of the hydrogen was then transferred to internal proportional counters for tritium measurement.

The internal proportional counters used should be capable of absolute measurements of the tritium. However, to be sure of the absolute accuracy, they are being calibrated against a National Bureau of Standards tritium standard. The calibration is not yet complete, but it appears that our final numbers should have an uncertainty of less than 5% on an absolute scale. The relative precision will probably be 2% or better.

This work was supported by the LLNL Nuclear Design Program.

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Test Diagnostics Based on Isotopic Measurements of Xenon

G. B. Hudson

Isotopic analyses of xenon recovered from underground nuclear explosions can supply considerable information for nuclear-fission diagnostics. Xenon's low natural abundance and many relatively long-lived isotopes make it uniquely useful in providing fission-product data. Since the data are derived from isotopic measurements, they are not affected by elemental fractionation effects. Preliminary measurements of xenon from a nuclear test demonstrated our ability to precisely determine xenon fission products.

Xenon is a unique element for the measurement of fission products in underground nuclear explosives tests. If isomeric states are counted as isotopes, xenon has 18 isotopes with half-lives greater than 9 h. Of those, fission products can be measured for eight. For these nuclides (^{127}Xe , $^{129\text{m}}\text{Xe}$, $^{131\text{m}}\text{Xe}$, ^{133}Xe , $^{133\text{m}}\text{Xe}$, ^{134}Xe , ^{135}Xe , and ^{136}Xe), relative fission yields vary by a factor of about 10^7 ; the composition of fission xenon is thus quite sensitive to the species that is fissioning and to the nature of the fission reaction.

Because of its low natural abundance, xenon can be traced isotopically. That is, stable separated ^{124}Xe of known quantity can be placed with the device to produce accurately measurable shifts in ^{124}Xe abundance in the underground gases. Table 3 outlines the economics of noble gas isotopic tracers. The advantage of an isotopic tracer is that the resolution of the tracer from the background element relies on isotopic ratios as opposed to elemental ratios. Elemental ratios are more difficult to measure and are liable to significant physical fractionation because different elements are not treated identically in transport processes.

The technique of noble gas mass spectrometry plays an important role in the measurement of several xenon isotopes. Figure 17 shows an example of the precision of the isotopic analyses. Isotopic ratios are shown as deviations, δ_i , in parts per thousand from the isotopic composition of xenon in air:

$$\delta_i = \left[\frac{(^i\text{Xe}/^{129}\text{Xe})_{\text{measured}}}{(^i\text{Xe}/^{129}\text{Xe})_{\text{air}}} - 1 \right] \times 1000 \quad ,$$

where the index i refers to different xenon isotopes. We estimate that δ_{136} values can be measured to an accuracy of better than 1%, even when a large amount of isotopically normal xenon is present as a tracer.

An essential ingredient in obtaining this high precision is the ability to make corrections for mass fractionation effects from the raw data. For promptly sampled cavity gas (less than 10 h after detonation), fission products make effectively no contributions to ^{132}Xe , ^{131}Xe , ^{130}Xe , ^{129}Xe , ^{128}Xe , etc. Thus these isotopes can be used to measure mass fractionation effects, typically 3 to 6 parts per thousand per mass unit. These effects are linear and are corrected for by making a linear transformation such that $\delta_{132} = 0$:

$$\delta_i^{\text{corrected}} = \delta_i^{\text{measured}} - \left(\frac{i - 129}{3} \right) \delta_{132}^{\text{measured}} \quad .$$

Figure 17 shows an example of the nature of the data reduction. In addition to stable isotopes, ^{135}Xe ($t_{1/2} = 9.09$ h) can be measured using mass spectrometry, allowing an independent check on the gamma-ray spectrometry measurements. Table 4 shows several preliminary comparisons.

The most serious problem in interpreting xenon fission products is that some are formed as iodine and tellurium, which subsequently

Table 3. Costs and abundances of separated noble gas isotopes.

Isotope	Cost (\$/cm ³ STP ^a)	Concentration in air (ppm)	Tracer required ^b (cm ³ STP/kt)	Cost (\$/kt)
²² Ne	0.25	1.0	10,000	2500
²¹ Ne	10	0.05	500	6000
³⁶ Ar	3	30	3 × 10 ⁵	10 ⁶
⁷⁸ Kr	25	0.004	40	1000
¹²⁴ Xe	150	0.0001	1	150
Natural xenon	0.01	0.1	10 ⁴	100

^a At standard temperature and pressure.

^b Based on the assumption of 10¹⁰ cm³ STP of air per kiloton and sufficient tracer to double the abundance of the isotope in the cavity gas. For natural xenon, 10 times the background abundance is used.

Figure 17. Measured isotopic compositions of xenon in three replicate analyses of one sample from a recent nuclear test. Data are shown in terms of their deviation δ_i (in parts per thousand) from the isotopic composition of xenon in air. Note that the δ_i measurements are reproducible to better than 1%.

Figure 18. Three-isotope graph of $^{134}\text{Xe}/^{136}\text{Xe}$ vs $^{135}\text{Xe}/^{136}\text{Xe}$ as measured by mass spectrometry of noble gases, illustrating the time evolution of fission-produced xenon. Numbers next to data points indicate the time of sampling in days.

decay to xenon. Thus the isotopic composition of fission xenon evolves in time in a potentially complex fashion. Some simple systems exist, however. About one-half of the ^{136}Xe is produced directly in fission and the other half as ^{136}I , which decays rapidly to ^{136}Xe ($t_{1/2} = 85$ s). Thus there is relatively little chance of iodine-xenon fractionation, and the measured fission ^{136}Xe represents the entire chain yield. $^{131\text{m}}\text{Xe}$ is due almost entirely to independent-yield fission yield since ^{131}I produced in fission decays predominantly to the ground state of ^{131}Xe .

The ^{134}Xe and ^{135}Xe abundances, however, change substantially during the time involved in prompt gas sampling. Figure 18 shows the evolution of the isotopic composition of xenon. The isotopes ^{134}Xe , ^{135}Xe , and ^{136}Xe are a mixture from independent-yield fission and from the decay of fission-produced iodine. (Tellurium and antimony fission products make only small contributions to xenon at early times and are neglected in this discussion.) The dashed curve represents the integrated xenon composition due to the decay of iodine precursors, and the numbers denote the time (in days) at the positions on the curve. The large circle in the lower right-hand corner represents independent-yield fission xenon, and the solid curve represents the mixture of independent xenon and iodine-derived xenon. The curves are based on measured fission chain yields and estimates of the most probable charge in fission.^{1,2} Data obtained from mass spectrometry are shown for four prompt gas samples (solid circles) from a recent nuclear test.

The isotopic evolution shown in Fig. 18 is similar to that estimated. Differences in apparent ages (i.e., position on the evolution curve) and in the times the samples were taken appear to represent the transport time between separation

of the gas from iodine precursors and its collection in the sample bottles. At least two factors are at work. First, the gas is sampled outside the cavity and must pass through several meters of crushed rock. Second, the cooling of the cavity leads to progressive condensation and segregation of iodine in the cavity gas, providing a slower evolution of the composition of xenon than expected.

While the data in Fig. 18 conform well to the ideal model (i.e., no iodine-xenon fractionation is seen), sufficient information is available to resolve more complex situations. Given the half-lives of ^{134}I and ^{135}I , the ingrowth to ^{134}Xe and ^{135}Xe is approximately linear for the time scales considered here. Thus in Fig. 18, the dashed curve for iodine-derived xenon is effectively a straight line until 0.04 d. This means that iodine-derived xenon can be represented by a linear combination of only two iodine components. Combining this with one independent-yield fission component of xenon gives three components, and there are three isotopes measured. Therefore, if the independent fission yields of iodine and xenon are known, the fraction of each isotope due to each component can be computed, whence the fraction of ^{136}Xe from independent yield can be determined regardless of complex iodine-xenon fractionations.

Table 4. Comparison of ^{135}Xe concentrations measured by mass spectrometry and two different gamma-spectrometry techniques.

Sample	Ratio of measured concentrations ^a	
	Gross-gas technique ^b	Separated-gas technique ^c
A	1.07	1.10
B	1.11	1.15
C	1.10	1.04
D	1.07	1.10
E	1.08	1.21

^a ^{135}Xe concentration measured by gamma spectrometry divided by ^{135}Xe concentration measured by mass spectrometry.

^b Gamma-spectrometry measurements of xenon in gross unseparated cavity gas.

^c Gamma-spectrometry measurements of xenon separated from cavity gas.

In summary, isotopic determinations of xenon can provide accurate and detailed fission-product measurements which are very sensitive to the nature of the fission process.

This research was supported by the LLNL Nuclear Test Program.

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Detector Development for the Advanced Weapons Concepts Program

A. A. Delucchi and H. G. Hicks

Diagnostics for the first test in the Advanced Weapons Concepts Program will be very difficult because of the low output expected in the experiment. We identified a number of radiochemical detectors that can supply the desired fluence information. The candidates are required both to have low background productions in nuclear tests and to be amenable to low-level measurements in post-shot samples. For two of the candidates, we developed suitable low-level counting methods, and we determined device background productions on two nuclear tests.

Radiochemical Diagnostics Developments for Primaries

R. W. Loughheed and R. J. Nagle

In the past year we alloyed zirconium, ^{237}Np , and ^{238}U detectors with plutonium to measure fast neutron fluences and fluence gradients. Detectors were also positioned external to the fissile material. In addition, we used the gaseous detectors ^{78}Kr and AsH_3 . The detector measurements make possible a more accurate and detailed understanding of device performance by providing the design physicist with data that he can compare against modeling calculations.

Development of an Analytical Method for Zirconium in Plutonium Metal

J. H. Landrum, R. W. Lougheed, H. G. Hicks, E. H. Willes, and N. E. Henry

We developed an isotopic dilution procedure to accurately determine the amount of zirconium alloyed with plutonium metal. (The low concentration of zirconium usually alloyed with plutonium is too small for most standard analytical methods to detect accurately.) After the addition of a known amount of zirconium enriched in ^{91}Zr to a weighed sample of plutonium-zirconium, ion exchange is used to separate zirconium from plutonium and americium. The zirconium isotope ratios are then used to determine the amount of zirconium of natural isotopic composition contained in the plutonium.

In nuclear-explosives diagnostics, there is a need for techniques to provide information about the geometry of device parts at critical times. One approach has been to include detectors that are sensitive to high-energy neutrons, and observe both the first- and second-order ($n,2n$) conversion products. Zirconium, one of several such detectors, has been chosen for this application—primarily because the metallurgy of plutonium excludes most of the other candidates. Although zirconium can be alloyed with plutonium, it is very difficult to do so, and homogeneous mixtures have not been obtained. It is, therefore, important to sample and analyze the alloy thoroughly and accurately. The low concentration of zirconium usually alloyed with plutonium is too small for most standard analytical methods to detect accurately. We chose isotope dilution mass spectrometry, since precise results can be obtained over a wide range of very low concentrations.

Isotope dilution mass spectrometry is a very sensitive technique, but is subject to pitfalls not usually encountered in other analytical procedures. For example, one must avoid, or remove, any species with the same mass or masses as the item of interest and one must ensure that isotopic exchange occurs between the tracer and sample. Also, for this study, extraneous zirconium from glassware and reagents had to be scrupulously avoided. The following precautions were taken, since

microgram amounts of extraneous zirconium could compromise a measurement:

1. Only special high-purity reagents, known to be low in zirconium by our analysis, were used.
2. The volumes of all reagents were kept to a minimum.
3. Reagent blanks were carried through the same treatment as the sample with each set of analyses.
4. All glassware was prewashed with warm 4M HCl, then rinsed with high-purity water before use.
5. Ion exchange resins were thoroughly washed with high-purity 5M HCl before use.
6. Acid concentrations were maintained at $>4\text{M}$ HCl to avoid the formation of zirconyl complexes.
7. The plutonium-zirconium sample was always dissolved in the presence of the ^{91}Zr tracer to ensure isotopic exchange.
8. Filaments for the mass spectrometer were prebaked before use.

The enriched zirconium was obtained from the Isotopes Division of Oak Ridge National Laboratory and was supplied as zirconium dioxide. The oxide was accurately weighed, dissolved in high-purity 6M HCl, and standardized versus an analyzed standard solution of natural zirconium. Table 5 gives the isotopic composition and chemical concentrations of both standard solutions. The zirconium

separation procedure, described below, typically reduces alpha contamination by a factor greater than 10^5 while yielding 40–70% of the zirconium.

In a glove box, the sample is dissolved in the presence of a known quantity of ^{91}Zr while the HCl concentration is kept greater than 4M. Pu^{+3} is oxidized to Pu^{+4} with HNO_3 and gentle heat. The main separation from plutonium and americium is accomplished by sequential elution with different concentrations of HCl from an anion-exchange resin column. The column is loaded and rinsed with concentrated HCl, allowing the Am^{+3} to pass through the column while Pu^{+4} and Zr^{+4} are retained. Zirconium is then eluted from the column with 7M HCl, leaving Pu^{+4} on the resin. A solution of HI is then added to the zirconium fraction and the solution is heated to reduce any residual plutonium to Pu^{+3} . The HCl concentration is adjusted to greater than 10M and passed through a second anion-exchange column. Pu^{+3} and Am^{+3} pass through the column while Zr^{+4} is retained. Zirconium is then eluted from the column with 5M HCl, assayed for alpha activity, removed from the glove box, and evaporated to dryness in preparation for isotopic analysis.

Zirconium isotopic analyses are performed using MSXL, a tandem 60-degree magnetic sector mass spectrometer. The zirconium samples are loaded into a double-filament surface-ionization source using two prebaked rhenium filaments. Typical filament loadings of 2 μg of zirconium give stable 10^{-13} -A zirconium-ion beams for several hours. Data are obtained by ion counting over the mass range 90 through 96. Small isobaric contributions due to molybdenum at

masses 92, 94, and 96 are subtracted using the signal at mass 95, for which there is no stable zirconium isotope, as the molybdenum indicator. A least-squares fit to the corrected ratios is used to determine the relative amounts of isotopically natural zirconium and ^{91}Zr diluent tracer in each sample, treating a linear mass bias as a second fitted parameter. Table 6 shows a typical set of results obtained on the parts of one device. The errors quoted reflect only the statistical uncertainties of mass ratios, propagated through the calculations to obtain parts per million of zirconium, and the uncertainty of the ^{91}Zr tracer calibration (Table 5). A 10% uncertainty was assumed for the reagent blank measurements. No estimate was made for systematic errors such as in weighing or pipetting.

This research was supported by the LLNL Nuclear Test Program.

Table 5. Isotopic composition of the enriched-zirconium and natural-zirconium solutions.

Isotope	Enriched in $^{91}\text{Zr}^a$ (atomic %)	Natural zirconium ^b (atomic %)
90	3.24	51.44
91	94.59	11.21
92	1.63	17.15
94	0.46	17.39
96	0.08	2.80

^a Concentration = 167.4 $\mu\text{g}/\text{ml}$ (average of 4 determinations; std dev = 0.42).

^b Concentration = 200.0 $\mu\text{g}/\text{ml}$.

Table 6. Amounts of zirconium in samples of a plutonium-zirconium alloy, determined by isotope dilution mass spectrometry.

Sample	Total measured zirconium (ppm)	Zirconium in blank (ppm)	Net zirconium (ppm) ^a
1A	786.7	4.6	782.1
1B	788.7	4.6	784.1
2A	794.9	4.4	790.5
2B	795.3	4.4	790.9
3A	651.3	4.0	647.3
3B	651.1	4.0	647.1
4A	682.9	4.3	678.6
4B	683.0	4.3	678.7

^a Std dev = 0.9%.

Method for Measuring Parts-Per-Million Levels of ^{237}Np in Plutonium

R. Gunnink, R. Stone,¹ R. J. Nagle, and J. H. Landrum

The interpretation of certain characteristics of the device performance on some nuclear tests requires an accurate measurement of the ^{237}Np content in the pre-shot plutonium used in the device. We developed a radiochemical procedure whereby the ^{237}Np abundance in plutonium can be measured by germanium gamma-ray spectrometry down to the parts-per-million (ppm) level with an accuracy of a few percent.

A method was required that could accurately measure ^{237}Np in plutonium at the ppm level. ^{237}Np is an alpha-decaying isotope with a relatively long half-life of 2.14×10^6 y. Its presence in plutonium generally stems from the ^{241}Pu isotope of plutonium, which decays principally to ^{241}Am , which in turn decays to ^{237}Np . Although the buildup of ^{237}Np is slow because of the relatively low abundance of ^{241}Pu ($\sim 0.5\%$), and because of the long half-lives of ^{241}Pu and ^{241}Am (14.35 y and 433 y, respectively), a ppm level of ^{237}Np will nonetheless be generated in a few years. Another obvious source of ^{237}Np is its incomplete separation from plutonium in the chemical processing plant.

Several methods were considered for measuring ^{237}Np in plutonium; however, the most promising appeared to be one in which the ^{237}Np would be chemically separated from a sample of plutonium and then quantitatively measured using a germanium gamma-ray detector.

The general procedure is as follows: First, about 1 g of plutonium sample is accurately weighed and dissolved in HCl in the presence of a measured quantity of ^{243}Am tracer. The ^{243}Am decays to ^{239}Np , which can be used to trace the ^{237}Np and can therefore be measured to determine the chemical yield of the separated neptunium fraction. Care is taken to ensure complete exchange of the two neptunium isotopes prior to any chemical separations. In these separations we use specially prepared columns containing Dowex 1 $\times$ 8 (100-200 mesh) anion-exchange resin. The sample solution, which is 12M in HCl and contains

sufficient HI to reduce the plutonium to Pu^{+4} , is first loaded onto a column and then washed with 10M HCl to remove the plutonium. After all but trace quantities of the plutonium is removed, the neptunium is eluted from the column using 5M HCl. A second column separation is used to provide additional purification from ^{237}U , ^{239}Pu , and ^{241}Am , which are the three principal interferences. In the second column separation, the sample is oxidized with HNO_3 and then treated with formic acid to selectively reduce the neptunium while leaving the uranium in the U^{+6} oxidation state.

The ^{237}Np decays by alpha emission to ^{233}Pa . However, an 86-keV gamma ray is also emitted, with a probability of 12.5%. The approximate counting efficiency is 5%. Therefore, a 1-ppm level of ^{237}Np in a gram of plutonium will give a counting rate of approximately 10 counts/min, assuming good chemical yields. Adequate precision can be obtained by counting for a few hours. However, the sample must be decontaminated from uranium, plutonium, and americium by factors of $\sim 10^4$ to remove spectral interferences. The ^{239}Np daughter product of ^{243}Am emits several gamma rays and is used to measure the overall chemical yield. The time of the americium-neptunium separation must be noted, since ^{239}Np has a half-life of only 2.35 d and will begin to disappear as soon as it is separated from its ^{243}Am parent.

Instead of using the 86-keV gamma ray to measure the ^{237}Np in the final sample, one can alternatively measure the ^{233}Pa daughter product of its decay or alpha-count a portion of the sample. Although these are also viable

techniques, our results to date do not justify the additional effort they require.

We have not yet processed a sufficient number of samples to report a precision or accuracy for the method, but our results thus far show that the required decontamination factors are being

attained (see Fig. 19), and sufficient counts are being obtained in the 86-keV peak to indicate that the method is a viable one.

This research was supported by the LLNL Nuclear Test Program.

Figure 19. This portion of a germanium detector spectrum shows that the ^{237}Np content in a plutonium sample is easily detected after it is chemically separated from the sample. ^{243}Am - ^{239}Np tracer is added to determine the chemical yield.

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Isomer-Ratio Calculations for Neutron-Induced Reactions on Deformed Nuclei: the Effect of Recent Discrete-Level Modeling Studies

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We made significant progress in our capability to calculate accurate isomer ratios for neutron-induced reaction cross sections for deformed nuclei. The improvements are primarily due to the recent development of a technique for modeling levels in odd-odd deformed nuclei. We find that the hundreds of discrete levels and their gamma-ray branching ratios provided by this modeling are necessary to calculate isomer ratios in agreement with experiment for the reactions $^{175}\text{Lu}(n,\gamma)^{176\text{m,g}}\text{Lu}$, $^{175}\text{Lu}(n,2n)^{174\text{m,g}}\text{Lu}$, and $^{237}\text{Np}(n,2n)^{236\text{m,g}}\text{Np}$.

We made significant progress this year in our capability to calculate accurate isomer ratios for neutron-induced reaction cross sections for deformed nuclei. The improvements are primarily due to the recent development of a technique for modeling levels in odd-odd nuclei; the details of its development are described elsewhere in this report (see "Level-Structure Modeling of Odd-Odd Deformed Nuclei" in Section 6). Here we present the results of using such discrete-level information in the calculation of isomer ratios for three important radiochemical detector reactions:

1. $^{175}\text{Lu}(n,\gamma)^{176\text{m}}\text{Lu}$ ($t_{1/2} = 3.7$ h, 0.123 MeV, 1^-)
 $^{176\text{g}}\text{Lu}$ (stable, 0.000 MeV, 7^-)
2. $^{175}\text{Lu}(n,2n)^{174\text{m}}\text{Lu}$ ($t_{1/2} = 142$ d, 0.171 MeV, 6^-)
 $^{174\text{g}}\text{Lu}$ ($t_{1/2} = 3.4$ y, 0.000 MeV, 1^-)
3. $^{237}\text{Np}(n,2n)^{236\text{m}}\text{Np}$ ($t_{1/2} = 22.5$ h, 0.076 MeV, 1^+)
 $^{236\text{g}}\text{Np}$ ($t_{1/2} = 1.2 \times 10^5$ y, 0.000 MeV, 6^-)

In the case of the two (n,2n) detectors, both the short- and long-lived products can be measured directly in samples of debris from nuclear detonations. The $^{175}\text{Lu}(n,\gamma)$ reaction is important because both the $^{176\text{m}}\text{Lu}$ and $^{176\text{g}}\text{Lu}$ that are made undergo further (n, γ) reactions to produce

$^{177\text{m}}\text{Lu}$ ($t_{1/2} = 161$ d, 0.970 MeV, $23/2^-$) and $^{177\text{g}}\text{Lu}$ ($t_{1/2} = 6.7$ d, 0.000 MeV, $7/2^+$), which are measured in debris samples.

We performed all of the cross-section calculations with the Nuclear Chemistry Division's version of the STAPRE code,¹ which uses the Hauser-Feshbach statistical-model (compound-nucleus) formalism of complete conservation of angular momentum and parity. For a given incident neutron energy, the STAPRE code calculates isomeric-state populations and gamma-ray cascades in the nuclei of interest. Each nucleus in the calculation is described by a number of discrete levels above the ground state, and above these discrete levels, by a continuum of levels represented by a level-density formula and divided into energy intervals or bins of equal size. Beginning with the highest energy bin, the code allows each state of a particular spin and parity (J^π) to decay: (1) by gamma-ray transitions to another J^π state in a lower-energy bin or to one of the discrete levels in the nucleus, or (2) by particle emission to some other nucleus. The gamma-ray transitions take place according to our E1 and M1 strength function systematics,^{2,3} where the E1 strength function is modeled to have an energy-dependent Breit-Wigner line shape and the M1 strength function follows the usual Weisskopf E^3 energy dependence. The M1 strength function is

usually specified to be 15 to 20% of the total radiation width. The continuum bins are depopulated, one by one, until the discrete levels are reached.

The depopulation of the discrete levels then proceeds according to a method that is part of the technique devised by Hoff *et al.* for modeling deformed odd-odd nuclei (see "Level-Structure Modeling of Odd-Odd Deformed Nuclei"). The essentials for this gamma-ray depopulation scheme are empirical. Within a rotational band, gamma-ray de-excitation feeding of lower-lying members of the band is known to be more probable than transitions that lead out of the band; the M1 + E2 intraband transitions tend to be intrinsically much faster than interband transitions, where a change in at least one of the odd-nucleon orbitals usually occurs. Thus, the features of this model are the following:

1. Excited members of a rotational band will de-excite only to the next lowest member of the band (via M1 transitions).
2. Band-head levels will de-excite according to the rule $\delta K = 0, \pm 1$. If no such level is available, the band head is identified as a potential isomeric state.
3. The energy dependence for all transitions depopulating a band-head level (M1 and E1 transitions) is E^3 .

4. M1 transitions are intrinsically faster than E1 transitions by a factor of 6.

This zero-order approximation of how the de-excitation of a real deformed nucleus proceeds has certain features that model reality rather well and others that are only crude approximations.

Given this version of a de-excitation cascade, it becomes necessary to characterize the odd-odd product nucleus with a sufficient number of discrete levels if a statistically representative result is to be obtained for the calculated isomer ratio. Within this set of levels, many rotational bands must be included in order to obtain a representative selection of K quantum numbers. Also, the levels of each band should be extended to appropriately high values of angular momentum. In order to achieve these conditions, we have modeled the discrete levels of the odd-odd species in question up to 1500 keV for the lutetium isotopes and up to 1200 keV for neptunium.

Table 7 summarizes these modeled sets of levels. They are all derived from empirical data for single-particle excitations and rotational parameters in neighboring odd-mass and even-even nuclei. Existing experimental data for the odd-odd nuclei, including level energies, spins, and parities, but not gamma-ray de-excitation data, were incorporated in the modeled sets. In

Table 7. Comparison of modeled and experimental sets of discrete levels in ^{176}Lu , ^{174}Lu , and ^{236}Np .

Level-set designation	Energy range	Number of levels	Number of rotational bands	Range of K quantum Nos.	Range of J quantum Nos.
^{176}Lu					
A	0-620 keV	27	9	0-9	0-12
B	0-770	49	18	0-9	0-12
C	0-1000	109	31	0-9	0-12
D	0-1500	291	64	0-9	0-12
Experiment	0-992	38	7	0-7	0-10
^{174}Lu					
A	0-490 keV	27	10	0-7	0-12
B	0-730	72	24	0-8	0-12
C	0-900	131	36	0-8	0-12
D	0-1040	194	46	0-8	0-12
E	0-1200	264	62	0-9	0-12
F	0-1500	433	82	0-9	0-12
Experiment	0-970	57	10	0-7	0-7
^{236}Np					
A	0-1200 keV	714	92	0-6	0-12
Experiment	0-216	5	3	1-6	1-6

Table 7, the experimental lists for ^{176}Lu and ^{174}Lu are seen to account for about 35% of the total in the first 1000 keV of excitation; up to 1500 keV, the experimental lists are only 13% of the total. For ^{236}Np , experimental data are nearly nonexistent. This illustrates a very desirable property of the model: one can construct detailed level schemes even in the absence of direct experimental observations of the odd-odd nucleus.

Figures 20 and 21 show the results of our calculations for lutetium. We computed the neutron-transmission coefficients employed in these calculations by using a spherical equivalent potential derived from a deformed potential for ^{169}Tm ,⁴ and applying it to the lutetium isotopes. Figure 20 shows the results of the calculation of the m/g ratio arising from the $^{175}\text{Lu}(n,\gamma)^{176\text{m,g}}\text{Lu}$ reaction as a function of the incident neutron energy. The lower curves give the best agreement with an m/g ratio derived from recent experimental work. Beer and Kappeler⁵ measured the $^{175}\text{Lu}(n,\gamma)^{176\text{m}}\text{Lu}$ cross section to be 0.906 b at about 30 keV, while Allen *et al.*⁶ measured it to be 0.958 b. The measured total $^{175}\text{Lu}(n,\gamma)$ cross section at 30 keV ranges from about 1.2 b^{7,8} to 1.4 b,⁹ leading to an experimental m/g ratio between 1.8 and 4.0. For comparison, an earlier calculation of the m/g ratio is shown in Fig. 20. The calculation was based on an older set of discrete-level and gamma-ray branching

information for ^{176}Lu . In this case, ^{176}Lu was described up to an energy of 0.77 MeV by only 37 levels.

Figure 21 shows our calculated m/g ratios for the $^{175}\text{Lu}(n,2n)^{174\text{m,g}}\text{Lu}$ reaction as a function of incident neutron energy. The six curves again represent ratio calculations obtained by using the level sets A-F given in Table 7 for ^{174}Lu . The two lowest curves agree best with the m/g ratio measured by Nethaway at 14.8 MeV.¹⁰ Again, as in the case of the calculation of the $^{175}\text{Lu}(n,\gamma)$ isomer ratio, a few hundred discrete levels are required before the correct m/g ratio can be computed.

Figure 22 shows the calculated isomer ratio that we obtained for the $^{237}\text{Np}(n,2n)^{236\text{m,g}}\text{Np}$ reaction. We computed the neutron-transmission coefficients with the coupled-channel code ECIS,¹¹ using a deformed potential derived for ^{239}Pu by Arthur.¹² Our calculation, which uses 714 levels to describe ^{236}Np up to an energy of 1.2 MeV (set A for ^{236}Np in Table 7), is in excellent agreement with the measured data point of Myers *et al.*¹³ Note that those authors reported the ratio of the long-lived isomer ^{236}Np ($t_{1/2} = 1.2 \times 10^5$ y) to the short-lived isomer ^{236}Np ($t_{1/2} = 22.5$ h), $^{236}\text{Np}(\text{l})/^{236}\text{Np}(\text{s})$, to be 0.35, an average value obtained from neutron spectra produced in several underground thermonuclear tests. We plotted their data point at 14 MeV, the most probable incident neutron

Figure 20. $^{175}\text{Lu}(n,\gamma)^{176}\text{Lu}$ isomer ratio as a function of incident neutron energy. Solid curves show our calculations of the ratio, which were done with the ^{176}Lu level sets A-D given in Table 7. Sets C and D gave essentially the same results, and only one curve is shown. The dashed curve shows an earlier calculation, obtained with an older set of ^{176}Lu levels. The data are derived from Refs. 5-9.

Figure 21. $^{175}\text{Lu}(n,2n)^{174}\text{Lu}$ isomer ratio as a function of incident neutron energy. Solid curves show our calculations of the ratio, which were done with the ^{174}Lu level sets A-F given in Table 7. The measured isomer ratios (data points) are from the work of Nethaway.¹⁰

Figure 22. $^{237}\text{Np}(n,2n)^{236}\text{Np}$ isomer ratio as a function of incident neutron energy. The solid curve shows our calculation, which uses 714 levels for ^{236}Np up to 1.2 MeV of energy. The dashed curve shows a calculation made by Mann in 1977.¹⁴ The data point is from the work of Myers *et al.*¹³

energy. In addition, our level scheme assigns the long-lived ^{236}Np to be the ground state and the 22.5-h ^{236}Np to be the isomer. Included in Fig. 22, for comparison, is a 1977 calculation made by Mann.¹⁴

The results of the three calculations presented here clearly show that hundreds of discrete levels and their gamma-ray branching ratios are required to compute accurate isomer ratios for deformed nuclei. Studies are now under way to investigate whether a broader range of spins is required for these levels in such calculations, and to determine how to use all of this discrete-level information in an appropriate deformed-nucleus level-density formulation.

This work was supported by the LLNL Nuclear Test Program.

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How Accurately Can (n,2n) and (n,3n) Cross Sections Be Calculated for ^{237}Np , ^{238}Pu , and ^{239}Pu ?

M. A. Gardner and D. G. Gardner

We estimated (n,2n) and (n,3n) cross sections for ^{237}Np and $^{238,239}\text{Pu}$, and investigated the problem of how accurately the cross sections can be calculated. Since the fission cross section over most of the neutron energy range is 70 to 95% of the total reaction cross section for these isotopes, we found that uncertainties of $\pm 5\%$ in the fission cross section can lead to uncertainties of 10 to 50% in the calculated (n,2n) and (n,3n) cross sections. Reliable (n,xn) cross-section values can probably be obtained only from experimental measurements.

We estimated the (n,2n) and (n,3n) cross sections for ^{237}Np , ^{238}Pu , and ^{239}Pu from their threshold energy up to 16 MeV, and investigated the problem of just how accurately one can calculate the cross sections. We refer to the cross sections as "estimated" rather than calculated, because we did not include fission competition in computing them. Our method of estimating the various (n,xn) cross sections was based on the following expression:

$$\begin{aligned} \sigma_{n,xn}(\text{estimated}) \\ &= \sigma_{n,xn}(\text{calculated with no fission}) \\ &\quad \times \left(1 - \frac{\sigma_f(\text{evaluated library})}{\sigma_{\text{CN}}(\text{optical model})} \right), \end{aligned}$$

where $\sigma_{n,xn}$ (calculated with no fission) is the cross section for an (n,2n) or (n,3n) reaction calculated with no fission competition, using the Nuclear Chemistry Division's version of the STAPRE statistical model code¹; σ_f (evaluated library) is the fission cross section from the ENDL library (Evaluated Neutron Nuclear Data Library)² or the ENDF/B-V library (Evaluated Nuclear Data File)³; and σ_{CN} (optical model) is the compound-nucleus formation cross section computed from the neutron-transmission coefficients T_ℓ generated by the coupled-channel code ECIS⁴:

$$\sigma_{\text{CN}}(E_n) = \pi \lambda^2 \sum_{\ell} (2\ell + 1) T_\ell(E_n) .$$

In other words, each (n,2n) and (n,3n) cross section, calculated without fission competition, was scaled downward by the fraction of the total compound-nucleus reaction cross section remaining after a fission cross section (obtained from one of two evaluated libraries) was accounted for. Since high excitation energies and broad spin ranges are involved, the lack of fission competition did not significantly perturb the spin distributions and the relative production cross sections of the (n,xn) products.

We computed the neutron-transmission coefficients with the ECIS code, using a deformed potential derived for ^{239}Pu by Arthur.⁵ This one set of transmission coefficients was used in the calculations for all three isotopes. The STAPRE calculations were made with allowance for precompound evaporation and with Gilbert and Cameron level-density parameters that were modified to reproduce known values of D_{obs} , the average level spacing at the neutron separation energy. In the case of the neptunium calculations, we used 90 discrete levels to describe ^{237}Np up to an energy of 1.3 MeV, and 714 discrete levels for ^{236}Np up to 1.2 MeV. Almost all of this level information on neptunium was calculated theoretically, and

more details on the modeling used can be found elsewhere in this report (see "Isomer-Ratio Calculations for Neutron-Induced Reactions on Deformed Nuclei . . ." in this section and "Level-Structure Modeling of Odd-Odd Deformed Nuclei" in Section 6). Similar discrete-level calculations have not been made yet for the plutonium isotopes, so only the experimentally available levels were used. This meant that no more than 12 discrete levels were used for any one of the plutonium isotopes.

Figure 23 shows some of the results that we obtained for ^{239}Pu . The estimated (n,2n) and (n,3n) cross sections were obtained with a scaling factor that used the evaluated ^{239}Pu fission cross section from the ENDF/B-V library. The shaded bands indicate the effect of changing this fission cross section by $\pm 5\%$. For example, at an incident neutron energy of 14 MeV, a 5% decrease in the fission cross section leads to a 55% increase in the (n,2n) cross section! This sensitivity is due to the fact that the ^{239}Pu fission cross section, throughout this energy range, is from 65 to 97% of the total compound-nucleus reaction cross section. Therefore, small uncertainties in the fission cross section can result in (n,2n) and (n,3n) cross-section uncertainties of considerable magnitude. Unfortunately, for ^{239}Pu as well as for most of the other actinides, very little experimental fission cross-section data is available for neutron energies above 6 MeV. Furthermore, among the measured cross-section sets that are available, it is not uncommon to see disagreements of 20% and more. Included in Fig. 23 are the (n,2n) cross-section measurements of Loughheed *et al.*⁶ and a recent (n,2n) cross-section calculation made by Arthur.⁵

Figure 24 shows the excitation function that we estimated for the production of the short-lived ^{236}Np ($t_{1/2} = 22.5$ h) via the ^{237}Np (n,2n) reaction. The scaling factor used here was based on the evaluated ^{237}Np fission cross section from the ENDL library. Again, the shaded bands indicate the effect of changing this fission cross section by $\pm 5\%$. Corresponding changes of 15 to 20% in the (n,2n) cross section are seen over most of the energy range. Also plotted in Fig. 24 are the experimental cross-section values for the production of the short-lived ^{236}Np as measured by Landrum *et al.*⁷ and by Lindeke *et al.*⁸

We observed similar sensitivities to the uncertainty in the fission cross section with the (n,xn) cross-section estimates for ^{238}Pu . Over the entire neutron energy range, the ^{238}Pu fission cross section found in both of the evaluated

libraries was always more than 85% of the total compound-nucleus reaction cross section obtained from optical-model calculations. In fact, the fission cross section from one of the libraries was actually larger than the calculated reaction cross section for neutron energies between 12 and 13 MeV.

We reached the important conclusion from this study that reliable (n,2n) and (n,3n) cross-section values for ^{237}Np and $^{238,239}\text{Pu}$ probably can be obtained only from experimental measurements. Calculated (n,xn) cross sections are extremely sensitive to the uncertainties in the fission cross section, since the fission cross section, for all three of these isotopes, is a significant fraction of the total compound-nucleus reaction cross section between neutron energies of 6 and 16 MeV. Even if one were to use the best possible

Figure 23. Estimated (n,2n) and (n,3n) cross sections of ^{239}Pu as a function of incident neutron energy. Shaded bands indicate the effect of $\pm 5\%$ uncertainties in the ^{239}Pu fission cross section obtained from the ENDF/B-V library. Also shown are the (n,2n) cross-section measurements of Loughheed *et al.*⁶ (data points) and a recent (n,2n) cross-section calculation made by Arthur⁵ (dashed curve).

Figure 24. Estimated cross section of $^{237}\text{Np}(n,2n)^{236}\text{Np}$ ($t_{1/2} = 22.5$ h) as a function of incident neutron energy. Shaded bands indicate the effect of $\pm 5\%$ uncertainties in the ^{237}Np fission cross section obtained from the ENDL library. Data points show the (n,2n) cross-section measurements of Landrum *et al.*⁷ and Lindeke *et al.*⁸

fission model to carry out calculations, it would be difficult to assign uncertainties of less than 5% to the calculated fission cross sections. This region of high neutron energy requires either fission cross-section measurements with very

small associated uncertainties, or accurate measurements of the (n,xn) cross sections themselves.

This research was supported by the LLNL Nuclear Test Program.

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Absolute Determination of the K Through O Conversion Coefficients of the M4 Transition in $^{193\text{m}}\text{Ir}$

R. Gunnink, M. Lindner, R. J. Nagle, and D. H. Sisson

We produced carrier-free, nearly isotopically pure $^{193\text{m}}\text{Ir}$ from thermal-neutron irradiation of ^{192}Os . From studies of the conversion-electron spectrum, the photon spectrum, and the absolute counting rates, we calculated the absolute K, L1, L2, L3, M1, M3, M5, N, and O conversion coefficients for the first time. With possibly two exceptions, these values agree closely with theoretical calculations. The value for the energy of the unconverted gamma ray was found to be 80.22 ± 0.02 keV. The half-life for $^{193\text{m}}\text{Ir}$ determined by absolute electron counting in a proportional counter of 100% counting efficiency ($4\pi\beta$) was found to be 10.53 ± 0.04 d.

Boehm and Marmier¹ produced and identified the nuclide $^{193\text{m}}\text{Ir}$ by second-order capture of thermal neutrons in ^{191}Ir . From L- and M-subshell measurements with an electron spectrometer, they characterized an M4 transition of 80.19 keV and measured a half-life of 11.9 d for the decay. Bisgård and Hanson² pointed out that $^{193\text{m}}\text{Ir}$ was also formed through neutron capture in ^{192}Os and conjectured that the level was populated from an isomeric state in ^{193}Os of 17-min half-life. However, Bäcklin, Berg, and Malmkog³ showed that about 0.25% of the beta-decays of the well-known 31-h ^{193}Os populated the 80-keV level in ^{193}Ir . Finally, Martin, Merkert, and Campbell⁴ measured the L, M, N, and O conversion-electron ratios for $^{193\text{m}}\text{Ir}$. Although they did not publish their values for these ratios, they normalized their ratios to theoretically calculated values and found generally good agreement for an M4 transition of this energy.

All of the foregoing studies were made with magnetic electron spectrometers on sources that had been intensely irradiated with thermal neutrons. Thus, the $^{193\text{m}}\text{Ir}$ was a very small fraction of the source intensity, and only the high instrumental resolution made possible the detection of the conversion electrons; no other characteristics, except for decay rate, were observable.

During the course of our work in radiochemical detector diagnostics we performed counter calibrations of $^{193\text{m}}\text{Ir}$ that required sources of isotopically pure $^{193\text{m}}\text{Ir}$. This study allowed us to observe not only the conversion electrons, but also L x rays, K x rays, and even the low-intensity unconverted gamma transition.

Enriched ^{192}Os (99.4%) was irradiated in the Omega West reactor at Los Alamos National Laboratory to produce a relatively low level of $^{193\text{m}}\text{Ir}$. A pure, carrier-free iridium chemical fraction isolated from the osmium was free of all contaminant radioactivities except for a relatively low level (compared with the $^{193\text{m}}\text{Ir}$) of ^{192}Ir . The source of this contamination is unclear. Its presence necessitated only a minor correction to a few of our measurements.

From the supply of isolated $^{193\text{m}}\text{Ir}$ we prepared sources for several different detectors:

1. Solution-evaporated sources on ultrathin plastic VYNS films were used for $4\pi\beta$ and for Ge(Li) spectral measurements.
2. Samples electroplated on 5-mil titanium foils provided uniform "massless" sources for conversion-electron spectrometry with an evacuated windowless silicon diode and for Ge(Li) spectral measurements.
3. A solution-evaporated source was prepared in a tubular container for well-type Ge(Li) detector measurements.

From these we were able to compare intensities of all the sources and to measure spectra for K x rays, L x rays, the 80-keV unconverted gamma ray, the L, M, N, and O electrons, and the absolute total electron decay rate (from $4\pi\beta$). All spectral analyses were performed with the GRPANL peak-shape-fitting code of Gunnink and Ruhter.⁵

Figures 25 and 26 show, respectively, the L-shell electron spectrum and the M through P electrons. In Fig. 27 we give that portion of the photon spectrum which includes the K x rays and the unconverted 80-keV gamma ray.

We have also analyzed the L x-ray spectrum. It will be described in next year's annual report.

The energy resolution of our silicon detector is obviously much lower than those attained from any of the magnetic spectrometers.¹⁻⁴ However, because of the high purity of the source and the relatively high sample counting rate, there is little doubt as to the energy and intensity of the constituent peaks.⁵

K-shell conversion electrons were not observed in our spectra ($e_{K^-} \approx 4$ keV). However,

the K x rays of Fig. 27 were mainly from atomic transitions in iridium (i.e., ^{193m}Ir K conversion), with only a small correction for osmium and platinum K x rays arising from contaminant ^{192}Ir .

We found the energy of the unconverted gamma ray to be 80.22 ± 0.02 keV. This value does not differ greatly from the value 80.19 keV,⁴ but conversion coefficients are extremely energy-sensitive; a difference of only 0.03 keV would result in a difference of 0.25% in interpolation of a calculated conversion coefficient.

In comparing results with theoretical calculations, several assumptions were made:

- It was assumed that the $4\pi\beta$ proportional counter detected 100% of the ^{193m}Ir decays.
- The true electron subshell intensities were assumed to be proportional to the peak intensities observed in the windowless silicon detector.
- The limited resolution of the silicon detector precluded resolution of the M2 or M4 subshell peaks. For purposes of comparison with calculations, the M2 was divided evenly between the M1 and M3 peaks. The unresolved M4 and

Figure 25. Windowless-silicon-detector spectrum of L conversion electrons from the 80-keV M4 transition in ^{193m}Ir .

M5 peaks were compared with the sum of the calculated M4 and M5 conversions.

• The N and O + P subshells were resolved only from each other, with no subshell resolution.

Table 8 lists the observed conversion coefficients (α) in column 2. Column 1 gives the subshell, while column 4 lists the values interpolated from the tables of Rösel *et al.*⁶ with a cubic spline fit.⁷ The values listed in column 2

Figure 26. Windowless-silicon-detector spectrum of M, N, and O conversion electrons from the 80-keV M4 transition in ^{193m}Ir .

Table 8. Comparison of observed and calculated ^{193m}Ir conversion coefficients.

Subshell	α (obs)	Std dev (%)	α (calc)	$\frac{\alpha \text{ (calc)}}{\alpha \text{ (obs)}}$
K	104	—	103	0.99
L1	2744	3.0	2773	1.01
L2	549	3.3	493	0.90
L3	11,324	3.0	11,842	1.04
M1	1053	3.3	925	0.96
M2			161	
M3			3869	
M4			78	
M5			263	
N	1341	3.2	1400	1.04
O	194	4.2	224	1.16

Figure 27. Ge(Li)-detector spectrum showing unconverted gamma-ray and iridium-region K x rays.

 Table 9. Comparison of conversion-electron intensity ratios.^a

Shell or subshell	This work	Std dev (%)	Boehm and Marmier ¹	Bisgård and Hanson ²	Bäcklin <i>et al.</i> ³	Calc (Rösel <i>et al.</i> ⁶)
L1	54.5	0.46	55	61	54.8	52.7
L2	10.9	1.30	10	<12	<13	9.4
L3	225	0.3 ^b	225	225	225	225
M1 + 1/2 M2	20.9	1.42	—	—	—	19.1
M3 + 1/2 M2	74.4	0.56	—	—	—	75.0
M4 + M5	6.1	3.90	—	—	—	6.5
K ^c	2.08	—	—	—	—	1.96
L	290.4	0.18	—	—	—	287.1
M	101.4	0.56	—	—	—	100.6
N	26.6	1.17	—	—	—	26.6
O + P	3.8	2.89	—	—	—	4.3

^a Normalized to L3 = 225.

^b Std dev of five determinations of fractional L-shell conversion.

^c From K x-ray intensity determination and Auger $\omega = 0.95$.

are the averages of five independent spectral determinations. The standard deviations listed in column 3 were limited to a minimum of 3% because of the systematic uncertainty of 3% associated with the intensity of the unconverted 80-keV gamma ray. As shown in column 5, the overall agreement between the observed and the calculated conversion coefficients appears to be good.

Because of the low intensity of the 80-keV gamma ray, it represents a large systematic uncertainty in all of our measurements. For this reason, a comparison of electron shell and subshell intensities (rather than of the coefficients α) allows use of maximum precision and also of comparison with values reported by previous investigations. In Table 9 we present the intensity ratios for the L and M subshells and for K, L, M, N, and O shells. Column 3 lists the experimental uncertainties of our values. For historical reasons, our values are arbitrarily normalized to a value of 225 for the L3 intensity.⁸

Our observed values for the L subshell conversions are in good agreement with the data of Boehm and Marmier¹ and Bäcklin *et al.*³ Since

no reasonable parameter adjustments in the peak-shape fitting could lead to an appreciable change in the L2 ratio, we conclude that the discrepancy between our observed L2 value and the calculated value of Rösel *et al.* is real. Except for the M1 conversion, there are no further obvious conflicts with theory.

Since our source was nearly pure ^{193m}Ir, we followed the decay of both of our $4\pi\beta$ samples for about six months in order to correct for the minor ¹⁹²Ir component. In Table 10 we compare our value with those of previous investigators.

This research was supported by the LLNL Nuclear Test Program.

Table 10. Comparison of values of ^{193m}Ir half-life.

Reference	Half-life (d)
This work	10.53 (± 0.04)
Bayhurst <i>et al.</i> ⁹	10.6
Bisgård and Hanson ²	10.8
Bäcklin <i>et al.</i> ³	12
Boehm and Marmier ¹	11.9

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Measurement of 14-MeV Fission Yields

D. R. Nethaway and F. F. Momyer

We irradiated ^{235}U , ^{238}U , and ^{239}Pu targets with neutrons of 14 to 15 MeV at the LLNL Rotating Target Neutron Source facility (RTNS-II). These irradiations are part of a program to measure the fission yields of 10.76-y ^{85}Kr , ^{156}Eu , and ^{161}Tb . Accurate knowledge of these yields is necessary for the interpretation of diagnostic measurements made on gas and core samples of debris from nuclear tests. The fission yield of ^{85}Kr had not been measured previously. The yields of ^{156}Eu and ^{161}Tb had been measured before at 14.8 MeV, but we made new measurements because recent advances permitted greater accuracy.

We began a program to measure the yields of several fission products from fission of uranium and plutonium induced by neutrons of 14 to 15 MeV. We plan to repeat the measurements using fission-spectrum neutrons from a critical assembly at the Los Alamos National Laboratory (Los Alamos, NM). Our main interest is in the products ^{85}Kr (10.76 y), ^{156}Eu , and ^{161}Tb . Accurate knowledge of the fission yields of these products is necessary for the interpretation of diagnostic measurements made on gas and core samples of debris from nuclear tests at the Nevada Test Site. ^{85}Kr is one of the peak-yield products used to infer the total number of device fissions. The fission yield of this product does not change significantly with energy in the 14- to 15-MeV range. In contrast, ^{156}Eu and ^{161}Tb are low-yield products whose yields are sensitive to the energy of the neutron that causes fission. Hence, we use them to obtain information on the fission split (both neutron energy and fissioning species) in device tests, by means of a two-group energy approximation (fission spectrum and 14.1 MeV).

The fission yield of ^{85}Kr had not been measured before because of its long half-life. Although the yields of ^{156}Eu and ^{161}Tb had been measured at 14.8 MeV, we made new measurements because recent advances in counting techniques permitted greater accuracy and because knowledge of the fission yields at 14.1 MeV is needed.

Figure 28 shows the relevant formation and decay processes for ^{85}Kr . Since the fission yield

of ^{85}Kr had not been measured before, we had to rely on the measured yield of $^{85\text{m}}\text{Kr}$ and the branching fraction of $^{85\text{m}}\text{Kr}$ to the ground state to estimate the yield of ^{85}Kr . This method is correct if the independent fission yield of ^{85}Kr is negligible. If the independent yield of ^{85}Kr is appreciable, however, the estimated yield for ^{85}Kr would be too low. We have long suspected that the independent yield of ^{85}Kr was the cause of past anomalous diagnostic results. While we would like to understand our results in terms of branching fractions and independent yields, the fact is that direct measurement of ^{85}Kr production in fission tells us what we need to know for diagnostic purposes.

We irradiated four targets at the Rotating Target Neutron Source (RTNS-II) at LLNL: ^{235}U (oralloy) at 14.3 and 14.7 MeV, ^{238}U at 14.4 MeV, and ^{239}Pu (weapons-grade plutonium) at 14.8 MeV. The general method that we used was to dissolve the irradiated target material in a closed system and collect the gaseous products for measurements of krypton and xenon. We repeated the gas collection twice later for the xenon decay products of tellurium and iodine. Aliquots of the target solution were then taken for direct gamma-ray assay and for rare-earth analyses.¹ Table 11 summarizes the results for ^{85}Kr , ^{156}Eu , and ^{161}Tb .

We plan to irradiate two more targets to complete the series: ^{238}U at 14.7 MeV, and ^{239}Pu at 14.3 MeV. We irradiate each target at two energies so that we can measure the change in

^{156}Eu and ^{161}Tb yields in the 14- to 15-MeV range. The results obtained for ^{235}U at 14.3 and 14.7 MeV show a significant increase in yield for ^{156}Eu and ^{161}Tb , and allow us to make reasonable extrapolations to 14.1 MeV.

For 14.8-MeV fission of ^{239}Pu , we measured a yield of 0.00232 atoms/fission for ^{85}Kr , 18% higher than our historical yield, which is based on the decay of $^{85\text{m}}\text{Kr}$ and assumes no independent formation of ^{85}Kr . If the 0.204 decay branch of $^{85\text{m}}\text{Kr}$ is correct, this is equivalent to an independent fractional chain yield of 0.046 for ^{85}Kr , quite consistent with the value of about 0.041 predicted for $^{85\text{m}}\text{Kr} + ^{85}\text{Kr}$.² Similarly, for 14.3- and 14.7-MeV fission of ^{235}U , we measured

a yield of 0.00400 atoms/fission for ^{85}Kr , 4.5% higher than our historical yield. This is equivalent to an independent fractional chain yield of 0.012 for ^{85}Kr , again consistent with the value of about 0.014 predicted for $^{85\text{m}}\text{Kr} + ^{85}\text{Kr}$.²

We also measured the yields of several xenon isotopes, including the independent yields of ^{127}Xe and $^{129\text{m}}\text{Xe}$. From the gamma-ray assay of the solution samples, we were able to determine the yields of dozens of other products, including the nuclides ^{95}Zr , ^{99}Mo , ^{144}Ce , and ^{147}Nd , which we used to calculate the number of fissions in the sample.

This work was supported by the LLNL Nuclear Test Program.

Figure 28. Formation and decay processes for ^{85}Kr . The predicted fission split is shown for 14.8-MeV fission of ^{239}Pu to ^{85}Br and $^{85\text{m}}\text{Kr} + ^{85}\text{Kr}$.

Table 11. Measured fission yields of ^{85}Kr , ^{156}Eu , and ^{161}Tb . (Statistical errors are about 1%, and do not include decay-scheme uncertainties or the error in the number of fissions.)

Target	Neutron energy (MeV)	Fission yield (atoms/fission)		
		^{85}Kr	^{156}Eu	^{161}Tb
^{235}U	14.3 ₃	3.98×10^{-3}	5.14×10^{-4}	4.09×10^{-5}
	14.7 ₃	4.00×10^{-3}	5.35×10^{-4}	4.38×10^{-5}
^{238}U	14.4	2.50×10^{-3}	1.04×10^{-3}	6.89×10^{-5}
^{239}Pu	14.8	2.32×10^{-3}	1.94×10^{-3}	2.29×10^{-4}

¹ We gratefully acknowledge the help of J. Fontanilla, W. Culham, C. Smith, and N. Bonner with the rare-gas separations, J. Landrum and H. Hicks with the chemical separations, and J. Andrews and E. Delucchi with the rare-earth separations.

² D. R. Nethaway, *Tables of Values of Z_p , the Most Probable Charge in Fission*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-51640 (1974).

Fission Yields of ^{161}Tb from Fission-Spectrum Neutrons on Plutonium

H. G. Hicks

Measurements of the yields of a number of fission products from 14-MeV neutrons on ^{235}U , ^{238}U , and $^{239,240}\text{Pu}$ led to an improved value of the ^{161}Tb fission yield from fission-spectrum neutrons on plutonium. Data from six recent underground nuclear events were used to determine the value, $(6.62 \pm 0.10) \times 10^{-3}\%$.

Samples of underground nuclear detonation debris are dissolved and analyzed for ^{95}Zr , ^{144}Ce , and ^{147}Nd to determine the number of fissions represented by one millilitre of the solution. The conversion of ^{95}Zr , ^{144}Ce and ^{147}Nd atoms per millilitre to fissions per millilitre requires the knowledge of the fission yields of these nuclides. In analyzing a nuclear detonation, the fission-product-yield distribution is described by a linear combination of the fission-yield distributions caused by fission-spectrum (f.s.) neutrons and 14-MeV neutrons on each fissioning nucleus. The fraction of fissions caused by 14-MeV neutrons is determined from measurements of a nuclide, such as ^{161}Tb . The fission yield of such a nuclide is greatly different in the two fission-product-yield distributions. A much more accurate measurement of the fraction of 14-MeV-neutron fission of plutonium ($\pm 10\%$) is provided by the independent fission yield of $^{131\text{m}}\text{Xe}$, $1.07 \times 10^{-3}\%$ with f.s. neutrons and 0.0496% with 14-MeV neutrons. The fission of ^{235}U produces a small correction because of the low fission yields of $^{131\text{m}}\text{Xe}$ ($3.1 \times 10^{-5}\%$ with f.s. neutrons and $6.70 \times 10^{-3}\%$ with 14-MeV neutrons), and the fission yields of $^{131\text{m}}\text{Xe}$ from ^{238}U are at most 1% of those from ^{235}U .

Momyer and Nethaway recently reported measurements of the yields of fission products from 14-MeV neutrons on ^{235}U , ^{238}U , and $^{239,240}\text{Pu}$ (see "Measurement of 14-MeV Fission Yields" in this section). Included in these measurements are the values for the independent yield of $^{131\text{m}}\text{Xe}$ and the total chain yields of ^{147}Nd and ^{161}Tb . These data enabled us to use data from underground nuclear events to

improve the value of the fission yield of ^{161}Tb from f.s. neutrons on plutonium.

Six underground nuclear events that had the following three criteria were selected:

- $^{131\text{m}}\text{Xe}$ data from prompt gas samples.
- ^{147}Nd , ^{160}Tb , and ^{161}Tb data.
- Large fraction of fissions due to plutonium.

The ^{161}Tb data must be corrected for that grown in from the 3.7-min ^{161}Gd formed from neutron capture by ^{160}Gd in the surrounding soil. For each sample, 1.6% of the ^{160}Tb atoms/ml was subtracted from the ^{161}Tb atoms/ml to account for this background,¹ at most a 3% correction. The contribution of ^{160}Tb from fission was considered to be negligible. The fractions of fissions in plutonium by 14-MeV neutrons were obtained from the interpretation of the $^{131\text{m}}\text{Xe}$ data by C. F. Smith.

The fission yield of ^{161}Tb by f.s. neutrons, f_1 , was calculated from

$$\frac{{}^{161}\text{Tb}}{{}^{147}\text{Nd}} = \frac{(1-x)f_1 + xf_2 + u_1}{(1-x)f_3 + xf_4 + u_2} \quad (1)$$

where

- x = fraction of 14-MeV neutron fission;
- f_1 = fission yield of ^{161}Tb , Pu + f.s. neutrons;
- f_2 = fission yield of ^{161}Tb , Pu + 14-MeV neutrons;
- f_3 = fission yield of ^{147}Nd , Pu + f.s. neutrons;
- f_4 = fission yield of ^{147}Nd , Pu + 14-MeV neutrons;

u_1, u_2 = contribution to ^{161}Tb and ^{147}Nd production, respectively, from uranium fissions.

The fission yields for f.s. neutrons were obtained from Ref. 2 except for f_1 . The fission yields for 14-MeV neutrons were obtained from the data reported in "Measurement of 14-MeV Fission Yields." The results, summarized in Table 12, give an average value of $(6.62 \pm 0.10) \times 10^{-3}\%$ for the fission yield of ^{161}Tb from f.s. neutrons on plutonium.

This research was supported by the LLNL Nuclear Test Program.

Table 12. Fission yield of ^{161}Tb from fission-spectrum neutrons on plutonium.

Event	^{161}Tb fission yield (%)
Burzet	6.99×10^{-3}
Tilci	6.34×10^{-3}
Caboc	6.86×10^{-3}
Kesti	6.50×10^{-3}
Manteca	6.60×10^{-3}
Crowdie	6.42×10^{-3}
Average	$(6.62 \pm 0.10) \times 10^{-3}$

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In Situ Measurement of Carbon in Potential Shot-Site Holes

H. I. West, Jr., J. E. Glasgow,¹ and J. R. Hearst²

Knowledge of the carbonate content of proposed nuclear test sites is important in evaluating the sites for containment. We are using oil-well logging techniques based on the detection of ^{12}C and ^{16}O gamma rays from inelastically scattered 14-MeV neutrons. We expect to achieve an accuracy of ± 0.01 weight fraction for the CO_2 determination.

To evaluate a potential nuclear test site for containment, one needs to know the carbonate content of the surrounding medium. Containment scientists rely on chemical assays (reported as CO_2) of bore-hole cuttings and side-wall samples. Obtaining and assaying such

samples is a slow, expensive process. A laboratory experiment³ gave evidence that the detection of 4.43-MeV gamma rays following the inelastic scattering of 14-MeV neutrons from ^{12}C could provide an accurate measure of the carbon content. In this technique, 6.13-MeV gamma rays

Downhole tool

Timing

Figure 29. Schematic of the logging tool. At the top are the tool and associated electronics. The bottom shows the timing used in data acquisition.

are also measured from ^{16}O following inelastic neutron scattering, and the results are reported as a carbon-oxygen weight ratio (C/O). Since the medium is almost universally 50% oxygen, such normalization eliminates the need for absolute measurements. Containment scientists require an accuracy of ± 0.01 weight fraction of CO_2 (equivalent to ± 0.006 for C/O).

The approach has been used in the oil-well logging business (for example, see Refs. 4 and 5) for a number of years. Logging tools have been developed that approach the required accuracy but which are calibrated to work only in water-filled holes 8 to 12 in. in diameter; they are not expected to work well in our dry holes, which are usually 5 to 10 ft in diameter. The experienced logging companies have not shown much interest in modifying their logging tools to meet our needs.

Our major problem is the efficient detection of the ^{12}C gamma rays in the presence of other radiations. Unfortunately, the kinematics of the $^{12}\text{C}(n,n'\gamma)$ reaction result in Doppler-broadened gamma-ray line widths of more than 50 keV; hence the high resolution of the germanium gamma-ray detector cannot be put to use. Low detection efficiency limits the usefulness of germanium also. We therefore chose the NaI(Tl) scintillation detector. Since we are not limited by size, we can use large-volume detectors to enhance detection into the full-energy peak and thus improve sensitivity. We are working with crystals 5 in. in diameter by 5 in. thick and 6 in. in diameter by 6 in. thick, and there is evidence that even larger crystals would be worthwhile.

Figure 29 shows a plan of the future system. Our present system differs from it only in two ways: signals come directly from the detector preamplifier to the pulse-height analyzer in the trailer, and the small computers are not in use.

In our system, the neutron generator is used in the pulsed mode at a repetition rate of 10 kHz. We normally collect data in the periods 0–15, 15–30, and 30–100 μs . The neutron burst, lasting about 10 μs , appears in the first period; spectra are acquired in all three periods. Data from the third period are due mostly to neutron capture and are used to correct the inelastic data acquired in the first period. Most of the inelastic gamma rays observed are due to carbon, oxygen, and silicon. The capture spectrum comes from a wide variety of elements, but fortunately this contribution can be measured accurately for subtraction from the first period.

Our work to date has been with boxes of calibrated materials, graphite, and various mixtures of limestone and sand. Figures 30 and 31 show spectra for graphite and limestone, respectively. We had some success in measuring the carbon contribution in the various samples. Figure 32 shows the intensity of the carbon signal as a function of the carbon content of the sample, determined by simple unfolding procedures. The signal for graphite had to be corrected for differences in self-shielding for neutrons and gamma rays with respect to limestone-sand mixtures. The abscissa is given in grams of carbon per cm^3 but, since the density of the medium is about 2 g/cm^3 , the data are approximately C/O as they stand. The output should be directly proportional to the input, so we expect the results to fall on the 45° line, as shown. At the lower values there are marked deviations from expectation; the bottom point can be interpreted as $\text{C/O} = 0.05 \pm 0.02$. We need results about four times more accurate. To achieve greater accuracy, improvements are required in the detection system and in data analysis. Accurate detector-response functions must be determined before more sophisticated computer codes can be used to analyze the data. We plan to make extensive use of the GAMANAL⁶ code in future data analysis.

Our immediate goals are to achieve the required sensitivity, at least in the laboratory,

Figure 30. Spectrum obtained from the inelastic scattering of 14-MeV neutrons off ^{12}C in the form of graphite.

and to further define the system. We anticipate that it will take 2 years to produce a fieldable instrument that has the accuracy needed. Work on calibration and on improvement in data

Figure 31. Spectrum obtained from the inelastic scattering of 14-MeV neutrons off ^{12}C in granulated limestone (CaCO_3).

analysis will extend beyond that time.

This work was supported by the LLNL Containment Program.

Figure 32. Response of the logging tool in measurements of the ^{12}C content of calibrated samples. The abscissa is approximately equal to C/O .

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Geochemistry and Environmental Sciences

4

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Overview

Because the Laboratory handles large quantities of special nuclear materials, conducts underground tests at the Nevada Test Site, and, in past years, conducted atmospheric tests in Nevada and the Pacific Ocean, it must determine the possible sources of environmental contamination and understand the mechanisms for the distribution and biological uptake of radioactive species. Much of this effort involves careful and routine sampling in areas that have a history of contamination or the potential for contamination by radioactive nuclides. Whenever possible, we use these data to draw more far-reaching conclusions about the behavior of radionuclides in the environment.

In this section, we report on work that utilizes data on tritium levels in rain, soil moisture, groundwater, and vegetation to characterize the pathways and inventories of tritium in the local hydrologic cycle. This year we found tantalizing empirical evidence that tritium may not be a reliable tracer of groundwater movement.

We also report progress on the study of atoll islands in the Pacific Ocean. Through the use of numerical models we conclude that seismic profiling and surface resistivity measurements are sufficient for resource assessment and island groundwater research. Additionally, we gained insights into atoll geochemistry, geology, and biology by studying pore-water chemistry and motion within an Australian coral reef. Such studies yield insights into both reef and atoll geohydrology that in turn aid in modeling the natural leaching of radionuclides from the soils of atoll islands contaminated by nuclear-device tests.

Our experience in determining low levels of radioactivity in the environment and measuring biological uptake offers the opportunity to develop new techniques for monitoring

radionuclide concentration levels and detecting undocumented releases of radioactivity in the southern hemisphere. In this region, much of the radioactivity entering the atmosphere is deposited in the ocean rather than on land. We determined that marine plankton, which are abundant and ubiquitous, can concentrate many radionuclides to the point where they can be readily detected. This year, we report on the progress toward establishing a data base of the background inventories of fission-product radionuclides at various locations in the South Pacific. These data will be necessary to interpret results if we monitor the southern hemisphere for changes in the levels of marine radionuclide concentrations.

The Division is also involved in developing techniques to store nuclear wastes safely. We developed the methodology for dynamic laboratory leaching studies to test the stability of nuclear waste forms and studied both glass beads and simulated uranium fuel rods. From these experiments, we determined quantitative values for the leach rates of relevant radioactive species.

The integrity of nuclear-waste repositories depends not only on the matrix containing the radionuclides but also on the integrity of the packaging. Therefore, we are studying the effects of radiation on the corrosion of metal canisters in relevant geochemical environments and in high-radiation fields.

We also studied the containment properties of geologic media at the Nevada Test Site, by laboratory leaching studies of melt glass and through radiometric analyses of groundwater taken from wells adjacent to test cavities. We conclude that leaching from vitrified cavity material should not be a contributor to groundwater contamination.

Livermore Valley Tritium-Distribution Study

M. R. Ruggieri

We are investigating tritium distributions in the Livermore Valley of California in order to better understand flow pathways within the local hydrologic cycle. Soil-water analyses utilizing two different extraction methods have identified a disequilibrium within the soil water. A conceptual model is being developed which describes this phenomenon and has important relevance to predicting and understanding the movement of other mobile radionuclides in porous media.

The Livermore Valley ^3H -distribution study is investigating the local hydrologic cycle by utilizing Laboratory tritium effluent as a water tracer. Data gathered on tritiated water (HTO) levels in rain, soil moisture, groundwater, and vegetation allow us to characterize the flow pathways and inventories of tritium within the environs and to make inferences about water-flow mechanisms and the potential transport of mobile radionuclides. A special study within this effort involves the movement of ^3H in an irrigated-vineyard environment.

Previous reports¹ described tritium distributions following a minor HTO release in

December 1981. Results indicated that tritium-labelled precipitation occurred up to 10 mi away and that several months of uncontaminated rainfall were required to wash the HTO out of the root zone of most natural grass vegetation (a depth of 0-5 ft).

In order to further investigate the fate of the HTO pulse, additional samples collected in the spring of 1982 were analyzed. Results from these analyses show that by the late spring of 1982 the HTO pulse was not detected in the soil column. Interpretations of these results along with neutron-probe soil moisture data (see Fig. 33) indicate that the pulse was diluted by flow dispersion as it moved downward and eventually became indistinguishable from the soil-water background.

Of particular interest is the difference in tritium values observed between suction lysimeter samples and soil-core moisture samples extracted using a freeze-drying technique (see Table 13). The results show that the ^3H in soil water extracted with suction lysimeters is significantly different than in the freeze-dried core water. Inspection of average LLNL tritium levels in rain and lysimeter soil water prior to the 1981 release indicates that the 36- to 84-in. soil-core values represent "normal" Lab rainwater, while the lysimeter water values at these depths are elevated to levels similar to those observed during the release. This suggests that a soil-water disequilibrium exists in the soil column and that different moisture compartments are differentiated by the two soil-

Table 13. Analysis of tritium in soil water.

Depth (in.)	Method	
	Lysimeter samples ^a (tritium units) ^c	Soil cores, freeze-dried ^b (tritium units)
6	865 ± 52	—
12	705 ± 47	846 ± 51
36	704 ± 47	371 ± 34
60	757 ± 49	—
84	—	403 ± 35
132	—	349 ± 33
180	—	128 ± 20

^a Collected 4-16-82.

^b Collected 5-19-82.

^c 1 tritium unit = 3.23×10^{-3} pCi/ml.

water-extraction methods. Additional sample analysis will be carried out to expand and refine this phenomenological model.

Results from a corollary study at a local irrigated vineyard also show evidence of a soil-water disequilibrium. As noted in previous reports, the source and uptake of tritium into grape plants has been under investigation. Through extensive analysis of soil, plant, and water samples the input source of tritium into the grape plant has been determined to be winter rain. The comprehensive instrumentation at the experimental site (piezometer with recorder, lysimeters, and soil-moisture monitoring equipment) has provided us with information to develop a useful model of the unsaturated zone in which the grape plants are rooted. Saturated-groundwater surface monitoring has shown that the water table at this location routinely rises within 15 ft of the soil surface during the winter (it has been observed as shallow as 12 ft after heavy rains). This annual groundwater movement probably dilutes and removes elevated tritium levels in soil below 15 ft (see Fig. 34). Results from the analysis of soil-core samples collected throughout the first 30 ft of depth concur with this observed groundwater movement and theorized dilution mechanism. These soil-core

results also indicate that soil water held in shallow soil (0–15 ft deep) is high enough in tritium to account for the tritium levels found in grape plant parts (see Table 14). Comparison of very shallow lysimeter samples and soil cores suggests that soil-water compartments are not in equilibrium (see Table 15). The presence of soil water that has winter-rainfall tritium values well into the summer irrigation period indicates that low-tritium irrigation water is not displacing or equilibrating with residual winter rain. Although the soil-water conditions at the irrigated vineyard differ greatly from those at the LLNL site, we observe similar examples of soil-water disequilibria. This conceptual model involving

Table 14. Analysis of tritium in vineyard soil cores.

Depth (ft)	Tritium units
5	195 ± 12
10	210 ± 12
15	105 ± 9
20	88.3 ± 8
25	63.4 ± 7
30	60.5 ± 7

Figure 33. Amount of water at 5- and 15-ft depths on the LLNL site.

soil-water compartments has relevance to all porous-media flow phenomena, but especially to those within the unsaturated zone. Movement of mobile radionuclides through unsaturated porous media may be strongly influenced by water

partitioning. In an effort to further elucidate and refine the current model, additional sample analysis will be carried out.

This work was supported by the LLNL Site Environmental Monitoring Program.

Figure 34 Rise of the water table with rainfall (cumulative), as indicated by the depth of a local vineyard well (ground surface to water level)

Table 15. Analysis of tritium in very shallow samples from the vineyard.

Collection date	Irrigation water	Lysimeter samples from 1-ft depth (tritium units)	Soil cores from 6-in. depth, freeze-dried (tritium units)
6-23	—	—	171 ± 34
7-7	32 ± 2.6	—	223 ± 36
7-24	—	31 ± 6.4	117 ± 7.4
7-26	66 ± 8.6	33 ± 7.2	—
7-29	—	59 ± 6.8	—
8-20	38 ± 6.8	—	—

1 M R Ruggieri Livermore Valley Tritium Distribution Study Nuclear Chemistry Division Annual Report FY82 Lawrence Livermore National Laboratory Livermore CA UCAR 10062 82/1 (1982)

Marshall Islands Hydrogeology

R. W. Buddemeier, M. Herman,¹ and S. W. Wheatcraft¹

Trial calculations with a numerical model indicate that our conceptual model of a layered aquifer system represents a useful approach to understanding the hydrology of atoll islands. We are now making detailed calculations to explore the interactions of the different variables and to determine best-fit hydraulic conductivity values on the basis of field data. A review of the relationship between large-scale permeability and carbonate cementation has led us to propose that island groundwater research and resource assessment can be effectively conducted with a combination of surface resistivity measurements, surface seismic profiling, and shallow-water-table studies in locations where drilling programs are not feasible.

Our previous report² mentioned the development of a numerical model of a vertical two-dimensional section of an atoll island characterized by a two-layer aquifer system. This work has proceeded along two parallel and complementary pathways: (1) development of a

model of an idealized island (symmetric and dimensionally convenient) for study of the interaction of variables, and (2) construction of a dimensionally and physically accurate model of Enjebi Island (Enewetak Atoll) to attempt to reproduce by model calculations the extensive set of field hydrologic data available for that site.^{3,4} The two-layer model describes a high-permeability Pleistocene limestone overlain by unconsolidated Holocene carbonates of lower permeability, and preliminary calculations indicate that application of this model modifies water-table-head and tidal-response patterns calculated from the conventional Ghyben-Herzberg model³ in a manner consistent with the field observations. Figure 35 shows a comparison of the field data with the results of the two different model calculations. Present work is directed toward optimizing the data fit by varying the ratio of permeabilities of the upper and lower aquifers. Besides validating the model, this exercise will provide an indirect experimental determination of the lower-aquifer permeability.

In addition to the continuation of the modeling efforts, further consideration of the surface resistivity studies² in conjunction with the hydrologic and geophysical data previously reported^{3,4} has led to a proposal for a groundwater-resource evaluation and research technique suitable for application to remote atoll and reef islands.⁵ Since the hydrologically

Figure 35. Comparison of calculated and measured water-table tidal responses. Circles indicate data from Enjebi Island; curves show calculations based on the Ghyben-Herzberg model³; shaded zones show the range of values calculated from the two-layer model using input values appropriate to Enjebi Island.

significant boundary between the Pleistocene and Holocene aquifers corresponds to a change in degree of lithification, surface seismic profiling can map the interface. Surface resistivity studies have been shown useful in estimating total fresh water and the vertical distribution of salinity, and tidal response, water quality, and head measurements on shallow wells can be used in conjunction with the two-aquifer model to estimate the overall hydrologic characteristics of

the system. Since these field measurements use portable and relatively inexpensive equipment, their combined application will permit hydrologic characterization of islands where the more conventional approach of core drilling and hydrologic testing is either economically or logistically impractical.

This research was funded by the LLNL Nuclear Chemistry Research Program.

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Reef Hydrogeology and Geochemistry

R. W. Buddemeier, J. A. Oberdorfer,¹ D. W. Kinsey,² and J. C. Andrews²

Studies of pore-water chemistry and water motion within an Australian coral reef showed that water residence times are of the order of months to a few years, that biological activity maintains a reducing environment in spite of advection of dissolved oxygen into the reef fabric, and that both carbonate precipitation and solution are occurring in response to biogenic CO₂ inputs to the pore water. The results have important implications for marine geochemistry, geology, and biology.

Because of questions raised as a result of studies of atoll island hydrology (see "Marshall Islands Hydrogeology" in this report) and the general lack of information about physical and chemical processes in the interior of coral reef systems, a collaborative study of Davies Reef (central Great Barrier Reef, Australia) was undertaken during January–April 1983.

The reef was instrumented with tide gauges and current meters to obtain data on water levels, current velocities, and wave spectra. Seventeen test wells were drilled and cased in the reef, which is submerged at all tide stages. Three pairs of wells were established in a cross-reef transect which was used primarily for geochemical sampling. Eleven wells were emplaced in two concentric circles for dye-transport studies and field permeability measurements (slug tests). Figure 36 shows the layout of the study area and the transect well depths.

Reef permeabilities were measured both by slug-testing of the shallow wells and by laboratory permeametry of hand samples and material cored from a deep drilling site near the study transect. Unconsolidated samples and formations yielded permeabilities of 100–2000 m/d, while consolidated materials (including the surface "reef plate," which acts as a barrier between the overlying water and the shallow, unconsolidated sediments) generally had permeabilities in the range of 0.1–10 m/d. Preliminary data from the tide and current meters suggest that sustained cross-reef water level differences may be 5–10 cm as a result of wind and current effects, and that transient

heads of 25–30 cm may occur at periods of high or low tide. These head data can be combined with formation permeabilities to estimate pore-water velocities and residence times in the reef structure. Some of these estimates are presented in Table 16.

Also given in Table 16 are velocity and residence-time estimates based on dye-transport studies and on variations in oxygen content observed over the tidal cycle. Dye studies were performed by injecting dye into one well and monitoring adjacent wells for appearance. Oxygen-renewal times were calculated from the

Table 16. Pore-water velocity and residence-time estimates.

Method	Velocity (m/d)	Residence time (d)
Calculated		
Horizontal	0.4	1000
Vertical	7	4
Dye transport		
Horizontal	400	1
	<8	40
	<54	6
Vertical	9	3
	≥1.4	20
	≥2.8	10
O₂ renewal		
1–3%/tide = 2–6%/d	–	20–50

Figure 36. The Davies Reef study area, showing locations of the instrumentation and the test wells. (The two letters that identify each well stand for: F = forward, M = middle, B = back, D = deep, and S = shallow. MT was a surface sample. The well depths are shown in parentheses.)

percent of saturated surface water required to produce the observed tidal cycle variations in pore-water dissolved oxygen. Although many of the techniques provide limits or estimates only, and the system under study is quite heterogeneous, it is clear that shallow pore waters have residence times of the general magnitude of months. This is a hydrologically dynamic system rather than the solid and impermeable barrier it is often considered to be.

Intensive chemical sampling and *in situ* on-shipboard analysis over several tidal cycles revealed a number of interesting features. All of the wells contained sulfide and had low but measurable levels of dissolved oxygen. Relative to the surface seawater, all well waters showed increased total CO_2 (indicating oxidation of

Figure 37. A comparison of the relationship between alkalinity and ΣCO_2 for supersaturated surface seawater and saturated pore-water samples.

organic matter, probably by sulfate-reducing bacteria). However, both positive (carbonate solution) and negative (carbonate precipitation) alkalinity changes were discovered, and all of the pore-water samples were at saturation equilibrium for aragonite (unlike the surface water). Figure 37 shows these features, as well as the unexpectedly strong correlation between alkalinity and ΣCO_2 .

These results suggest that the organic carbon supply (either endogenous or advected) is adequate to maintain anaerobic conditions in spite of continuous oxygen injection, and that the supersaturated seawater goes to CaCO_3 equilibrium rather quickly after entering the reef. Thereafter, the history of precipitation and solution depends on the amount of biogenic CO_2

released into the system, and probably therefore on its relative "age" and trajectory through the biologically active reef framework.

These results have raised a number of basic questions about the budgets of organic and inorganic carbon within the reef structure, and further analysis of the data is expected to have important implications for subjects as diverse as the mechanisms of carbonate diagenesis and the coupling between the benthic ecosystem and its substrate.

This work was supported by the LLNL Nuclear Chemistry Research Program.

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Marine Radioactivity in the Southern Hemisphere

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The Southern Hemisphere Aquatic Monitoring Project is exploring the potential utility of analyzing various marine biota to detect and identify radionuclides released to the marine environment of the southern hemisphere. The project is in its second year; we completed a literature search for relevant data and became aware of current trends in recent research. A total of 50 plankton samples have been collected from the southern hemisphere, and 38 of them have been analyzed so far. We have found the samples easy to collect in a reasonable time with simple equipment, and capable of providing high sensitivity for detection of radionuclides. Radionuclides in the natural decay chains of uranium and thorium were in the ranges expected and do not affect our sensitivity for artificial radionuclides. ^{95}Nb and ^{144}Ce were found in very low concentrations in eight of the plankton samples as well as in associated air filters and fallout collections. The source of these radionuclides has not been determined.

The Southern Hemisphere Aquatic Monitoring Project is exploring the potential utility of the analysis of various marine biota to detect and identify releases of radioactivity occurring in the southern hemisphere. In the event of an atmospheric release, much of the radioactivity entering the troposphere would be deposited very quickly on the surface of the ocean, leaving a minimal amount to be detected by atmospheric sampling. There is also the possibility of a release directly to the marine environment, which could produce a relatively large volume of radioactive water but little, if anything, detectable by atmospheric sampling. In the ocean, dilution and dispersion would make the radioactivity progressively more difficult to detect if it were not for the fact that various marine organisms, especially the plankton, concentrate the radioactive isotopes of many elements in their bodies. The concentration factors relative to water are often 1000-fold or greater, and the plankton are relatively abundant and easy to collect and analyze. Some of the plankton-associated organisms, as well as some

of the larger marine algae, have similar characteristics.

This project, now in its second year, originally consisted of four stages: (1) a literature search to ascertain what relevant work had been done in the field of bioconcentration and the direction of current research; (2) collection of background samples from the southern hemisphere, both to gather data and to develop experience; (3) development of a capability for a fast sampling response in the case of a release; and (4) development of a continuous-monitoring capability. The literature search is completed, at least as far as obtaining reports relevant to our work, but the large task of digesting this information will require more time. During the period of November 1981 to May 1982, we collected 32 samples on board the USCGC *Glacier* from the stations shown in Fig. 38, and we reported the results of the analysis of eleven of them in last year's annual report. Emphasis on the third and fourth stages, development of fast response and continuous-monitoring capabilities, has decreased over the last year. While both

Figure 38. Sampling locations where plankton samples were successfully collected. Fission products were detected at the circled stations.

these areas continue to receive some attention, we feel that monitoring and sample collection are best left to experts in the field of surveillance, so we are concentrating more on development of new types of samples and techniques.

This year's field work was similar to last year's. We again used the USCGC *Glacier* which followed a similar cruise track except that this year, instead of returning up the west coast of South America, the ship retraced its route south. We again worked jointly with Battelle Pacific Northwest Laboratory personnel (Richland, WA), who took air filter and wet and dry fallout samples. We collected only 18 plankton samples this year, but they were generally from latitudes greater than 60°S where we collected relatively few last year. The samples were also of generally better quality because they were collected by net tow, rather than pumped through the ship's fire mains and filtered. Collection by net tow is also much faster; a typical 2-hour tow yields a sample equivalent to 24 hours of pumping. All of these

samples have been counted on our standard Ge(Li) systems to detect the shorter-lived nuclides such as ^7Be , and will also be counted on our high-sensitivity Compton-suppressed spectrometer as time permits. Subsamples were also sent to our project biological advisor, Prof. John Wormuth of Texas A&M University, for species analysis. We report no data from this year's samples, since we concentrated on the completion of last year's analyses and the literature work.

All 32 of last year's samples have been counted on our low-level spectrometer. It is very interesting to note that the fission products ^{95}Nb and ^{144}Ce were detected from eight locations. The concentrations for both nuclides were very low but, in general, significantly above detection limits. These two nuclides were also detected in the atmospheric samples from these same stations, but it is not yet clear whether this is indicative of atmospheric input or sea-surface resuspension. Both of these nuclides are common components of fission waste, have high

concentration factors in plankton, and have been reported previously from other localities. In fact, after the Pacific nuclear test series in 1956, ^{95}Zr – ^{95}Nb and ^{144}Ce – ^{144}Pr were the predominant radionuclides detected in plankton in the Marshall Islands.¹

Concentrations of the natural radionuclides of the uranium and thorium series were all within the ranges expected and did not affect the detection sensitivity for artificial radionuclides. At present we cannot say much about the ^7Be in plankton except that it, too, is highly correlated with the air and fallout samples. Upper limit values for the nuclides ^{54}Mn , ^{65}Zn , and ^{103}Ru were very low, making the positive detection of these nuclides good evidence for a recent nuclear event of some kind. With the upper limit values as determined and assuming an average concentration factor of a few hundred, we have a sensitivity for detection equivalent to a seawater concentration of about 0.1 pCi/l.

Our experience of the last 2 years has shown that:

1. Adequate samples can be collected in a few hours to a day with simple and inexpensive equipment.
2. Sensitivity is high.
3. Naturally occurring radionuclides do not compromise the sensitivity of detection of anthropogenic radionuclides by direct gamma-ray spectrometry.

4. Concentrations of artificial radionuclides are very low, but not zero.

This last is an important finding which indicates that when searching for a release, a few samples containing common fission products at very low levels would not constitute sufficient evidence that the contaminated area had been located.

In the future, we would like to collect additional plankton samples from other areas of the southern ocean to see if our results so far are typical. Techniques and equipment, though basically simple, could use some refinement, especially as applied to sampling platforms other than oceanographic ships. We would like to analyze selected samples from both cruises for plutonium, americium, and possibly ^{106}Ru and ^{55}Fe , all of which require dissolution of the sample and subsequent radiochemical purification. Other bioaccumulators can be investigated, such as plankton-associated organisms (lantern fish and jelly fish), marine algae from various coastal zones (especially western Australia), and freshwater organisms that are known to have large concentration factors for trace elements. We shall publish one or two reports on the Southern Hemisphere Project during FY84.

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Nuclear-Waste-Form Leaching Studies

F Bazan

Leaching studies are the ultimate criteria for predicting the long-term chemical stability of a nuclear waste form. The first approach to understanding how a nuclear waste form reacts with different types of repository groundwater and rock media is laboratory simulation of interactions between waste forms and rock; this was the objective of the Bead Leach II experiment. We used PNL 76-68 glass beads and simulated uranium fuel rods as the waste forms in a single-pass continuous-flow leaching experiment, with simulated basalt groundwater and simulated brine-sandstone groundwater as the leachants and crushed basalt and crushed sandstone as the rock media.

Since 1979 the Nuclear Chemistry Division has been conducting studies on the leaching of nuclear waste forms as part of an overall effort to find a suitable form in which to store high level nuclear defense waste. These studies have been laboratory experiments, many of which simulate actual repository conditions. The first in these series of experiments was Bead Leach I.¹ A second experiment, Bead Leach II, was begun in 1981 and work continues to date. Both of these experiments were long-term (400-d) single-pass continuous flow leaching experiments. Whereas Bead Leach I was confined to the leaching of 76-68 glass beads with water and bicarbonate leachants, the Bead Leach II experiment used a simulated basalt groundwater and a simulated brine-sandstone groundwater. Furthermore, the Bead Leach II experiment was designed to test

the chemical durability of the glass beads in two separate rock media. The beads were assembled in leaching cells, some of which contained crushed basalt and crushed sandstone in addition to the glass beads. This arrangement is depicted in Fig. 39. Other experimental conditions were temperature = 25°C and 75°C, flow rate = 1 ml/d, 10 ml/d, and 300 ml/d.

The Bead Leach II experiment was further complicated by the addition of simulated uranium fuel rods in the form of UO_2 monoliths. These were leached in the same manner as the glass beads. Leachates from all samples were collected at prescribed intervals during the experiment, and selected leachates were analyzed for plutonium, neptunium, uranium, boron, and molybdenum in the case of the glass beads and only for uranium in the case of the

Figure 39 Leaching cells used in the Bead Leach II experiment

UO₂ samples. Also, the rock columns from each cell were analyzed at the conclusion of the leaching phase to establish the amount of dissolved material attached to the rock. The rock column was divided in four portions and each of these was analyzed for plutonium, neptunium, uranium, boron, and molybdenum retention. Because of the complexity of the experiment and the great number of leachates and rock samples to be analyzed, we report the results of the Bead Leach II experiment in two parts. The results pertaining to the leaching of the glass beads with the simulated basalt groundwater constitute Part I.² The work on the glass beads with the simulated brine-sandstone groundwater is still in progress, as is all of the work on the UO₂ monoliths. This portion of Bead Leach II will be reported at the end of this year.

Results from Part I of the Bead Leach II experiment are:

- Plutonium and neptunium concentrations were determined in all but a few of the leachates from the glass-only samples. The measured concentrations and the known surface area of the beads were used to calculate leach rates. (Leach rate is defined as the amount of glass material dissolved per unit area and unit time—usually expressed as g/m²·d.) These calculated leach rates range from 0.0001 to 0.01 g/m²·d for plutonium and from 0.001 to 0.1 g/m²·d for neptunium. They are consistent with previous results obtained in Bead Leach I.

- There is both a temperature effect and a flow-rate effect on the leach rates exhibited by plutonium and neptunium; the leach rates increase with increasing temperature and increasing flow rate.

- With the exception of very few leachates, most analyses show nondetectable amounts of

boron, uranium, molybdenum, and cesium at 25°C. At the higher temperature, 75°C, concentrations of these elements in the leachates translate into leach rates ranging from 0.1 to 1.0 g/m²·d.

- Leachates from the leaching cells containing both glass beads and crushed basalt were also analyzed for plutonium, neptunium, uranium, boron, molybdenum, and cesium. Their concentrations were compared with concentrations for the same elements adsorbed on the rock surfaces.

- At 25°C, the plutonium adsorbed on the rock is 2–10 times larger than the amount found in the leachate. The plutonium deposited on the rock is located mostly in the layer nearest to the glass beads as shown in Fig. 40 (Ch 9, 11, and 12). Neptunium, uranium, molybdenum, and cesium were not detected on any of the rock layers. Therefore, it is not possible to state if there is any adsorption of these elements at 25°C.

- At 75°C, the plutonium adsorbed on the rock is 10–100 times larger than the amount found in the leachate. About 80–90% of the plutonium on the rock is located in the layer nearest to the glass beads as shown in Fig. 40 (Ch 40, 41, 44, 67, and 71). The neptunium adsorption appears to be flow-rate dependent; at 1 ml/d and 10 ml/d, there is about twice as much neptunium on the rock as there is in the leachate, while the opposite appears to be true at 300 ml/d. In the case of uranium, the total amount adsorbed on the rock is always less (from 2 to 6 times) than what is found in the leachates, depending on the flow rate. The distribution of both neptunium and uranium on the rock column is similar, as shown in Figs. 41 and 42.

- Molybdenum was not detected on any of the rock samples. Therefore, a comparison of molybdenum concentration in the leachate and on the rock is not possible. Boron appears to behave similarly. We conclude that both are not readily adsorbed on basalt.

- Cesium does not adsorb on the rock at 25°C, but it appears to adsorb at 75°C only at 1 ml/d and 10 ml/d. About 80% of the leached cesium remains on the rock at 1 ml/d and about 50% at 10 ml/d. Thus, both temperature and flow rate appear to affect adsorption of cesium on basalt.

This research was supported by a contract with Battelle Pacific Northwest Laboratories as part of the Waste/Rock Interactions Technology Program.

Figure 40. Plutonium adsorption (%) on rock.

Figure 41. Neptunium adsorption (%) on rock. (ND = not detectable.)

Figure 42. Uranium adsorption (%) on rock. (ND = not detectable.)

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Radiation Chemistry in a High-Level Nuclear-Waste Repository in Unsaturated Tuff

R. A. Van Konynenburg

One of the candidate nuclear-waste repository sites proposed by the Department of Energy is in a tuff formation inside Yucca Mountain in Nevada. Radiation chemical reactions will occur in the media surrounding high-level nuclear-waste packages in the repository. We are studying their likely effects on corrosion of metal canisters and release of radionuclides.

The Nuclear Waste Policy Act of 1982 established a timetable for activities leading to construction of repositories for high-level nuclear waste generated by commercial nuclear power and defense activities. Lawrence Livermore National Laboratory is responsible for designing the waste packages for the Yucca Mountain tuff repository and predicting their behavior over long time periods. Federal regulation 10CFR60 specifies that the waste packages must provide substantially complete containment of all radionuclides for the first 300 to 1000 y, and release of no more than one part in 10^5 per year for the next 100,000 y. The Nuclear Chemistry Division is providing technical advice on design and interpretation of experiments to study corrosion, dissolution of radionuclides, and chemical interaction of components of the waste package with each other and with the environment, under gamma irradiation and various temperature conditions. This work is being carried out in cooperation with workers from the Chemistry and Materials Science Department and the Earth Sciences Department.

The principal mechanism that could lead to penetration of the containment offered by the waste package is thought to be corrosion. The principal mechanism that could remove radionuclides from the waste is believed to be leaching or dissolution in groundwater by chemical reactions in the environment of the packages. Both processes could be influenced by chemical effects produced by the radiation emitted from the waste. As a result, we are

exploring the likely radiation chemical behavior in this environment, primarily emphasizing effects on corrosion.

Although a variety of radiation types are emitted by the waste, only the gamma radiation is sufficiently penetrating and intense to produce significant effects on corrosion while the containment is intact. Once groundwater and air have penetrated the package, alpha particles could have an influence on the leaching or dissolution. During both these periods, radiation damage to the solid waste form itself can arise, primarily due to recoil nuclei from internal alpha decays. This could be important if it led to swelling of the waste form, producing mechanical stresses in the package, or to a damaged structure which could be more readily leached. Past work by others has generally shown those effects to be small for waste forms currently being considered.

There are three waste forms currently under consideration: two borosilicate glasses from reprocessing of both defense and commercial nuclear fuel, and commercial spent fuel itself. The initial gamma-ray dose rate in the vicinity of the packages will be in the range 10^3 to 10^5 rads per hour, depending on the waste form. The mean energy of the source gamma rays will be about 0.6 MeV. The gamma rays will interact via the Compton and photoelectric effects, producing high-energy recoil electrons. These in turn will produce ionization in the media around the waste packages, which can lead to chemical changes.

Figure 43. Capsule used to study the corrosion of metals in a nuclear-waste-repository environment.

The probable environment includes the tuff itself, perhaps a metallic hole-liner, perhaps some backfill material, and the fluid medium. The Topopah Spring tuff, which is the member proposed for the repository, is a welded, devitrified volcanic rock, consisting mainly of fine grains of alkali and plagioclase feldspars, quartz, and cristobalite, with large phenocrysts of various mineral species. This rock has about 13% porosity, and is located above the water table so that the pores are only about 70% filled with water, the rest being air. The annual percolation of water due to rainfall is expected to be only a few millimeters, since most of the rainfall runs off the surface.

After the waste packages are emplaced, the surrounding media will be subjected to gamma irradiation as well as heating. When the rock is heated above the boiling point of water, the resulting steam is expected to drive most of the air out of the immediate package environment. The period during which steam will prevail near the packages will range from about 100 to 1000 y, depending on the waste form. After this time, the packages will have cooled to below the boiling point, allowing the steam to condense and drawing air back into the package environment. At this stage, however, the gamma flux will be much lower, resulting as it does primarily from ^{137}Cs , which has about a 30-y half-life.

Because of these thermal effects, the medium that will prevail during most of the high-dose-rate period is steam. Corrosion during this period is expected to be slow, and radiation chemical effects are expected to be minor. When the waste package (probably constructed of stainless steel) drops below the boiling point, it will be possible for a liquid film to exist on its surface, and air will be present. Past work by others has shown that nitric acid and other species such as O_2^- ions, HO_2 radicals, and H_2O_2 molecules are produced by radiolysis under these conditions. These species can lead to faster corrosion. Past work by others has shown that feldspar minerals will rapidly exchange potassium and sodium ions for hydrogen ions. This should serve to buffer the nitric acid, and produce sodium and potassium nitrates and clays.

The first experiment has been in progress for about 6 months. It is designed to study the corrosion of 304L stainless steel at 30°C in the presence of crushed tuff, aerated J-13 water

(from a well that drains the tuff deposit in Nevada), and a gamma field of about 6×10^5 rads/h. The experimental capsule is shown in Fig. 43. Water is circulated slowly through the capsule during the irradiation. There are some samples in the "sensitized" and some in the "non-sensitized" conditions. (Sensitization involves annealing the material under the appropriate time and temperature conditions to form chromium carbide precipitates, thereby making the steel more vulnerable to intergranular-stress corrosion cracking. This is important near welds in the waste canisters.) The capsule is located a few centimeters from a 20-kCi ^{60}Co source, submerged in a water-filled pit.

The samples have been visually examined twice so far. The last examination took place at 4000 h into the experiment. No appreciable corrosion was observed on either the sensitized or non-sensitized samples. Duplicate samples in a second capsule running without gamma irradiation likewise have shown no appreciable corrosion. We plan to continue this experiment for a total duration of 1 y. At that time the water will be chemically analyzed, the samples will be checked for weight loss, and any observed corrosion will be characterized by metallography and scanning electron microscopy.

Meanwhile, more elaborate apparatus is being assembled to permit experiments at elevated temperatures, to simulate the heating due to radioactive decay of fission products and actinides in the waste.

Although these corrosion results are long in coming and have not been very dramatic so far, this is exactly what one hopes for in a material to be used for containment of high-level nuclear waste. The task that lies before us is to project results obtained in times of the order of a year to behavior of high-level waste packages over geologic time periods. This will necessitate gathering of data under more extreme conditions of irradiation, temperature, and environmental chemistry than are expected, and projecting to longer-time behavior under more moderate conditions, using models for corrosion processes. While we cannot hope for absolute predictability with our current understanding of corrosion processes, it should be possible to help in making intelligent choices of materials that will perform the task at hand.

This research was sponsored by the Nevada Nuclear Waste Storage Investigations.

Field and Laboratory Studies of Radionuclide Migration at the Nevada Test Site

J. H. Rego

The radionuclide migration project investigates radionuclide transport away from sites of high radionuclide deposition resulting from underground nuclear tests; groundwater contamination is a primary but not exclusive focus. Our goal is to predict migration in a way that will be relevant to other possible sites of radioactive contamination such as nuclear power plants, nuclear-waste disposal sites, spills and releases, etc. We continued our studies of leaching from melt glass (vitrified cavity material), and devised new techniques for validating very-low-level determinations of radionuclides in groundwater samples. We also developed a chemical procedure to improve detection sensitivity for ^{106}Ru .

The radionuclide migration project consists of three major efforts:

- We analyze samples and review data from present and potential field experimental sites at the Nevada Test Site in order to detect and characterize actual radionuclide migration.
- We conduct laboratory experiments to identify the release and transport characteristics of relevant radionuclides and to characterize potential sources.
- We design new experiments and develop methods in support of both the laboratory and field efforts.

The laboratory studies to date have concentrated on the radionuclide-release characteristics of nuclear melt glasses. We have developed a practical approach using a single-pass continuous-flow leaching method to study this phenomenon.¹ In 1983 we published the results of the first part, *A Leaching Study of Nuclear Melt Glass: Part I*,² and completed the data analyses for the second part (Melt Glass II). The experiments are intended to simulate conditions in which a cavity formed below the water table is refilled with the local groundwater, creating the potential to transport radionuclides downstream under natural conditions. The melt glasses for the second part of the experiment were specifically selected for

studying leaching effects on glasses 5–18 y after detonation, and to represent sites at which there are also field experiments in progress or proposed. Because of the ages, the activities of the melt glasses are low, often resulting in counting rates at or very near background levels. Consistent and measurable levels of ^{137}Cs were observed, and less consistent but generally measurable levels of ^{60}Co activity were found. The nuclides ^{54}Mn , ^{57}Co , ^{106}Ru , ^{125}Sb , ^{144}Ce , and ^{241}Am were only sporadically detected.

Figure 44 shows the leach-rate data obtained in experiments I and II for ^{60}Co removal from a glass that was run as control in both experiments. There appears to be a modest but significant difference between the two experimental runs; however, all of the glasses measured in both experiments generally yield leach rates of comparable magnitude and with comparable time dependence. The cumulative amount of each radionuclide leached over 420 d was typically 0.1 to 0.3% of the original total glass activity, regardless of the age of activity of the glass. Although leach rates decreased with time, measurable leaching was still occurring at the end of the 420-d period.

On the basis of these experiments and the available data for *in situ* surface areas and activities, it is clear that melt glass is unlikely to

Figure 44. Leach rates of ^{60}Co from a melt glass used as a control in experiments Melt Glass I and II.

be a source of significant release of short-lived radionuclides to the environment. Other sources (e.g., chimney rubble) and longer-lived radionuclides are still under consideration.

The applicability of the laboratory experiments is limited by the lack of reliable field data on which to base predictions that can be extrapolated over large distances or significant times. In collaboration with the Earth Sciences Department at LLNL, we are preparing a field report that documents the history of and studies at sites where the geology, hydrology, and radioactivity are of interest for possible radionuclide-migration studies. The report will contain a summary of all the analyses of field samples completed by Nuclear Chemistry to date and will be used for both interpretation of existing data and design of future experiments.

Two reopened "holes of opportunity" are being evaluated for possible further field studies. The Faultless Event site is unique in that it is located in a isolated central Nevada valley that was hydrologically characterized before the detonation and where there are no other potential sources of radionuclide migration. The second site is Bourbon, where the event was detonated below the top of the Paleozoic rocks in the unsaturated zone. A satellite well that penetrates the saturated zone 120 m below the detonation point is located 137 m (horizontal distance) away from the working point. The well has recently been equipped with a solar pump to provide continuous withdrawal from the very-low-yield aquifer, and water can now be

analyzed for possible contamination of the saturated Paleozoic formation.

A significant effort has been devoted to validation of very-low-level determinations of radionuclides in groundwater samples. In particular, the consistent presence of small amounts of ^{137}Cs in well RNM-2S (Cambrian Event satellite well) was investigated by use of chemical separation techniques and comparison of low-level beta and gamma measurements on samples taken by different methods. We demonstrated that the observed ^{137}Cs was almost certainly traceable to contamination (probably by resuspended NTS soil) during sampling and not to radionuclide migration in the groundwater. This has sparked a renewed interest in investigation of the sensitivity and validity of commonly accepted sampling and analytical techniques.

In 1978 to 1980 we detected low but measurable levels of ^{106}Ru (less than 0.1 pCi/l) in well RNM-2S water. The ratio of tritium to ^{106}Ru indicated that some of the ruthenium produced by the Cambrian detonation was migrating away from the cavity at a rate close to the groundwater flow rate. The presence of the ruthenium surprised many workers since laboratory studies predicted much slower migration rates for ruthenium.³ The short half-life of ^{106}Ru (1.01 y) and very low concentration in the water from the cavity eliminate concern that ruthenium migration presents a hazardous situation. However, we feel further data based on more careful monitoring techniques are needed to understand the migration of such mobile radionuclides.

We developed a chemical procedure to improve the detection sensitivity for ^{106}Ru . The separation procedure reduces the concentration of other radioactive species present in the sample. This reduces the 511-keV annihilation peak, thereby reducing the interference with one of the principal gamma rays used to detect ^{106}Ru (511.8 keV). The procedure also reduces the sample volume and improves the counting geometry, which allows us to determine ^{106}Ru at concentrations of approximately 2 fCi/l. (Our previous detection limit was about 0.02 pCi/l when the salts from 200 l of groundwater were counted using our low-level Compton-suppression detection system.)

This method will be used in the future to check the analysis of samples when the presence of ^{106}Ru is possible or suspected. Currently, none

of the wells being sampled is from a sufficiently recent test to contain measurable ^{106}Ru in the water.

The Cambic Event site is also one of the field sites of potential interest for further mechanistic studies. Water has been pumped continuously from the satellite well for the past nine years; this water has been carefully monitored ⁴ and has flowed through a canal to an evaporation area on the desert floor for that entire time. This unusual condition—a constantly saturated recharge source with a well-characterized history of radionuclide concentration imposed on the

desert soil—has created a shallow subsurface plume of water that is reasonably accessible for study of both water and radionuclide transport under conditions of saturated flow, unsaturated flow, and vapor transport. A site for some preliminary experiments has been established, and we anticipate that the results obtained will be useful for understanding radionuclide movement in geologically similar deeper aquifers.

This research was supported by the DOE Nevada Operations Office.

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Safeguards Technology and Radiation Effects

5

Overview

The Division supports improvements in the safeguarding of special nuclear materials by developing new instruments, methods, and techniques to enhance their control and accountability. In addition, we have the charter to transfer these new developments to DOE contractor sites such as the Savannah River Plant (Savannah, GA), the Idaho Chemical Processing Plant (Idaho Falls, ID), and the Hanford Engineering and Development Laboratory (Hanford, WA). Since 1971, we have focused our research and development activities in the area of material control and accountability and, more recently, on the best ways that gamma-ray and x-ray spectrometry can be used for process control and the inventory and accountability of special nuclear materials (in particular, the fissile isotopes of plutonium and uranium). The Division is also involved in international safeguards activities through the Program of Technical Assistance to Safeguards, funded primarily by the U. S. Department of State.

In this section we report on a computer-based energy-dispersive x-ray fluorescence analysis system that we designed for the Savannah River Plant. This system determines the concentration of solutions containing quantities of plutonium as large as 100 grams per litre and occasionally mixtures of uranium and plutonium. It provides both day-to-day process control information and monthly accountability and inventory data for plutonium process and product lines. A similar system installed at the Allied General Nuclear-Service Plant (Barnwell, SC) gave the first stream-concentration information ever obtained on-line and in near-real time at any reprocessing plant.

We also report work that describes a formula to properly treat the propagation of errors associated with gamma-ray spectroscopic and calorimetric measurements used for accountability of large quantities of plutonium.

Finally, we discuss the work sponsored by the Program of Technical Assistance to Safeguards. Here we made improvements to a portable microcomputer that we developed in FY82 for the International Atomic Energy Agency (Vienna, Austria). This computer supplements a spectrometer used by inspectors for in-field analysis of plutonium gamma-ray spectra. We added additional operational features and improved the data-analysis software.

The Division also conducts research on radiation damage to materials of particular interest to the Magnetic Fusion Energy Program. In this section, we report on calculations to assess thermal-neutron-radiation damage in natural copper, molybdenum, vanadium, and tungsten. Here we used the statistical model for thermal-neutron capture and calculated the damage due to recoils from the subsequent gamma-ray emission.

We also measured the change that occurs in the magnetoresistivity of copper at magnetic fields up to 12 tesla during fusion-neutron irradiations at about 4 degrees Kelvin alternating with anneals to room temperature. These measurements will provide data for the design of the stabilizers required for the superconducting magnets used in fusion reactors.

X-Ray Fluorescence Analysis of Uranium and Plutonium in Chemical Processing Streams

D. C. Camp and W. D. Ruhter

When spent nuclear fuels are chemically processed to recover uranium and/or plutonium, the concentrations of these products must be monitored for process- and inventory-control purposes. Energy-dispersive x-ray fluorescence analysis (EDXRFA) is one technique that can monitor uranium and plutonium concentrations in processing streams rapidly, quantitatively, and nondestructively.^{1,2} During the past year we designed and developed uranium- and plutonium-measurement systems utilizing EDXRFA for installation at the Idaho Chemical Processing Plant (ICPP) and the Savannah River Plant (SRP). The ICPP system will measure uranium concentrations in an organic process stream as often as every 3 min. The SRP system will measure plutonium concentrations in nitric solutions automatically drawn from a process storage tank into a measurement cell.

In x-ray fluorescence analysis (XRFA), atoms of interest within a sample are fluoresced and the intensities of the characteristic x rays emitted are measured. From the measured intensities the concentrations of the atoms being fluoresced can be determined. In this particular application of XRFA, 122-keV gamma rays from collimated ⁵⁷Co sources are used to fluoresce uranium and/or plutonium atoms in solutions. The stimulated K x rays from the uranium and/or plutonium atoms are detected by a collimated high-purity-germanium (HPGe) detector. Pulses from the detector are analyzed by a multichannel analyzer and the resulting spectral data are processed by computer programs to yield uranium and/or plutonium concentrations.

Both the 122-keV exciting radiation and the K x rays have sufficient energy to penetrate several millimeters of stainless steel. Therefore, this technique is easily adapted to on-line measurements, since in most processing facilities the process solutions flow within stainless steel pipes. The source-detector geometry has been optimized for a cylindrical cell geometry, i.e., pipes, cylindrical cells, or test tubes.

At the ICPP (Idaho Falls, ID) uranium is separated from fission products in three

successive decontamination columns. The need is to monitor continually the uranium concentration in the product stream after the first column. A "normal" stream at this point contains a few grams of uranium per litre in an organic solution of 93% dodecane + 7% tributyl phosphate. Most of the fission products have been removed; however, the amount remaining will vary depending on stream conditions and the type of fuel being processed.

A portion of this stream will be diverted through a stainless steel cell which is welded in-line. The XRFA assembly is coupled to this cell. The assembly, sample cell, and pipes are shielded by lead and tungsten in a stainless steel enclosure to reduce radiation exposure to personnel. Since ⁵⁷Co has a 270-d half-life, it is necessary to change the exciting sources only once per year. To facilitate this change without disassembly of the lead enclosure, the ⁵⁷Co sources are mounted on the ends of 12-in.-long rods that extend outside the enclosure.

Since a variable amount of fission product activity remains in the product stream, some means must exist to control the maximum count rate experienced by the HPGe detector. We designed a mechanism (shown in Fig. 45) that

provides manual selection of three different collimator openings, which limit the amount of fission-product radiation that can be seen by the detector.

The ^{57}Co -source strengths of 50 mCi each were chosen to excite sufficient uranium x-ray intensity for the range of uranium concentrations expected, regardless of the fission-product radiation intensity. The XRFA system yields a 2% accurate (1σ) concentration value in 500 s for 1 g uranium per litre, using the medium-sized collimator (7.0-mm-diam). The large (12.1-mm-diam) and small (2.6-mm-diam) collimators allow a net uranium K x-ray intensity three times greater and seven times smaller, respectively, than the medium collimator does.

Uranium concentrations are determined by obtaining the ratio of the net uranium K_{α_1} x-ray intensity to the gross number of counts within a window that includes the incoherent scattering peak of the 122-keV exciting radiation (the gross intensity, or GI).¹ By calibrating the system using uranium solution standards, the relationship between this ratio and the measured concentrations can be established. This ratio is independent of source exciter half-life, changes in dead time, and small changes in geometry. If excessive fission-product activity is present, it

will contribute to GI, and decrease the concentration. The system is currently programmed to test for excessive fission-product background once each hour. If it is excessive, a computer-controlled shutter is used to eclipse the exciter sources and then a background spectrum is taken. This background spectrum is subtracted from the fluorescence spectrum to obtain the correct GI value.

The computer-based multichannel analyzer system reports concentration values at the end of each analysis. Every hour an average value is calculated and printed for all measurements made during the past hour. Every 24 h the preceding day's hourly averages are listed.

At the Savannah River Plant (Savannah, GA) the XRFA assembly will be coupled to a process accountability tank. Plutonium solution will be automatically pulled by vacuum from the tank into a specially constructed stainless steel cell. A "normal" solution at this point contains 2-4N nitric acid and fresh, chemically separated plutonium (no ^{241}Am). The plutonium concentration will be several grams per litre.

Plutonium K x rays are excited by two 20-mCi ^{57}Co sources. A third highly collimated ^{57}Co source is located to allow a 122-keV beam to be transmitted through the solution into the

Figure 45. Cross section of the ICPP's XRFA system designed to yield near-real-time concentrations of uranium in processing streams. The three manually selectable collimator openings provide detector-count-rate adjustment for partially separated fission products. The removable rods that position the ^{57}Co sources are not shown.

detector. The ratio of the net plutonium K_{α_1} x-ray intensity to the 122-keV transmission intensity is obtained to determine the plutonium concentration. A calibration procedure establishes the relationship between this ratio and the concentration. This method measures concentrations independent of minor variations in the composition and density of the plutonium solution.

To achieve better than 0.5% accuracy for accountability measurements, it is necessary to remove the solution's natural x- and gamma-ray contributions from the fluorescence spectrum. This is accomplished by eclipsing the exciting sources with a computer-controlled shutter and then measuring the solution's natural radioactivity spectrum. The system's data-analysis software subtracts this spectrum from the fluorescence spectrum, thus obtaining

plutonium concentration information independent of plutonium isotope variation. The system is designed to yield 2-5% process control information in less than 5 min, and concentrations for accountability purposes in times not exceeding 30 min.

The ICPP uranium XRFA system is scheduled for on-line installation in October 1983. The SRP plutonium XRFA system is to be tested at the Savannah River Laboratory throughout October and November 1983. Installation in the Plant for on-line plutonium analysis is scheduled for January 1984. It should be noted that although the measurement technique, XRFA, is the same for both installations, the engineering required to apply the technique is different in many aspects for each installation.

This research was supported by the DOE Office of Safeguards and Security.

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The Propagation of Errors in the Measurement of Plutonium-Isotope Composition by Gamma-Ray Spectroscopy

T E Sampson¹ and R Gunnink

Accountability measurements of large quantities of plutonium are frequently made by combining a gamma-ray spectrometric measurement of the relative isotopic abundances with a calorimetric measurement of the power output. The error associated with such an accountability measurement results from the errors arising from these two measurements. We found that the method for propagating these errors reported in the 1975 ANSI calorimetry standard is incorrect, and we developed appropriate error formulas that are applicable to three measurement cases.

In recent years, several U S laboratories have developed computer codes to analyze the gamma-ray spectra from plutonium samples in order to determine the isotopic composition of the plutonium.²⁻⁵ These techniques are, in general, versatile, accurate, and precise. For some cases, the precision of the measurement approaches that obtained using conventional mass spectrometric techniques. The measurements have been applied to reprocessing-plant solutions as well as to solids and many forms of scrap and waste.

These nondestructive techniques are particularly useful in conjunction with calorimetry when applied to bulk solids to obtain accountability information and to resolve shipper/receiver differences. For use with calorimetry, it is important that the measurement uncertainty in the specific power arising from the gamma-ray measurement be determined accurately so that the uncertainty in the total plutonium content can be estimated properly.

The plutonium mass of a sample measured by calorimetry is given by

$$M = W P \quad (1)$$

where M = plutonium mass in grams, W = sample power in watts, and P = sample specific power in watts per gram of plutonium.

The sample specific power P is given by

$$P = \sum K_i f_i \quad (2)$$

where K_i = specific power of the i^{th} isotope in the sample in watts per gram of isotope, $i = {}^{238}\text{Pu}, {}^{239}\text{Pu}, {}^{240}\text{Pu}, {}^{241}\text{Pu}, {}^{242}\text{Pu}, {}^{241}\text{Am}$ for most samples, f_i = isotopic fraction of the i^{th} isotope in the sample in grams of isotope per grams of plutonium.

In Eq. (1) the measurement of W is independent of the measurement of the specific power P and thus the combination of uncertainties in the two measurements is straightforward. In Eq. (2) the uncertainty in P is a function of the uncertainties of the isotopic fractions, f_i . For this analysis we neglect the uncertainties in the isotopic specific powers, K_i , because they only contribute a bias to the final result but do not contribute to the statistical precision of P .

It is tempting to compute the uncertainty in P by propagating assumed independent errors in the isotopic fractions in Eq. (2).⁶ However, this is incorrect. The isotopic fractions, f_i , are correlated because of the constraint that

$$\sum_i f_i = 1.0$$

where $i = {}^{238}\text{Pu}, {}^{239}\text{Pu}, {}^{240}\text{Pu}, {}^{241}\text{Pu}, {}^{242}\text{Pu}$ for most samples.

Thus the isotopic fractions, f_i , are not independent variables. Although it can be seen that Eq. (21) of ANSI N15.22⁶ is in error in this regard, it is more difficult to write the correct expression in the general case because the correct expression depends upon the details of the gamma-ray measurement.

We derived error expressions for the isotopic fractions and the specific power values of the sample by considering the uncertainties in the fundamentally measured independent variables of the problem. In our study, we considered three different analysis schemes that are in use or could possibly be used in the future.

In the first case we assumed that the ${}^{238}\text{Pu}$, ${}^{239}\text{Pu}$, ${}^{240}\text{Pu}$, ${}^{241}\text{Pu}$, and ${}^{241}\text{Am}$ were independently measured, resulting in independent measures of the corresponding standard deviations. In the second case, we assumed the measurement of four independent isotopic ratios, as measured with respect to ${}^{241}\text{Pu}$ ("independent" meaning that there are no common peaks among the eight different peaks

forming the four ratios). In the third case, we again assumed independently measured isotopic ratios, except this time with respect to ${}^{239}\text{Pu}$.

The equation derived for the variance in the power, $\sigma(P)$, for the first case is given by

$$\sigma^2(P) = \sum_i (P - K_i)^2 f_i \sigma_r^2(A_i) + K_{\text{Am}}^2 f_{\text{Am}}^2 \sigma_r^2(A_{\text{Am}}) \quad (3)$$

where K_i = specific power (watts/gram) of isotope i , A_i = amount of isotope i in sample, f_i = isotopic fraction of i (i.e., $A_i/\Sigma A_i$), $P = \Sigma K_i f_i$, $\sigma_r = \sigma(f_i)/f_i$.

Similar but more complex expressions were derived for the other two cases.

The agreement of the precisions predicted by our equations with the precision of observed measurements was found to be satisfactory for two published data-analysis methods. We also illustrated the sensitivity of the specific power to biases in any of the independent measured variables.

This research was supported by the LLNL Safeguards Technology Program.

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Implementation of LLNL Microcomputer for IAEA Plutonium-Isotope Analysis

W. D. Ruhter

We assisted the International Atomic Energy Agency (IAEA) in implementing a portable microcomputer for in-field analysis of plutonium gamma-ray spectra. We designed and developed the portable microcomputer and the data-analysis software.^{1,2} The implementation included upgrading of the data-analysis software, participation with the IAEA in testing and evaluating the software, construction and delivery of three additional units, and training of IAEA inspectors in the use of the units. We have completed this work except for parts of the training program and are preparing a final report on the hardware, software, and operation of the portable microcomputer. The IAEA finds the results obtained with the portable microcomputer data-analysis software quite satisfactory and has requested the manufacturer of its multichannel analyzer to incorporate this software into the analyzer.

Early in FY81 we delivered to the IAEA (Vienna, Austria) a prototype of a portable microcomputer for in-field analysis of plutonium gamma-ray spectra.^{1,2} Such spectra are taken by IAEA inspectors in the normal course of their duties using a 1000-channel pulse-height analyzer and amplifier that are 10 years old in technology. In late FY81, IAEA personnel completed an extensive in-field test and evaluation of the prototype microcomputer unit and issued a report on the results.³ In general, the report found the results obtained with the microcomputer satisfactory, but it recommended that several modifications and additions be made to the data-analysis software. It included a recommendation that the software be modified to compensate for effects of pulse pileup in the gamma-ray spectral data. Most of the pulse pileup and resolution problems were due to the poor performance of the older amplifier and analyzer.

In March 1982, we received funding to help the IAEA implement the portable microcomputer for field use. This work included upgrading of the data-analysis software, construction and delivery of three additional units, test and evaluation of the new software, and

development of a training program for use and maintenance of the microcomputer. We modified the data-analysis software extensively, including modifications to alleviate the problems caused by pileup. We shipped a new microcomputer with upgraded software to the IAEA in September 1982. We were to participate in a test and evaluation of the new software at IAEA Headquarters in October 1982. However, because of a boycott action taken by the U.S. Department of State against the IAEA, we were not permitted to work with the IAEA and the test and evaluation did not occur as planned.

While this was being resolved, the IAEA decided to begin using a new multichannel analyzer from Silena which contains a much-improved amplifier. This unit, shown in Fig. 46, should give significant improvement in energy resolution and pulse pileup, particularly at high input count rates. This should also improve the plutonium-isotope results obtained with the portable microcomputer. The IAEA decided to conduct the test and evaluation of the new data-analysis software with the new analyzer/amplifier and to collect the test spectra in 2000 channels. This required that we further modify the microcomputer software to allow it to

Figure 46. The portable microcomputer connected to the new Silena multichannel analyzer. The amplifier is contained within the analyzer. (The amplifier controls are on the right side of the analyzer.)

communicate with the new analyzer and to process data contained in 2000 channels.

In May 1983, we were permitted to travel to IAEA headquarters to work with IAEA personnel on evaluating the new data-analysis software. We first modified the microcomputer software to permit its use with the new analyzer. We then proceeded with the evaluation using data that had been taken of well-characterized plutonium samples with the new analyzer/amplifier.

The results obtained in this test and evaluation were quite good.⁴ The IAEA is pleased with the performance of the data-analysis software and their next step is to transfer this software to a commercial microcomputer system. The four

portable microcomputer units that the IAEA has are inadequate for their needs. They have asked Silena, the manufacturer of the new analyzer, to incorporate our data-analysis software into their new analyzer. The analyzer has a microprocessor and memory space available for such purposes. This will provide the IAEA with additional data-analysis units and will eliminate the need for an additional piece of equipment. We expect to work with the IAEA and Silena in the transfer of our software and in testing the final product.

This work was supported by the U.S. Department of State and was administered by the International Safeguards Project Office.

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Tracer Techniques for Studying Pyrochemical Processing of Plutonium

W. D. Ruhter, M. S. Coops,¹ G. J. Koskinas,¹ R. Gunnink, and A. L. Prindle

We used gamma-ray spectrometry to determine distribution coefficients of the light-actinide elements uranium, neptunium, plutonium, and americium in high-temperature extractions (pyrochemical) of magnesium-based metal alloys by chloride salts. Our method is to use suitable masses of light-actinide elements "tagged" with gamma-ray-emitting radioactive isotopes; samples are withdrawn from the salt and metal phases and gamma-ray measurements are made with a Ge(Li) spectrometer. The resulting data are analyzed using the GRPANL computer code. Our most recent experiments determined the distribution of the light-actinide elements between phases of molten copper-magnesium alloys and MgCl_2 -NaCl-KCl molten salt.

Pyrochemical processes that utilize molten alloys as solvents and molten salts as extractants have unique advantages over traditional aqueous and organic processes for partitioning highly radioactive wastes²; pyrochemical processes are able to tolerate more readily the extreme environmental conditions encountered. Research is being conducted to obtain the extraction data needed to develop a highly compact, continuous-flow molten-salt processing system that can be used to recover the actinide elements from highly irradiated nuclear fuels without requiring years of cooling prior to chemical processing.

Recent measurements conducted by Chemistry and Materials Science Department personnel obtained distribution coefficients for the light-actinide elements between molten magnesium chloride extraction salt and varying compositions of copper-magnesium alloys. The distribution data for each actinide were obtained by gamma-ray spectroscopic analysis. Suitable masses of gamma-ray-emitting isotopes of uranium, neptunium, plutonium, and americium were added to the molten salt-metal mixture to trace the equilibrium distribution of the elements during the experiment. Small metal and salt samples were removed from the extraction phases between sequential additions of copper to the liquid metal extractor. After weighing, each

sample was dissolved and diluted to a predetermined volume in a standardized polyethylene counting vial. The gamma-ray spectra were measured with a well-calibrated Ge(Li) spectrometer system, and the resulting data were analyzed using the GRPANL computer program. Nuclear Chemistry Division personnel provided the calibrated Ge(Li) system and support for the design and implementation of the gamma-ray data analysis.

The GRPANL program, developed in the Nuclear Chemistry Division,³ is a general-purpose program for unfolding and interpreting specified spectral regions containing gamma- and x-ray peaks. Very complicated overlapping peak groupings can be fitted accurately because the user (in general, a well-trained spectroscopist is required) specifies and controls the analysis of the peak grouping. The GRPANL program can be run on a 16-bit 32K-word computer system and is a convenient, useful tool for gamma-ray spectra analysis.

For this particular experiment one of the peak groupings, shown in Fig. 47, required GRPANL to fit 11 overlapping gamma- and x-ray peaks to determine the ^{238}Pu and ^{243}Am abundances. The GRPANL program assembles the energy and intensity results of one or more groups from one or more spectra to form a file of reduced data.

Figure 47. Fit to 11 overlapping peaks using the GRPANL program. The peaks with long low- and high-energy tails are x rays with Lorentzian distributions. This plot was drawn from that generated by the GRPANL program on the Nuclear Chemistry Division's VAX 11/750.

This file, in conjunction with fundamental nuclear-decay-scheme information contained in other files, is automatically interpreted to identify the isotopic species present and to provide an accurate assay of each isotope. With these data the distribution coefficients, K_D , can be calculated for each element represented by a gamma-ray-emitting tracer. Gamma-ray analysis provides a powerful technique for determining extraction parameters for many different alloys and molten-salt systems.

The distribution coefficient results obtained from the molten magnesium chloride and copper-magnesium alloy system experiment are shown in Fig. 48. (The cerium results were obtained from a previous experiment.) These results indicate a sample extraction method for chemical partitioning of americium metal (and other 3^+ actinides) from metallic 3^+ lanthanide metals. Previous data⁴ from similar extractions between magnesium chloride extraction salt and zinc-magnesium alloys show no significant separation between 3^+ actinide and lanthanide

Figure 48. Distribution coefficients for cerium, uranium, neptunium, plutonium, and americium in MgCl_2 - NaCl - KCl salt vs copper-magnesium alloys (at 1100 K).

metals. This development provides a simple, rapid method for purification of americium (and other heavy actinides) from lanthanide elements such as fission products.

This work was supported by the LLNL Chemistry and Materials Science Department's research and resource funds.

Chemistry and Materials Science Department, Lawrence Livermore National Laboratory

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Application of Statistical Nuclear Models to Thermal-Neutron-Radiation Damage in Metals

J. H. Kinney,¹ M. W. Guinan, and Z. A. Munir²

We used a statistical nuclear model to calculate the recoil energies from thermal-neutron capture in natural copper, molybdenum, vanadium, and tungsten. These recoil calculations have allowed us to reexamine the results of previous thermal-neutron-radiation-damage experiments in terms of the modified Kinchin and Pease model of defect production in metals.

Electron, ion, and neutron irradiations of a number of metals have demonstrated that the defect-production efficiency, as defined by the modified Kinchin and Pease expression, decreases with increasing recoil energy up to roughly 100 times the threshold energy for producing damage.³ The parameters for displacement functions that are used to predict the responses of materials to energetic radiation have been fitted to the results of charged particle irradiations in this intermediate-recoil-energy region. Because the primary recoil spectra that are characteristic of these ion irradiations are broadly distributed over the recoil-energy region where the efficiency is rapidly changing, errors can be introduced into the resulting calculated displacement functions. These errors can significantly alter the estimates of the radiation damage expected in many components of fusion reactors.

It has been known for many years that thermal-neutron-capture reactions in many metals produce a source of narrowly distributed intermediate-energy recoils. Until now, however, the inability to accurately calculate the neutron-capture recoil spectrum has prevented researchers from fully utilizing the results of thermal-neutron-capture damage experiments. We have calculated the primary recoil spectra for thermal-neutron capture in copper, molybdenum, vanadium, and tungsten and have derived the damage-energy cross sections needed to determine the defect-production efficiencies from the measured resistivity damage rates.

In principle, it should be possible to calculate an exact thermal-neutron-capture recoil-energy distribution from knowledge of all nuclear energy levels, their corresponding spins and parities, and their populations after neutron

Figure 49. Comparison of calculated and experimental neutron-capture gamma-ray spectra in copper and tungsten.

Table 17. Results of thermal-neutron-capture recoil calculations.

Metal	Multiplicity		Median recoil energy (eV)	Damage-energy cross section (barns $\times$ keV)
	Experiment	Calculated		
Copper	1.98	2.18	480.0	1.276
Molybdenum	3.57	3.70	240.0	0.410
Vanadium	1.81	2.03	480.0	1.671
Tungsten	2.56	2.83	75.0-80.0	0.766

Figure 50 Integral damage energies in natural copper from electron, thermal-neutron, and reactor-neutron irradiations

capture The momentum transferred to the nucleus would be the vector sum of the momenta from the individual gamma rays in the cascade (neglecting the insignificant momentum transferred by the 0.025-eV neutron). Unfortunately, in most metals a high percentage of the primary gamma rays undergo transitions to continuum states. These transitions cannot be placed in level diagrams and therefore cannot be accounted for in recoil calculations that use the above method.

We have developed a model to calculate thermal neutron recoil energies which accounts for continuum transitions.⁴ In this model, the nuclear level diagram for the recoil nucleus of interest is divided into the measured discrete levels at low excitation energies and 250 keV 'bins' within the continuum. Experimental values for the transition probabilities are used for transitions from the capture state to the low-energy discrete levels as well as for the transitions between the discrete levels. For transitions into and out of the continuum, calculated transition probabilities based upon a statistical model are used.⁵

Table 18. Damage efficiencies from resistivity-damage-rate experiments on thermal-neutron-irradiated metals.

Metal	Resistivity damage rate ⁸⁻¹¹ ($10^{-31} \Omega \cdot m^3$)	Efficiency
Copper	0.019	0.43
Molybdenum	0.039	0.95
Vanadium	0.025	0.64
Tungsten	0.083	1.30

There are two experimental observations with which we can compare our results. The first is the experimental gamma-ray spectrum, and the second is the gamma-ray multiplicity. The gamma-ray spectra obtained from our statistical calculations for neutron capture in natural copper and tungsten are compared with the observed gamma-ray spectra of Orphan *et al.*⁶ in Fig. 49. Table 17 gives the experimental and calculated multiplicities for each of the metals we have studied. The calculated values appear to be within the experimental uncertainties in every case. This gives us confidence that the recoil energies obtained from the statistical model are accurate.

In Table 17 we have listed the results of the thermal neutron recoil calculations. Figure 50 shows the integral recoil spectrum from thermal-neutron capture in natural copper. Also shown in this figure are the integral recoil spectra from 1 MeV electron irradiations⁷ and reactor-neutron irradiations⁸ of the same metal. As we can see, the thermal neutron recoil spectrum is narrowly distributed in an intermediate energy region.

We used the calculated damage energy cross sections to determine the efficiency of thermal neutrons in producing damage. These efficiencies and the corresponding resistivity damage rates are listed in Table 18. The use of our recoil calculations greatly improves the agreement between previous thermal neutron irradiation

experiments and the results of electron irradiations and fully dynamic computer simulations. This is shown in Fig. 51 for molybdenum. In this figure, we have plotted the experimental efficiencies obtained from electron and high-energy-neutron irradiations as well as a previous efficiency for thermal-neutron irradiations determined with an old value of the thermal-neutron damage-energy cross section. We can see that the efficiency determined with our more accurate recoil calculation greatly improves the agreement with the other data as well as with our computer simulations. Improvement is observed for all of the metals studied so far.

This research was supported by the Materials and Radiation Effects Branch, DOE Office of Fusion Energy.

Figure 51. Damage efficiencies in natural molybdenum measured from electron, thermal-neutron, and high-energy-neutron irradiations. The previous value of the efficiency for thermal neutrons is given, and we have compared this data with the results of fully dynamic computer simulations.

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Fusion-Neutron-Induced Changes in Copper Magnetoresistivity

M. W. Guinan and R. A. Van Konynenburg

We measured the changes that occur in the magnetoresistivity of coppers (having various purities and pretreatments) at magnetic fields up to 12 T during the course of sequential fusion-neutron irradiations at about 4 K and anneals to room temperature. In conjunction with work in progress by Coltman and Klabunde of Oak Ridge National Laboratory (ORNL), the results should lead to engineering design data for the stabilizers of superconducting magnets in fusion reactors. These magnets are expected to be irradiated during reactor operation and warmed to room temperature periodically during maintenance.

Eight copper wires were repeatedly irradiated at 4.2–4.4 K with 14.8-MeV neutrons and isochronally annealed at temperatures up to 34°C for a total of five cycles. Their electrical resistances were monitored during irradiation under zero applied magnetic field. After each irradiation the magnetoresistances (the increases in resistance due to a magnetic field) were measured in applied transverse magnetic fields of up to 12 T. Then the samples were isochronally annealed to observe the recovery of the resistivity and magnetoresistivity. After each anneal at the highest temperature (34°C), some of the damage remained and contributed to the

damage state observed following the subsequent irradiation. In this way, we were able to observe how the changes in magnetoresistance would accumulate during the repeated irradiations and anneals expected to be characteristic of fusion-reactor magnets.

The copper materials used in this work were obtained from C. Klabunde of ORNL (Oak Ridge, TN) and are the same materials that R. Coltman and Klabunde have used for irradiations in the Bulk Shielding Reactor at ORNL and will use in the Intense Pulsed-Neutron Source at Argonne National Laboratory (Argonne, IL). The materials included both

Table 19. Descriptions of the unoxidized OFHC copper^a samples.

	Sample		
	CUUN0	CUUN1	CUUN2
Treatment	Recrystallized	7.5% cold-worked	14.3% cold-worked
Diameter (mm)	0.1259	0.1261	0.1261
Gauge length (mm)	11.0	11.1	10.1
Resistivity at 4.2 K ($10^{-11} \Omega \cdot m$) ^b	4.74	11.27	19.10
Ratio of resistance at 293 K to resistance at 4.2 K	363	153	91

^a OFHC copper, Intermagnetics General Corporation CDA-101.

^b Corrected for size effect.

Table 20. Pre-irradiation resistivities ($10^{-11} \Omega \cdot m \pm 0.05$)^a as a function of field.

Field (T ± 0.005)	Sample		
	CUUN0	CUUN1	CUUN2
0	4.99	11.14	17.04
1.816	12.39	17.54	23.31
3.827	22.43	26.84	31.86
5.791	31.96	36.51	40.93
7.694	40.50	45.98	50.08
9.767	49.39	56.12	60.13
11.783	57.60	65.83	69.83

^a Corrected for size effect.

oxygen-free high-conductivity (OFHC) copper and high-purity copper having different treatments in internal oxidation, cold work, and annealing.

In this report we will consider only the results for unoxidized OFHC copper, since these three samples are most typical of stabilizers in present use in large magnets. A complete set of results will be reported later. Table 19 summarizes the size and resistance data provided by Klabunde¹ for the OFHC copper used in this work.

Pre-irradiation values of the resistivity as a function of field for these samples are given in Table 20. The field and resistivity values shown are averages of measurements made in stepping the field up to and down from 11.783 T.

Irradiation was performed using the Rotating Target Neutron Source (RTNS-II) with 0.23-m-diam targets. The machine produced an average flux of 1.2×10^{16} n/m²·s on the samples. The neutrons were nearly monoenergetic at 14.8 MeV.

Sample CUUN1 was used as a monitor during the irradiation-anneal cycles with the objective of reaching the same resistivity at 12 T at the conclusion of each irradiation. A raw data plot of sample resistance at zero field vs proton recoil counts is shown in Fig. 52. The number of recoil counts is approximately proportional to the neutron fluence on the sample.

It is evident from Fig. 52 that a higher percentage of damage recovers in later cycles than in the first cycle. This is illustrated in Fig. 53 for all three OFHC samples. Here we have plotted the percentage of the accumulated resistivity change retained after 30-min anneals at 307 K as a function of the total resistivity change produced at 4.2 K. For all three samples the percentage of retained damage

Figure 52. Resistance of sample CUUN1 (7.5% cold-worked) as a function of neutron fluence (proton recoil counts) during five cycles of 14.8-MeV neutron irradiation at 4.2 K. The sample was annealed at 307 K between each irradiation cycle.

Figure 53. Percentage of the total accumulated resistivity change at 4.2 K retained after annealing at 307 K, as a function of the total resistivity change accumulated at 4.2 K during neutron irradiation.

Table 21. Resistivities ($10^{-11} \Omega \cdot m$) after the first and fifth irradiation-anneal cycles.

	Sample		
	CUUN0	CUUN1	CUUN2
Before irradiation			
At 0 ± 0.01 T	5.01	11.14	17.03
At 10.00 ± 0.01 T	50.34	57.23	61.25
After Cycle 1 irradiation			
At 0 ± 0.01 T	35.33	42.49	48.05
At 10.00 ± 0.01 T	76.43	81.26	85.51
After Cycle 1 anneal			
At 0 ± 0.01 T	11.01	16.55	21.59
At 10.00 ± 0.01 T	58.24	61.67	65.23
After Cycle 5 anneal			
At 0 ± 0.01 T	20.20	25.35	29.55
At 10.00 ± 0.01 T	65.49	69.08	72.41

decreases with increasing damage. As cold work is increased the damage retained decreases significantly.

For comparison, we have also plotted the results of six other annealing studies following low-temperature neutron irradiation. The three data points at low resistivities are from Takamura *et al.*,² and they also illustrate the effects of increasing amounts of cold work. The three points at the resistivities comparable to the maximum accumulated values attained here, in order of increasing damage, are from Burger *et al.*,³ Horak and Blewitt,⁴ and Brown *et al.*⁵

The approximate agreement seen in Fig. 53 between our present results and those of earlier studies at resistivities comparable to our total accumulated resistivity is encouraging, because it indicates that the percent retained is relatively insensitive to the detailed annealing actually employed after one or two of our cycles.

The magnetoresistivities measured as a function of field before irradiation are tabulated in Table 21. We interpolated the measurements on the three unoxidized OFHC samples to produce magnetoresistivity values for them at 10.00 ± 0.01 T in the following states: before irradiation, after the first irradiation, after the first irradiation-anneal cycle, and after the fifth irradiation-anneal cycle. These values, shown in Table 21, span the full range of values of zero-field resistivity encountered with these samples.

Resistivity changes as a function of field, H , are usually presented in the form of a Kohler

plot⁶ in which the fractional change in resistivity at field is plotted as a function of field divided by initial (zero-field) resistivity, $\rho(0)$:

$$\frac{\rho(H) - \rho(0)}{\rho(0)} = f\left[\frac{H}{\rho(0)}\right]$$

Our results for the variation of zero-field resistivity by cold work, irradiation, and repeated irradiation and anneal cycles all fall on a single Kohler line to within $\pm 2\%$. This plot is compared with that obtained from the pre-irradiation variation with field in Fig. 54.

Here the points are again all within $\pm 2\%$ of a single line covering 1-1/2 orders of magnitude. For comparison, the Kohler plot obtained by Fickett⁷ for pure copper samples when purity, temperature, and field were varied is also shown. At high values of $H/\rho(0)$ the magnetoresistances are comparable, but at low values, corresponding to increasing $\rho(0)$ at constant field, the curves diverge.

From a magnet-designer's point of view the quantity of interest is the resistivity at field. The data of Tables 20 and 21 are, therefore, recast in Fig. 55, in which the resistivity at 10 T is plotted as a function of the resistivity at zero field.

For evaluating stabilizer materials for superconducting magnets, the results given above are significant in two respects: (1) A Kohler plot determined on a conductor solely by varying the field can also be used to describe the variation of the magnetoresistance as a result of

cold work, high-energy-neutron irradiation, and subsequent annealing. This greatly simplifies the task of predicting the response of the magnetoresistance of actual conductors to changes resulting from fabrication of magnets and use in fusion reactors. (2) The fraction of accumulated damage remaining at 4.2 K after several irradiation-anneal cycles is nearly a factor of two lower than that previously estimated from studies of the annealing of lower-dose neutron damage.⁸ This result alone significantly increases lifetime estimates for magnets in conceptual fusion-reactor designs.

From consideration of these data and a preliminary design of a liquid-helium-cooled

magnet system (1.8 K), it appears that the magnetoresistivity of the stabilizer will not be the determining factor for shielding requirements in systems of this type that utilize polyimide thermal insulation.

Upon completion of planned experiments at the Intense Pulsed-Neutron Source by Coltman and Klabunde of ORNL using the same sample materials as used here, it will be possible, from all the data, to make accurate predictions of the magnetoresistive behavior of copper stabilizers under expected fusion-reactor conditions.

This research was supported by the Materials and Radiation Effects Branch, DOE Office of Fusion Energy.

Figure 54. Kohler plot for unoxidized OFHC copper. Solid symbols show pre-irradiation values (from Table 20); open symbols show measurements at 10 T after irradiation and annealing (from Table 21).

Figure 55. Resistivity at 10 T as a function of resistivity at zero field for unoxidized OFHC copper. Solid symbols are values deduced from the Kohler plot of Fig. 54; open symbols are measurements taken at 10 T.

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Supporting Fundamental Science

6

6 Supporting Fundamental Science

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Overview

The Nuclear Chemistry Division devotes a significant portion of its discretionary resources to basic research. We believe that the long-term health of our Division and our ability to find new approaches to weapons diagnostics depend on having small but high-quality research efforts in areas relevant to our programmatic missions.

Some of our research is directed to solving basic problems that will improve our understanding of diagnostic results, but more often the ultimate impact of the research is less well defined. This research frequently pushes our instrumentation to greater levels of sensitivity or sophistication, and new instruments or techniques often find immediate programmatic applications. By engaging in high-quality basic studies, we attract stimulating collaborators whose ideas extend far beyond a particular research project. By maintaining close contacts with many members of the international scientific community, we enhance the reputation of the Division and the Laboratory. This in turn makes it easier to attract quality people to our staff.

Most of the support for long-range basic studies comes from the weapons programs. However, we do receive additional support from DOE's Office of High-Energy and Nuclear Physics. Using such funds, we support research in nuclear chemistry and physics, inorganic chemistry, cosmochemistry and geochemistry.

This year, we report on research activities in cosmochemistry, heavy-element and fission studies, and nuclear-structure research.

The Division's activities in cosmochemical studies are only three years old, yet this year we report significant progress toward using the decay of ^{187}Re as a cosmological chronometer—both for dating the age of the universe and for dating more recent events in the solar system. We have developed quantitative techniques for determining the mass of nanogram quantities of osmium using both laser microprobe mass analysis and inductively coupled plasma atomic-emission spectroscopy.

We describe our continued research on the identification and interpretation of isotopic anomalies. This year we report our first results on

the production and mechanism of loss of noble gases in meteorites. These are obtained by the production of the gases *in situ* in a variety of naturally occurring minerals and pure chemical compounds by several nuclear reactions with 14-MeV neutrons.

We present significant new results in our attempt to use the decay of ^{163}Ho to determine the mass of the neutrino. We have also designed a spectrometer and source capable of setting an upper limit of 5 eV on the mass of the neutrino through a precise measurement of the beta-decay end point of tritium. A non-zero mass for the neutrino would not only have profound implications for fundamental-particle physics but also for the so-called missing-mass problem in the universe and for models of solar behavior.

In the area of heavy-element research, we have traditionally studied the nuclear and chemical properties of nuclei having $Z > 94$. In recent years, we have studied the mechanism of heavy-ion reactions on actinide targets and the spontaneous fission properties of the heaviest isotopes in order to design experiments to search for superheavy elements. This year we report on the use of ^{254}Es as a target material and on new calculations of decay properties of isotopes of the superheavy elements. We also performed a number of accelerator-based experiments that determined the chemical reduction potential of nobelium.

Nuclear-structure research is another established program. This year, we report some of the first results using our new superconducting-solenoid electron spectrometer. In a particularly significant experiment, we observed the decay from the second well to the ground state in ^{238}U in competition with a large fission background. We give the results from a new series of experiments designed to measure gamma rays and electrons in coincidence with protons following triton-induced reactions. We also report the remarkable success of our unified model using parameters taken from odd-mass nuclei in making detailed predictions of the low-energy properties of odd-odd actinide nuclei.

Precise Determination of the Lifetime of ^{187}Re —A Progress Report

M. Lindner, G. P. Russ, R. J. Borg, J. M. Bazan and D. Simons¹

In our continuing project to obtain a precise value for the lifetime of ^{187}Re by measuring the rate of growth of the daughter ^{187}Os atoms, we separated, isolated, and stored 13 osmium fractions from 5 separate purified sources of perrhenic acid. We continued to search for a means of concentrating 1 to 2 ng of osmium onto suitable targets for the laser microprobe mass analyzer (LAMMA), and greatly improved our ability to measure isotopic ratios with 1- to 2-ng samples of osmium on LAMMA.

The isotope ^{187}Re , with an approximate half-life of 4×10^{10} y, is considered to be the best nuclear cosmochronometer for determining the ages of metallic phases of meteorites. Ultimately, such determinations make possible a measurement of the galactic age. Hence, an accurate half-life for ^{187}Re is of considerable cosmological interest.

The decay of ^{187}Re to ^{187}Os involves a maximum beta-decay energy of 2.6 keV. The difficulties of absolute beta-intensity measurements at this energy are so great that we chose to determine the lifetime by periodic removal and measurement of the daughter ^{187}Os atoms from massive quantities (100 g to 1 kg) of rhenium, from which we had removed any osmium that might have initially been present.

We reported previously² the purification of five 1-kg batches of perrhenic acid as sources for radiogenic ^{187}Os growth. In that report we listed preliminary results of osmium isotopic ratio measurements on the laser microprobe mass analyzer (LAMMA) in a collaboration with the National Bureau of Standards. Since that report, we have divided our efforts nearly equally among the following:

1. Removal and isolation of radiogenic ^{187}Os from purified perrhenic acid.
2. A search for a method of concentrating 1 to 2 ng of osmium onto a small target suitable for LAMMA measurements.
3. A study of instrumental parameters that affect LAMMA isotopic ratio measurements on

targets containing 1 to 2 ng of osmium.

During the past year we removed two aliquots ($\sim 10\%$ each), at about 6-month intervals, from each of five 1-kg sources of purified perrhenic acid. Each source had been initially "inoculated" with an accurately known quantity of either ^{190}Os or ^{192}Os at the onset of growth. From these aliquots we separated and isolated the radiogenic ^{187}Os (together with the ^{190}Os or ^{192}Os). These isolated osmium samples are being stored until a satisfactory method can be found for concentrating the expected 1 to 2 ng of osmium onto the required substrate, designed for targets in the LAMMA.

We conducted a long search for some method of concentrating 1 to 2 ng of osmium from a large volume of solution (~ 50 ml) into a clean, massless spot. In particular, we investigated the use of anion-exchange resins (both strong-base and weak-base types), because the technique frequently provides a simple, convenient way of concentrating ionic species in high purity and good yield. We found no anion resin which fulfilled these requirements for osmium. The trapping of OsO_4 tracer vapor in cryogenically cooled organic reductants was also investigated, but the method created more problems than it solved.

A promising approach appears to be cryogenic trapping of the tetroxide tracer vapor by itself, followed by room-temperature transport of the vapor with a carrier gas (e.g., O_2) to a small chemical trap of very pure hydrochloric acid.

Initial experiments were mostly unsuccessful, but we attribute the failure to equipment design rather than to the concept. We have now redesigned our apparatus to test the method again.

We spent considerable effort in improving our ability to accurately determine isotopic ratios on LAMMA. Englert and Herpers³ and Simons⁴ have reported on osmium isotopic measurements with LAMMA. Simons⁴ showed that the major limitation to accuracy was a loss of gain in the electron-multiplier output with an increase in the input ion-pulse intensity. A possible solution to this problem was a correction factor based on a comparison of the known isotopic ratios of natural osmium with the intensity ratios as measured on LAMMA. Our own previous determination of osmium isotopic ratios² was based on this method. However, the risk in the use of somewhat arbitrary correction factors is better avoided, if possible. Furthermore, the situation was aggravated by apparent anomalies at masses 187 and 189. Thus, the potential presence of interfering ions (e.g., K_3Cl_2^+ and Cu_3^+) opened to question the overall feasibility of using LAMMA to measure ^{187}Os . We therefore considered it important to prepare new isotopic mixtures using ultrapure reagents to avoid mass contamination as much as possible.

We carefully prepared six solutions of different mixtures of ^{190}Os and ^{192}Os standards, using gravimetric (rather than volumetric) techniques, and reagents prepared from ultrapure water and hydrochloric acid. The ultimate concentrations were approximately 2 ng of osmium per microliter. These quantities were evaporated onto very thin (~ 10 nm) silicon monoxide films supported on gold electron microscope ("e.m.") grids. Microscopic examination showed almost no residues; occasional small reddish crystals, presumably of osmium chloride, could be seen. These were the richest sources of osmium ions in LAMMA. Figure 56 shows a micrograph of such a crystal. Its weight was estimated as 1 ng, and it provided an osmium source for many laser pulses, and thus, mass spectra. Figure 57 shows a typical spectrum for a 3:1 mixture of ^{190}Os and ^{192}Os . The data were recorded with a Tektronics 7612 200-MHz fast recorder. The absence of statistically significant events at mass 187 is noted.

Since the efficiency of a given laser pulse in producing a burst of osmium ions cannot be controlled, a distribution of intensities (of all isotopes) is produced in a series of successive

Figure 56. Crystal of osmium chloride (~ 1 ng) on silicon monoxide film and gold mesh. Lattice dimensions are about $150\ \mu\text{m}$.

Figure 57. Typical mass spectrum of a 3:1 mixture of ^{190}Os and ^{192}Os . No significant events occur at mass-187. The small peak at mass-197 is attributed to the gold grid.

pulses. This is illustrated in Fig. 58 for about 50 bursts recorded for our 3:1 mixture of ^{190}Os and ^{192}Os . The loss of gain for the more abundant isotope (abscissa) at high intensities shows up as a nonlinearity in the data. It is clear that no single data point can yield the proper isotopic ratio for this typical mixture, especially at the higher intensities (where the statistics should be the most favorable).

We determined the "observed" isotopic ratio by a least-squares fit to the quadratic expression

$$I_{192} = AI_{190} + B(I_{190})^2, \quad (1)$$

where I_{192} is the intensity of the mass-192 peak, and A and B are the coefficients to be solved for.

The parameter A is the "best-fit" isotopic ratio. Equivalently (but not identically) we also determined the isotopic ratio from a least-squares fit of the "differential" form of the above expression:

$$\frac{I_{192}}{I_{190}} = A + BI_{190} \quad (2)$$

In this case, A is the intercept of the "best-fit" straight line of a plot of the intensity ratio vs the intensity I_{190} .

Table 22 lists the isotopic ratios found by least-squares fits to both data forms. Fitting to

Eq (1) was made with the program LLFIT of Gunnink and Niday⁵; fitting to Eq. (2) was made independently with the program of Hudson.⁶ Results are listed for natural osmium and for five synthesized mixtures of osmium isotopes. The agreement is generally good, but not yet good enough. In the future we intend to improve our precision in several ways:

1. By increasing the number of data points to at least 100.

2. By improving our estimate of the uncertainties associated with each variable. This is one of our current problems. The assignment of rather arbitrary and unrealistic values to the

Table 22. Isotopic ratios determined from least-squares fit of LAMMA data.

Osmium isotopic mixture	Ratio measured	Number of data points	Method ^a	LAMMA value	Std dev (%)	Accepted value	LAMMA value / Accepted value
Natural	186/192	26	II	0.021	4.1	0.039	0.54
		26	II	0.029	3.5	0.039	0.74
			RI	0.039	—	0.039	1.00
	188/192	26	II	0.318	1.5	0.324	0.98
		25	RI	0.318	—	0.324	0.98
	189/192	26	II	0.408	1.5	0.393	1.04
		25	RI	0.404	—	0.393	1.03
	190/192	26	II	0.629	1.5	0.644	0.98
		25	RI	0.628	—	0.644	0.98
A	190/192	29	II	3.130	2.1	3.282	0.95
		22	II	3.291	1.5	3.282	1.00
		47	RI	3.039	—	3.282	0.93
B	190/192	40	II	9.333	1.7	9.274	1.01
		15	II	10.174	2.5	9.274	1.10
		54	RI	8.75	—	9.274	0.94
C	190/192	31	II	0.3764	1.5	0.3814	0.99
		29	II	0.3834	2.3	0.3814	1.01
		57	RI	0.373	—	0.3814	0.98
D	190/192	19	II	1.107	1.4	1.113	0.99
		22	II	1.027	1.4	1.113	0.92
		41	RI	1.037	—	1.113	0.93
E	190/192	33	II	5.099	1.5	5.327	0.96
		25	II	5.009	1.6	5.327	0.94
		55	RI	4.65	—	5.327	0.87

^a II is isotope vs isotope. RI is isotopic ratio vs isotope.

Figure 58. Intensity correlation for a 3:1 mixture of ^{190}Os and ^{192}Os , showing the nonlinearity at the upper end. Units are the fast-recorder output.

uncertainties is probably responsible for the poor agreement found for the low-abundance isotopes ^{186}Os and ^{187}Os in the fitting process.

3. By seeking new and improved algorithms, which would result from a better understanding of the causes of the nonlinear response of the detector. This approach is already being tested.

4. By possible improvements in the detector itself to improve linearity of response.

Finally, we compared the performances of the 100-MHz Biomation fast recorder and the 200-MHz Tektronics 7612 instrument by splitting the output of the electron multiplier. In the calculation of isotopic ratios derived from the two recorders, there seems to be a small systematic difference. There is, in addition, a consistently smaller standard deviation of a set of data points taken on the Tektronics instrument than for the same set of data recorded on the Biomation recorder.

This research was supported by the DOE Office of Basic Energy Sciences.

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Investigation of Optimal Parameters for the Detection of Osmium by Inductively Coupled Plasma Spectrometry

J. M. Bazan, A. L. Langhorst, Jr.,¹ and R. J. Borg

We used inductively coupled plasma (ICP) atomic-emission spectrometry to study the formation of the volatile species of osmium from solutions of different oxidizing agents. Osmium tetroxide (OsO_4) showed an enhancement of the ICP signal intensity by a factor of 10 to 300 over nonvolatile forms of osmium. Results were dependent on the osmium solution matrix, oxidizing agent, and mode of transportation of the volatile species to the ICP torch. We studied the stability of the OsO_4 signal intensity for use as a calibration standard for ICP analysis.

The objective of our study is to use inductively coupled plasma (ICP) spectrometry for the determination of osmium, concurrently with laser microprobe mass analysis (LAMMA). We are developing these methods for two programs: (1) terrestrial rhenium-osmium age dating, and (2) the development of back-up techniques for the half-life determination of ^{187}Os . Our ICP research was conducted on a Jarrell-Ash Model 975 ICP spectrometer with the volatile form of osmium (OsO_4) instead of the soluble form (presumably a halide complex). This technique had several advantages: (1) increased sensitivity for the detection of osmium by a factor of 10^2 , (2) removal of spectral interferences caused by nonvolatile elements, and (3) simplified sample preparation for the detection instrument.

Previously, the reported osmium detection limits obtained by ICP atomic-emission spectroscopy varied from about 1 to 0.36 ppm.²⁻⁴ These values indicated unexplained chemical phenomena. Summerhays *et. al.*⁵ observed that the signal intensity of osmium was enhanced when the solution matrix was 9N HNO_3 . They theorized that the osmium was being oxidized to the tetroxide (OsO_4), and that the volatile species was responsible for the increase in signal intensity. They determined that their intensity readings could not be reproduced over time

intervals of several days. We have improved their technique and identified a stable osmium solution suitable for producing the volatile tetroxide for ICP calibration.

Normally our instrument uses a cross-flow nebulizer to introduce samples, in the same manner as all others reported in the literature,⁵ but the nebulizer allows only 5% of the amount of solution sampled to be delivered to the ICP ionizing torch for analysis. Our new technique replaces the cross-flow nebulizer with a separation chamber preceded by a heated on-line mixture of oxidant and stabilized sample solution (Fig. 59). The argon transport gas carries the OsO_4 vapor to the ionizing plasma torch. In this way, the method is specific for osmium in the volatile state, since the preparation chamber eliminates the nonvolatile elements which could cause spectral interferences. To eliminate condensation of water vapor, which might impede the flow of OsO_4 into the ionizing torch, we have also built a new water-jacketed separation chamber. The effectiveness of this chamber has not yet been tested.

In searching for a system that would maximize the signal intensity of OsO_4 , we investigated four parameters: (1) the analytic matrix, (2) solution stability over time, (3) effect of oxidants, and (4) temperature dependence.

Figure 59. Schematic of OsO_4 separation system.

Using a Spex Industries, Inc., standardized osmium solution of $1000 \mu\text{g/ml}$ in 3.2N HCl , we prepared the following dilutions containing an osmium concentration of $9.3 \mu\text{g/ml}$: (1) 3.2N HCl , (2) $3\text{N H}_2\text{SO}_4$, (3) $6\text{N H}_2\text{SO}_4$, and (4) $1\text{N NH}_4\text{OH}$. Figure 60 shows a study of the enhanced OsO_4 signal as a function of time, produced in the various acid solutions with periodic acid (HIO_4) added to each. Figure 61 displays a time study showing the stability of the OsO_4 produced from an NH_4OH solution. We found an increase in the signal from the acid solutions over a period of 30 to 90 d, when the OsO_4 is produced. The OsO_4 produced from the $1\text{N NH}_4\text{OH}$ solution, however, appeared to yield a relatively constant signal over the same period of time. We prepared three separate $1\text{N NH}_4\text{OH}$ solutions on different days, to verify their stability for the production of OsO_4 with an oxidant over the time interval shown in Fig. 61. The solution prepared on 4/26/83 had an

osmium concentration of $9.3 \mu\text{g/ml}$ while the solutions prepared on 4/27/83 and 4/28/83 had a concentration of $10 \mu\text{g/ml}$. After the initial "growth" period of about 1 h, and within the limitations of the ICP instrument, the $1\text{N NH}_4\text{OH}$ solutions appeared to yield a satisfactorily stable signal intensity.

We tested seven oxidizing agents: solutions of $10\% \text{HIO}_4$, $10\% \text{KMnO}_4$, $10\% \text{K}_2\text{Cr}_2\text{O}_7$, $30\% \text{H}_2\text{O}_2$, concentrated HNO_3 , and fuming HNO_3 . Signal intensities and ease of handling were both used as criteria in determining the most suitable oxidizing agent. Varying the oxidants but maintaining the same osmium-solution matrix resulted in signal intensities that were comparable. Varying the osmium-solution matrix but maintaining the same oxidant resulted in distinctly different maximum signal intensities for the OsO_4 produced in our separation chamber. Figures 60 and 61 show these signal intensity values when a 10% periodic acid

solution was used as the oxidizing agent for all the osmium-solution matrices studied. Periodic acid was selected as the oxidizing medium for its ease of handling and long shelf life.

Figure 62 shows that the signal intensity was directly proportional to the temperature of the mixed reactants. This was true for all the osmium solutions and oxidants studied. The figure shows that a maximum signal intensity for OsO_4 was approached at 105°C .

Future research will include: (1) maximizing the signal intensity with temperature and flow rate, (2) comparing the stability of an alkaline solution with that of an ammoniacal solution, and (3) determining the detection limits for concentrations of osmium at optimum conditions.

This research was supported by the LLNL Nuclear Chemistry Research Program.

Figure 60. Time study of volatile OsO_4 production from acid solutions of osmium, using 10% HIO_4 at 105°C .

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Figure 61. Stability study of osmium in 1N NH_4OH and 10% HIO_4 at 105°C.

Figure 62. Signal intensity as a function of temperature, for osmium in 1N NH_4OH and 10% HIO_4 .

Production of Noble Gases by 14-MeV Neutrons

D. A. Leigh, R. J. Borg, and V. B. Lanier

We started an investigation into the production and mechanisms of loss of noble gases from meteorites. Neutrons with an energy of 14.2 MeV, generated by the LLNL Rotating Target Neutron Source facility (RTNS-II), were used to induce nuclear reactions in a selection of naturally occurring minerals and pure chemical compounds. The amounts and isotopic ratios of the resulting noble gases were determined by mass spectrometry. Measurements of outgassing rates are in progress to determine specific diffusion coefficients.

The aim of the research described here is to provide a better understanding of the chemical and physical factors that govern the loss of noble gases from meteorites. Although there has been abundant investigation of noble gases in meteoritic materials, with special emphasis on variations in isotopic composition, there have been virtually no investigations dealing directly

with the solid-state aspects of gas retention. It is clear that isotopic fractionation can be caused by differing recoil and trapping energies for different isotopes. It is also clear that diffusive loss is a function of mineralogy, temperature, and host particle size.

The *modus operandi* of our program is the measurement of the rates of outgassing in a suite

Table 23. Production of neon by 14.2-MeV neutrons.

Reaction	Q (MeV)	σ_{14} (mb)	
		Literature	This work ^a
(1) $^{23}\text{Na}(n,2n)^{22}\text{Na}(\beta^+)^{22}_{26a}\text{Ne}$	-12.42	25	304
(2) $^{23}\text{Na}(n,np)^{22}\text{Ne}$	-8.79	60	
(3) $^{23}\text{Na}(n,d)^{22}\text{Ne}$	-6.57		
(4) $^{23}\text{Na}(n,t)^{21}\text{Ne}$	-10.68		
(5) $^{23}\text{Na}(n,\alpha)^{20}\text{F}(\beta^-)^{20}_{11s}\text{Ne}$	-3.87	160	137
(6) $^{24}\text{Mg}(n,^3\text{He})^{22}\text{Ne}$	-12.76	130	$\leq 130^a$
(7) $^{25}\text{Mg}(n,\alpha)^{22}\text{Ne}$	0.48		
(8) $^{26}\text{Mg}(n,\alpha)^{22}\text{Ne}$	-10.61		
(9) $^{24}\text{Mg}(n,\alpha)^{21}\text{Ne}$	-2.55	200	172 ^a
(10) $^{25}\text{Mg}(n,\alpha)^{21}\text{Ne}$	-9.88		
(11) $^{24}\text{Mg}(n,\alpha)^{20}\text{Ne}$	-9.31		≤ 921

^a $^{21,22}\text{Ne}$ production measurements actually measure weighted sums of σ 's:

$$\sigma_9 + ({}^{25}\text{Mg}/{}^{24}\text{Mg})\sigma_{10} = \sigma_9 + 0.1266\sigma_{10} = 172 \text{ mb} ,$$

$$\sigma_7 + ({}^{24}\text{Mg}/{}^{25}\text{Mg})\sigma_6 + ({}^{26}\text{Mg}/{}^{25}\text{Mg})\sigma_8 = \sigma_7 + 7.899\sigma_6 + 1.101\sigma_8 ,$$

$$26 \text{ mb} \leq (\sigma_7 + 7.899\sigma_6 + 1.101\sigma_8) \leq 130 \text{ mb} .$$

of representative minerals as a function of temperature, for noble gases produced by different reaction mechanisms. For example, neon isotopes can be made from sodium and magnesium, using 14.2-MeV neutrons, by the reactions listed in Table 23.

In sodalite, $\text{Na}_4(\text{AlSiO}_4)_3\text{Cl}$, reactions 1–5 on the major element sodium dominate, but if significant magnesium is present as an impurity or in adjacent mineral grains, neon isotopes can also be made *in situ* or recoil-injected by reactions 6–11. Reactions 2, 3, 7, and 8 are alike in that ^{22}Ne is the initial product of the reaction, but reaction 1 produces a long-lived intermediate, ^{22}Na . Furthermore, sodium is a major constituent of sodalite. Annealing after irradiation should drive some if not most of the ^{22}Na back into its equilibrium position. After an interval to allow some ^{22}Na to decay, outgassing measurements should in principle reveal the differences between ^{22}Ne trapped at normal sodium sites and that retained in interstitial positions.

It is necessary to determine the relative isotopic composition of the noble gases in each mineral after irradiation prior to investigating the possibility of isotopic fractionation. Furthermore, because the relative cross sections for the production of noble gas isotopes are poorly known, in our experiments we irradiated and analyzed a suite of pure reagents as well as a separate suite of natural minerals. Table 24 lists the samples. Two sodalite specimens (labeled “Jacapuranga” and “African”), each with a

somewhat different trace-element content, were included in the irradiation.

All mineral specimens consisted of powders with an average particle diameter of $50 \pm 10 \mu\text{m}$, attained by grinding followed by differential screening. To remove excess water and trapped gases, the compounds were melted before encapsulation. These powders were sealed under vacuum ($\sim 10^{-7}$ torr) in quartz ampoules, and irradiated at ambient temperature with 14.2-MeV neutrons produced by the LLNL Rotating Target Neutron Source facility (RTNS-II). Dosimetry was performed by monitoring the fluence with natural iron foils wrapped around each ampoule. We counted the decay by electron capture of ^{54}Mn produced by $^{54}\text{Fe}(n,p)^{54}\text{Mn}$. The compounds and magnesium metal were treated in the same manner, with the omission of the grinding and sizing.

After irradiation, each of the two sodalite specimens was divided into two aliquots. An aliquot from each specimen was annealed at 600°C for 6 h in an attempt to drive the ^{22}Na back onto its rational lattice site. Specimens of each were sent to Prof. Till Kirsten of the Max Planck Institute (Heidelberg, FRG) for diffusion measurements, which are now under way.

We analyzed the noble gases released by each of the irradiated samples in a single 1650°C vacuum extraction. Aliquots of irradiated mineral powders and chips of irradiated reagents were weighed and wrapped in aluminum foil, then loaded into a 10-sample wheel attached to the gas-extraction system. After evacuation and

Table 24. Isotopic composition of noble gases in samples irradiated with 14.2-MeV neutrons.

Sample name	Chemical formula	Fluence (10^{16} n/cm ²)	Analysis weight (mg)	Major isotope	Concentration (10^{12} atoms/g)	Isotopic ratios	
Jacapuranga sodalite	$\text{Na}_4(\text{AlSiO}_4)_3\text{Cl}$	7.28 (annealed)	25.0 42.7	^{22}Ne ^{22}Ne	82 0.94	$^{21}\text{Ne}/^{22}\text{Ne} = 0.0169$ 0.091	$^{20}\text{Ne}/^{22}\text{Ne} = 0.460$ <1
African sodalite	$\text{Na}_4(\text{AlSiO}_4)_3\text{Cl}$	9.28 (annealed)	46.3 56.7	^{22}Ne ^{22}Ne	102 1.7	0.0177 0.0047	0.444 0.32
Albite	$\text{NaAlSi}_3\text{O}_8$	9.23	8.2	^{22}Ne	27	0.0174	0.454
Forsterite	Mg_2SiO_4	8.90	16.4	^{21}Ne	78	11.3	6.76
Estatite	MgSiO_3	10.47	23.0	^{21}Ne	65	11.0	6.77
Mg metal	Mg	0.824	48.3, 51.4	^{21}Ne	27.8	10.5	7.81
Salt	NaCl	0.884	91.3, 138.9	^{22}Ne	27.7	0.0169	0.43
Salt	KCl	0.808	197.0, 201.4	^{38}Ar	39.2	$^{39}\text{Ar}/^{38}\text{Ar} = 0.119$	$^{36}\text{Ar}/^{38}\text{Ar} = 0.00143$
Salt	CsCl	1.170	2.2, 3.0	^{132}Xe	266	$^{130}\text{Xe}/^{132}\text{Xe} = 0.0014$	
Nitrate	$\text{Ba}(\text{NO}_3)_2$	0.890	223.2, 254.7	^{131}Xe	0.45	$^{131}\text{Xe}/^{129}\text{Xe} = 1.084$ $^{134}\text{Xe}/^{129}\text{Xe} = 0.194$	$^{132}\text{Xe}/^{129}\text{Xe} = 0.221$ $^{130}\text{Xe}/^{129}\text{Xe} = 0.019$

several days of pump-down, extraction blanks were run and repeated until satisfactory blank levels were achieved. Reagent samples were loaded in duplicate for analysis by isotope dilution mass spectrometry (IDMS). Two separate extractions were performed on the first sample of each pair: a 400°C extraction to assess any tendency toward low-temperature gas loss and to remove atmospheric contamination, then a 1650°C extraction to remove the bulk of the noble gases produced *in situ*. A single 1650°C extraction was performed on the second sample of each pair; the extracted gases were mixed either with our usual noble gas spike mixture (^3He , ^{21}Ne , ^{38}Ar , ^{80}Kr , ^{124}Xe) or with our air standard, as appropriate for IDMS analysis. The mineral samples were analyzed only by a single 1650°C extraction, without spiking for IDMS, so while isotopic ratios were measured with good precision, absolute concentrations determined by the peak-height comparison method for the mineral samples are much less precise than those determined by the IDMS analyses of the reagent samples.

The results of these analyses are summarized for each element as follows.

Neon: The neon extracted from the unannealed sodalite and albite mineral specimens and from the NaCl reagent sample are all isotopically similar. Thus we can conclude that we adequately characterized the isotopic composition of neon produced by 14.2-MeV neutrons on ^{23}Na : $(^{20}\text{Ne}/^{22}\text{Ne})_{\text{Na}} = 0.45 \pm 0.01$, $(^{21}\text{Ne}/^{22}\text{Ne})_{\text{Na}} = 0.017 \pm 0.001$. The absolute production rates in the NaCl can be used to calculate the cross sections listed in Table 24. The annealing treatment succeeded in removing about 99% of the neon in the irradiated sodalites. The annealed African sodalite shows evidence for a small ingrowth of ^{22}Ne from ^{22}Na , but the annealed Jacapuranga sodalite gave inconsistent results, partly due to large blank corrections. Definitive results on ^{22}Na production must await reanalyses after an adequate decay time (~ 1 y).

The isotopic compositions of the neon extracted from the enstatite and forsterite samples are quite similar, but differ from the neon produced in magnesium metal. The primary difference is probably due to contamination with atmospheric neon, since substantial quantities of atmospheric neon were released in the 400°C magnesium sample extraction, implying a high probability of further release at higher temperatures. However, simply subtracting atmospheric neon from the neon

extracted from the magnesium sample cannot reproduce the isotopic compositions observed in the magnesium minerals. One way to reconcile the data is to postulate that about 10% of the ^{22}Ne in the enstatite and forsterite is from a sodium impurity (reactions 2 and 3). If so, the isotopic composition of neon produced by 14.2-MeV neutrons on pure magnesium is given by $(^{20}\text{Ne}/^{21}\text{Ne})_{\text{Mg}} \approx 0.60$ and $(^{22}\text{Ne}/^{21}\text{Ne})_{\text{Mg}} \approx 0.081$. A more conservative statement is to give the limits imposed by the measurement of neon from magnesium metal, assuming an unknown contribution from atmospheric neon: $(^{20}\text{Ne}/^{21}\text{Ne})_{\text{Mg}} \leq 0.744$, $0.0194 \leq (^{22}\text{Ne}/^{21}\text{Ne})_{\text{Mg}} \leq 0.0951$.

Using the neon isotopic production rates determined from the NaCl and magnesium samples, and assuming stoichiometry, all of the natural-mineral samples appear deficient in the neutron-induced neon. The forsterite, enstatite, and sodalites each have about 25% less neon than expected, but the albite is even more deficient, with 57% less ^{22}Ne than expected from the production rate in NaCl. The apparently uniform depletion of neon in four of the five minerals is suggestive of a systematic error in analytical sensitivity or in fluence monitoring, since the minerals were irradiated and analyzed in a separate batch from the reagents; however, no such systematic error has yet been identified. Pending confirmation by further analyses, we offer no explanation for the apparently uniform depletion of the forsterite, enstatite, and sodalites, but note that the additional depletion of the albite indicates a substantial loss of neon from this mineral.

Xenon: The $\text{Ba}(\text{NO}_3)_2$ and CsCl samples were included to determine production rates of xenon isotopes from barium and cesium. Only two xenon isotopes were detectable above blank levels in the CsCl sample, with $^{130}\text{Xe}/^{132}\text{Xe} = 0.0014 \pm 0.0002$. Three separate reactions, (n,2n), (n,np), and (n,d), can contribute to the production of ^{132}Xe , and the IDMS measurement of ^{132}Xe concentration in the CsCl sample implies a value of 6.35 b for the sum of cross sections for these three reactions leading to ^{132}Xe . That is about four times greater than the cross-section literature would indicate. The cross section for ^{130}Xe production by $^{133}\text{Cs}(\text{n},\alpha)^{130}\text{I}(\beta^-)^{130}\text{Xe}$ is 9.0 mb from our data, compared to literature values ranging from 1 to 2 mb. The xenon extracted from $\text{Ba}(\text{NO}_3)_2$ was dominated by ^{131}Xe , primarily from $^{132}\text{Ba}(\text{n},2\text{n})^{131}\text{Ba}(\beta^+)^{131}\text{Cs}(\beta^+)^{131}\text{Xe}$, and ^{129}Xe , primarily from $^{130}\text{Ba}(\text{n},2\text{n})^{129}\text{Ba}(\beta^+)^{129}\text{Cs}(\beta^+)^{129}\text{Xe}$.

Lesser amounts of ^{132}Xe , ^{134}Xe , and ^{130}Xe were also produced; their isotopic ratios are given in Table 24. The irradiated mineral samples were dominated by excesses in ^{126}Xe , presumably from $^{127}\text{I}(\text{n},2\text{n})^{126}\text{I}(\beta^-)^{126}\text{Xe}$, and showed little if any evidence of xenon from cesium or barium.

Argon: The two major isotopes of argon produced in the neutron-irradiated KCl sample were ^{38}Ar and ^{39}Ar , with $^{39}\text{Ar}/^{38}\text{Ar} = 0.119$. We attribute the production of these isotopes primarily to the reactions $^{39}\text{K}(\text{n},2\text{n})^{38}\text{K}(\beta^+)^{38}\text{Ar}$ and $^{39}\text{K}(\text{n},\text{p})^{39}\text{Ar}$, although $^{39}\text{K}(\text{n},\text{np})^{38}\text{Ar}$, $^{39}\text{K}(\text{n},\text{d})^{38}\text{Ar}$, and $^{41}\text{K}(\text{n},\alpha)^{38}\text{Cl}(\beta^-)^{38}\text{Ar}$ can also contribute to ^{38}Ar production. We can eliminate any significant contribution from chlorine by noting that only very small amounts of ^{38}Ar from $^{37}\text{Cl}(\text{n},\gamma)^{38}\text{Cl}(\beta^-)^{38}\text{Ar}$ were observed in the NaCl and CsCl samples. Since potassium is only a minor constituent in our mineral samples, a complete quantitative interpretation of the argon isotopic data will require accurate potassium analyses of these minerals.

Krypton: The most abundant krypton isotope produced in the sodalite, enstatite, and forsterite was ^{80}Kr , primarily from $^{81}\text{Br}(\text{n},2\text{n})^{80}\text{Br}(\beta^-)^{80}\text{Kr}$. Smaller amounts of ^{82}Kr , probably from $^{81}\text{Br}(\text{n},\gamma)^{82}\text{Br}(\beta^-)^{82}\text{Kr}$, were also observed. The krypton released from the irradiated albite was

isotopically quite different from that of the other minerals. The most abundant krypton isotope in the albite sample was ^{84}Kr , presumably from $^{85}\text{Rb}(\text{n},2\text{n})^{84}\text{Rb}(\beta^+)^{84}\text{Kr}$. Quantitative confirmation of these conclusions will require trace-element analyses for bromine and rubidium in these minerals. We observed no evidence for significant production of krypton isotopes in any of the irradiated reagents.

Helium: The irradiated sodalites, NaCl, and $\text{Ba}(\text{NO}_3)_2$ each contained measurable quantities of helium produced by 14.2-MeV neutrons. Helium concentrations from 1 to several nl/g were observed, with $^3\text{He}/^4\text{He} \approx 3 \times 10^{-4}$. The other samples contained little or no ^3He , with ^4He up to a few nl/g; however, analyses of unirradiated samples are needed to determine how much, if any, of the observed ^4He was already present prior to the irradiation. The obvious sources for neutron-produced ^3He and ^4He are $(\text{n},^3\text{He})$, (n,α) , and $(\text{n},\text{n}\alpha)$ reactions, but we have not yet determined whether the observed abundances can be quantitatively attributed to specific reactions with specific target nuclides.

This research was supported by the LLNL Nuclear Chemistry Research Program.

Decay of ^{163}Ho : Implications for the Mass of the Neutrino

P. A. Baisden, D. H. Sisson, S. Niemeyer, G. B. Hudson, C. L. Bennett,¹ and R. A. Naumann¹

In a continuing effort to explore electron-capture decay properties of ^{163}Ho as a means of measuring the mass of the neutrino, we inferred an electron-capture Q value of approximately 265 keV. This value was determined from the measured half-life using known atomic physics factors and an estimate of the nuclear matrix element based on the systematics of electron-capture ft values of other holmium isotopes.

During the past several years we have been studying the electron-capture decay properties of ^{163}Ho as a means of measuring the neutrino mass. This holmium isotope is especially interesting because the electron-capture Q value ($Q_{\text{EC}} = 2.6 \pm 2.1$ keV), though quite uncertain, is unusually small.^{2,3} As a result, only electron capture from M (2.05-keV) or higher shells is energetically feasible. We had hoped to infer the mass of the neutrino from the measurement of the half-life in combination with measurements of several relative capture rates from different electron orbits. However, the precision required for such measurements, and hence the success of this approach, depends very heavily on the electron-capture Q value. From our measurement of the x-ray spectrum of ^{163}Ho following M capture,⁴ we were able to set a lower limit on the Q value of 2.1 keV. More recently we measured the half-life of ^{163}Ho ,⁵ and in the following we infer the electron-capture Q value from our measured half-life using known atomic physics factors and an estimate of the nuclear matrix element based on the systematics of electron-capture ft values of other holmium isotopes.

Given the total ^{163}Ho half-life, the electron-capture Q value can be calculated from the following relation.⁶

$$\lambda_{\text{total}} = \sum_i \lambda_i = \frac{g^2}{4\pi^2} |M|^2 \sum_i n_i B_i \beta_i^2 q_i w_i, \quad ,$$

where g is the weak interaction coupling constant and $|M|$ is the nuclear matrix element.

For electron state i , n_i is the occupation probability, B_i is the overlap and exchange correction, β_i is approximately the electron wave function amplitude over the nuclear volume, and q_i and w_i are the momentum and total energy, respectively, of the neutrino. For $m_\nu = 0$, $q_i = Q_{\text{EC}} - E_i$, where E_i is the binding energy of the i electron in the daughter atom.

In order to compute Q_{EC} , we need to know $|M|^2$, n_i , β_i , and B_i . In general, the quantities n_i , β_i , and B_i depend on the properties of bound atomic states, common to all isotopes of the same element, and as such are known with a fair degree of confidence.^{6,7} Consequently, the reliability with which we can determine Q_{EC} rests primarily on our estimate of $|M|^2$.

We can obtain values of $|M|^2$ for ^{157}Ho , ^{159}Ho , and ^{161}Ho using measured values of Q_{EC} , $t_{1/2}$, and a branching ratio for the 7/2[523] to 5/2[523] electron-capture transition for each isotope.⁴ Figure 63 shows a graph of $|M|^2$ (normalized to $|M|^2 = 1$ for ^{161}Ho) as a function of mass number. The uncertainties indicated for $|M|^2$ are based on the uncertainties in Q_{EC} and $t_{1/2}$ only. The constancy of the electron-capture rates, as reflected by the similar values of $|M|^2$ calculated for $^{157,159,161}\text{Ho}$, suggests that $|M|^2(^{163}\text{Ho}) \approx |M|^2(^{161}\text{Ho})$.

If, instead, we use the single-particle Nilsson model with pairing correlation⁸ to extrapolate $|M|^2$ from the lighter isotopes (as done by Andersen *et al.*⁹), we obtain the solid curve shown in Fig. 63, which gives $|M|^2(^{163}\text{Ho}) \approx 0.7|M|^2(^{161}\text{Ho})$. We point out that the application of the pairing model is questionable in this case since it presumes that the proton and

Figure 63. Relative values of $|M|^2$ as a function of mass number. Values obtained from the electron-capture decay of ^{157}Ho , ^{159}Ho , and ^{161}Ho involving the $7/2[523]$ to $5/2[523]$ transition are shown as (●). Also included are the relative $|M|^2$ derived from a (d,p) reaction on even-mass dysprosium isotopes populating the $5/2[523]$ neutron state (Δ). The variation of $|M|^2$ with mass as predicted by the pairing model is shown as the curve. All values of $|M|^2$ have been normalized to $|M|^2 = 1$ for ^{161}Ho .

neutron orbits have only a weak interaction compared to the pairing energy. In the case of an allowed unhindered transition like $^{163}\text{Ho} \rightarrow ^{163}\text{Dy}$, the proton and neutron states have very similar spatial wave functions, differing primarily by the coupling of the nuclear spin to the orbital angular momentum. Thus the possibility for significant proton-neutron interaction exists, contrary to the premise of the pairing model.

We can also examine (d,p) direct-transfer reactions for the even- A dysprosium isotopes where a Nilsson $5/2[523]$ neutron is created. In terms of the pairing model, $|M|^2$ for the $7/2[523]$ proton orbit to the $5/2[523]$ neutron orbit is proportional to the vacancy factor u^2 for the $5/2[523]$ neutron state. Likewise, the (d,p) cross sections, when adjusted for Q -value differences, are also proportional to u^2 . However, the relative (d,p) cross sections shown in Fig. 63 show a much steeper, more linear decrease with increasing A than predicted by the pairing model.¹⁰

The relation between the (d,p) cross sections and $|M|^2$ is more general than the pairing model, provided that the (d,p) reaction really is a direct single-nucleon transfer. However, it is found

Figure 64. Expected variation of half-life $t_{1/2}$ with the electron-capture Q value, assuming that $|M|^2(^{161}\text{Ho}) = |M|^2(^{163}\text{Ho})$ is shown as the solid curve. The measured $t_{1/2}$ of 4570 y implies a Q_{EC} of about 2.65 keV.

experimentally that the (d,p) reaction on the dysprosium isotopes to the state of interest, $5/2[523]$, has a very small cross section and may not be excited by a direct reaction mechanism.¹¹ Thus neither the pairing model nor (d,p) cross sections provide a suitable estimate for $|M|^2$, and we adopted $|M|^2(^{163}\text{Ho}) = |M|^2(^{161}\text{Ho})$ based on the lack of variation of $|M|^2$ derived from electron-capture ft values in other holmium isotopes. We then calculated the absolute value of the nuclear matrix element from published values for $t_{1/2}$, Q_{EC} , and the atomic physics factors for ^{161}Ho .

Using this value of the matrix element, we determined the relation between $t_{1/2}$ and Q_{EC} for ^{163}Ho , shown in Fig. 64. The measured half-life of 4570 y implies a Q_{EC} of approximately 2.65 keV. We emphasize that the value of the nuclear matrix element enters critically in the determination of Q_{EC} , as an uncertainty of $\pm 30\%$ on the matrix element, which corresponds to a Q_{EC} range of 2.45 to 2.90 keV. This determination is in good agreement with the value of 2.58 keV estimated by Anderson *et al.*⁹ from measurements of the partial half-life for capture of M electrons.

In an earlier report⁴ we pointed out that a Q_{EC} value of 2.6 keV would require a precision of 0.04% for measurements of relative capture rates from different electron orbits in order to establish a neutrino mass of 35 eV at the 1σ confidence level. Thus the Q_{EC} value reported here makes this approach impractical.

However, we believe that the resonant enhancement of the inner bremsstrahlung spectrum¹² accompanying the electron-capture

decay of ^{163}Ho can be exploited to detect an electron-neutrino mass as low as 35 eV. In order to explore this possibility, we have prepared a counting sample of approximately $200\text{ }\mu\text{g}/\text{cm}^2$ on a platinum backing from an isotopically pure source of $^{163}\text{HoF}_3$, using vacuum sublimation.¹³ We are presently counting this sample on a

beryllium-window Si(Li) detector at Princeton University. Later, we plan to count the sample with about a ten-fold increase in efficiency on the recently constructed windowless Si(Li) detector¹⁴ at LLNL.

This research was supported by the LLNL Nuclear Chemistry Research Program.

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Determining Neutrino Mass: Design of a Magnetic Spectrometer to Measure the Beta-Decay End Point of Atomic Tritium

W. Stöfl

We designed an atomic tritium source and a toroidal spectrometer to measure the tritium beta-decay probability as a function of energy near the end point. The spectrometer we propose will be capable of setting an upper limit of 5 eV on the mass of the electron antineutrino.

One of the most challenging problems facing experimental physicists today is that of determining whether electron antineutrinos (hereafter simply called neutrinos) have a non-zero rest mass. A neutrino mass greater than about 2 eV would be very important for astrophysics, because it would represent the missing mass to close the expanding universe. A massive neutrino would also have a tremendous impact on elementary particle physics theory.

Over the years, a number of measurements of the tritium beta spectrum have been carried out which set an upper limit of about 50 eV for the neutrino mass. The two most successful experiments done so far used spectrometers with the following characteristics. Bergkvist¹ used a double-focusing spectrometer with an instrumental resolution of 40 eV and a solid, electrostatically corrected tritium-aluminum source. Lubimov² at the Moscow ITEP used a toroidal field spectrometer with a resolution of 45 eV. The source was tritiated Valine, a complex plastic compound. They claimed to have evidence for a 34 ± 4 eV neutrino mass, but there are many doubts about this result.

The limitations of all modern neutrino-mass experiments have not been statistical but rather systematic. To give an upper limit for the neutrino mass using an instrumental resolution of less than this limit, one has to know the actual resolution function of the spectrometer very well. This is not easy, because all available calibration lines (electron conversion lines) have a natural width, which is greater than 10 eV and not very well known.

Another problem is the solid source that contains the tritium. The actual thickness is not

always well known and a few monolayers more or less can affect the result drastically. But the main uncertainty for even lower mass limits comes from the molecular structure of the tritium sources. The tritium decays from its nuclear and atomic ground state via beta decay (18.6-keV end-point energy) to the nuclear ground state of ^3He , but a significant fraction of the decay ends up with excited atomic or molecular states in ^3He and the surrounding molecule. This is equivalent to a large number of different beta end-point energies. The atomic and molecular excitations are in the range of 0 to several tens of electron volts, right in the most interesting range. The molecular excitation probabilities after the beta decay are quite unknown and depend a lot on the chemical structure of the source material. For neutrino masses below 10 eV, the chemical potential of the ^3He atom inside the source molecule becomes also important. The beta decay gives a recoil energy of about 3 eV to the ^3He atom, which allows the ^3He to move to practically any location nearby and makes precise calculations impossible.

The solution to these problems is the use of an atomic tritium source. The different atomic excitations for the tritium- ^3He decay are known very accurately. There is practically no small energy loss possible on the way from the source atom to the energy-analyzing device because of the 13-eV gap between the ground state and the first excited state in atomic tritium. That allows a practically undisturbed measurement of the last 10 eV of the beta spectrum to determine the neutrino mass with an upper limit of less than 5 eV, if necessary.

We have designed such a source and a matching magnetic spectrometer. The present design of the spectrometer has a resolution of about 5 eV and a very large acceptance, because only 10^{-10} of all tritium beta decays fall in the last 10 eV of the spectrum. By sacrificing transmission, we can improve the resolution. A diagram of the source, the spectrometer, and the detector is shown in Fig. 65.

We chose a magnetically analyzed atomic beam to produce a source of 100% atomic tritium. Negative tritium ions are made in a conventional ion source, then magnetically analyzed and focused to produce the desired source geometry. Molecules and hydrogen are filtered out here. In the next step the excess electron is stripped off by an intense laser beam, tuned to the right frequency. The cold neutral beam flies into the area where the beta decays are observed; the leftover ions are deflected by passing another magnet. A 0.2-A, 1-eV neutral beam will provide the necessary source strength within 3 m for the proposed experiment.

To boost the counting efficiency several orders of magnitude, the tritium decay region is in a very high magnetic solenoid field of 1–2 T. The magnetic-mirror effect pushes practically all beta electrons into one direction regardless of their emission angle, without changing their initial energy. The large toroidal magnetic spectrometer sees the beam source along its axis. The electrons and the neutral beam are separated by a short deflecting magnetic field. The neutral beam is caught in a beam dump with high pumping speed (provided by turbopumps). The electrons fly around this dump, guided by the magnetic field lines.

The electrons, following the expanding field lines at the end of the superconducting source magnet, are brought back to an intermediate focus (this time non-adiabatic) and then enter the toroidal spectrometer. The energy resolution of this 8-m-long and 2-m-diam toroidal field spectrometer is about 5 eV, but can be improved at the cost of transmission. The electrons are then focused into a small, high-resolution silicon detector. With a careful data analysis, a background count rate of less than one count a day can be achieved. The overall counting efficiency will be about 20% of all beta decays in the source, using a resolution of 5 eV at 23 keV. The electrons will be accelerated about 5 kV after the tritium-beam dump, but still inside the high magnetic field, to make them distinguishable from contamination tritium in the

detector area. This makes the low background count rate possible.

The overall spectrometer will operate in a scanning mode, where only one electron-energy bit is measured at a time. The energy is scanned by varying the 5-kV accelerating voltage. The toroidal field spectrometer will operate at a fixed current. Since the toroidal deflection field has a strength of about 15 G, the earth's field must be compensated and stabilized to about 1 mG. This can be done by active Helmholtz-type shielding. Mu-metal shielding becomes too costly at those dimensions.

The spectrometer will be calibrated with a high-quality electron gun, since there are no natural radioactive calibration sources with a sufficiently narrow emission line. However, the spectrometer can then be used to measure the width of such decay lines.

The tritium collected from the beam dump will be cleaned with standard hot uranium filters and immediately recycled to the source to keep the tritium inventory small (less than 1 Ci activity).

The high vacuum in the spectrometer will be achieved by several cryo- and turbopumps. The whole vacuum system will be stainless steel, to obtain a good vacuum and to have low tritium absorption in the walls. The main spectrometer tank can be baked to remove most of the tritium in case of necessary maintenance inside the tank.

The entire spectrometer will be controlled by computers. The use of commercially available CAMAC systems throughout will provide high reliability of the system.

The present design of the spectrometer uses state-of-the-art technology and components. The superconducting source magnet and the Si(Li) detector system are upgraded versions of our electron spectrometer.³ The toroidal field spectrometer is designed, but still requires detailed engineering work. The tritium beam source, the vacuum system, and the tritium handling and recycling system use standard components and their design is complete.

The proposed spectrometer will eliminate all major difficulties encountered in previous attempts to measure the neutrino mass and will be sensitive to a possible neutrino mass of 5 eV. The spectrometer can also be used for general high-resolution and high-sensitivity electron spectroscopy of gaseous or solid radioactive sources.

This research was supported by the LLNL Nuclear Test Program.

Figure 65 Schematic layout of the proposed electron spectrometer (not to scale).

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On the Branching Ratio in the Decay of ${}^7\text{Be}$

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We measured the branching ratio for ${}^7\text{Be}$ electron-capture decay to the first excited state in ${}^7\text{Li}$ by implanting a 20-MeV ${}^7\text{Be}$ beam into a silicon-detector telescope and counting the subsequent gamma-ray decays with well-calibrated Ge(Li) detectors. A branching ratio of $10.7 \pm 0.2\%$ was obtained. This value is in agreement with past measurements but does not agree with a recently reported measurement by a similar technique.

The electron-capture decay of ${}^7\text{Be}$ proceeds mostly to the $3/2^-$ ground state of ${}^7\text{Li}$ but includes a significant branch to the $1/2^-$ first excited state at 477.61 keV. The value of the branching ratio between these two decays is an important experimental and theoretical quantity in nuclear physics. In particular, the branching ratio of ${}^7\text{Be}$ is intimately connected with a discrepancy between observed and theoretical fluxes of energetic solar neutrinos.

The ${}^3\text{He}(\alpha, \gamma){}^7\text{Be}$ reaction is an important link in the chain of reactions that lead to the production of observable solar neutrinos. It has been proposed that the discrepancy between a recent measurement of the reaction rate and other measurements may be related to the value for the ${}^7\text{Be}$ branching ratio used to normalize the ${}^3\text{He}(\alpha, \gamma){}^7\text{Be}$ cross section. A new measurement² of the branching ratio has suggested a value of $15.4 \pm 0.8\%$. This represents a substantial increase over the value of $10.39 \pm 0.6\%$ obtained from a weighted average of previous measurements,³ and is inconsistent with other recent measurements. If the much larger value for the branching ratio were correct, it would have a significant effect on the solar neutrino problem and on other cross-section measurements and theoretical studies as well.

In view of the importance of the ${}^7\text{Be}$ branching ratio, we made an independent measurement of this quantity. The experiment relied on the implantation of ${}^7\text{Be}$ ions into a silicon surface-barrier detector. A 24-MeV ${}^7\text{Li}^{3+}$ beam from the LLNL tandem Van de Graaff facility impinged on a thin (1 to 2 mg/cm²) polyethylene target, $(\text{CH}_2)_n$. Most of the incident beam passed through the target and struck a 0° shadow bar which also served as a split Faraday cup to

monitor beam intensity and position. The remainder of the ${}^7\text{Li}$ beam interacted with the target and produced a flux of ${}^7\text{Be}^{4+}$ ions through the ${}^1\text{H}({}^7\text{Li}, {}^7\text{Be})n$ reaction. The kinematics of this reaction process generated a strongly forward-peaked distribution of ${}^7\text{Be}$ ions with an energy of about 20 MeV, which were transported and focussed through a series of six quadrupole lenses (quadrupole sextuplet beam transport spectrometer, or QSBTS). This "secondary" beam then passed through a 5-mm-diam collimator and struck a solid-state detector telescope consisting of a 6.1- μm -thick ΔE detector and a 400- or 1500- μm -thick E detector. The E detectors were much thicker than the range of 20-MeV ${}^7\text{Be}$ ions ($\sim 60 \mu\text{m}$ of silicon). All impinging ${}^7\text{Be}$ ions that passed through the collimator were within the 50-mm² active area of the detectors. A two-parameter ΔE - E spectrum allowed the ${}^7\text{Be}$ ions to be easily separated from possible contaminants and gave a clear picture of the ${}^7\text{Be}$ energy spectrum. To ensure that all events were counted, no two-parameter coincidence was required in the ΔE - E spectrum. Instead, the two-dimensional spectrum was only gated by the E -detector signal. The ${}^7\text{Be}$ count rate was maintained at 500 to 2000/s during different runs with targets of different thicknesses by maintaining an incident ${}^7\text{Li}^{3+}$ current of 1.5 to 3.0 nA.

Figure 66 shows a typical map of ΔE vs E for events in the detector telescope. Most of the scattered ${}^7\text{Li}$ beam was stopped by the shadow bar or spectrometer walls. Nevertheless, ${}^7\text{Li}^{3+}$ ions that inelastically scattered to 11 MeV could have had approximately the same trajectory through the spectrometer as the 20-MeV ${}^7\text{Be}^{4+}$ ions. Therefore, in addition to the ${}^7\text{Be}$ peak in

Fig. 66 (which constitutes about 60% of the total number of events), there is a lower-energy peak due to 11-MeV ${}^7\text{Li}^{3+}$. Other heavy contaminants and pile-up events, barely identifiable in Fig. 66, contribute less than 1% to the total number of collected events and are easily separated from the ${}^7\text{Be}$ contour. It is clear in Fig. 66 that there are few lower-energy ${}^7\text{Be}$ ions, and hence it is unlikely that undetected below-threshold ${}^7\text{Be}$ ions were implanted in the detector. If such undetected ions were present they could lead to

an erroneously high value for the branching ratio.

A pulser peak at high E and ΔE (triggered by the ${}^7\text{Be}$ peak) was used to monitor the total system dead time by comparison with a real-time scaler. The total dead-time correction was typically less than 2% and it was consistently maintained at less than 5% during the runs.

We performed three separate runs lasting for 15, 41, and 42 h. The detectors (labeled A, B, and C in Table 25) were implanted with 2.7×10^7 ,

Figure 66. Contours of ΔE vs E for the ${}^7\text{Be}$ beam implanted into the E detector.

Table 25. Summary of branching ratio measurements for ${}^7\text{Be}$ decay.

Detector	${}^7\text{Be}$ production		Gamma-ray count					Branching ratio (%)
	Length of irradiation (h)	${}^7\text{Be}$ ions at end of irradiation ^a ($t = 0$)	Ge(Li) system No.	Distance to detector (cm)	Start time (d)	Stop time (d)	478-keV photopeak area ^b	
A	14.9	$2.598 (0.012) \times 10^7$	2	0.53	1.0290	1.9904	1057 (58)	10.5 ± 0.6
B	40.7	$7.671 (0.009) \times 10^7$	2	4.35	20.0159	33.6077	6160 (170)	10.4 ± 0.3
C	42.0	$12.46 (0.21) \times 10^7$	1	6.35	17.8002	31.1073	4600 (140)	10.7 ± 0.4
			1	1.89	42.1903	50.4111	9750 (150)	11.1 ± 0.3
Weighted average ^c								10.7 ± 0.2

^a Uncertainty in parentheses is mostly due to identification of ${}^7\text{Be}$ boundaries in the ΔE - E map.

^b Statistical uncertainty and uncertainty due to background subtraction.

^c Uncertainty is the quadrature sum of weighted statistical and systematic uncertainties.

8.0×10^7 , and 15×10^7 ^7Be ions, respectively, before correction for ^7Be decay. The total yield was corrected for ^7Be decay during the irradiation by numerically evaluating the integral

$$^7\text{Be}(t) = e^{-\lambda t} \int_0^t R(t') e^{\lambda t'} dt',$$

where $R(t)$ is the rate of ^7Be production (averaged over 1-h intervals during the run), and λ is the ^7Be decay rate. These corrected yields are summarized in Table 25.

Prior to ion implantation, the silicon E detectors were observed with Ge(Li) counters to ensure that the detectors were free of ^7Be contamination from previous usage. One detector (A), which had been used in a preceding ^7Be experiment, was observed to have a background of about 1.1 ± 0.6 dis/min of ^7Be activity. This is corrected for in Table 25, and contributes an uncertainty of less than 3% to the final branching ratio measured with the detector. The other detectors showed no contamination, the upper limit of uncertainty in the branching ratio due to background correction was less than 0.1%.

We measured the gamma-ray decay of the implanted ^7Be ions independently using two of a series of shielded Ge(Li) detector systems (indicated in Table 25 as system Nos. 1 and 2). The existing efficiency calibration of each system was checked against standard radioactive sources placed about 18 cm from the detector housing. Table 26 shows a detailed comparison of measured and quoted decay rates from a series of selected standard sources. In particular, the sources provided a check of the efficiency calibration around the ^7Be 477.6-keV decay line. The results suggest a standard deviation of $\pm 1.4\%$ in reproducing the standard source strengths. We take this standard deviation as the systematic uncertainty due to calibration source uncertainty.

Since the gamma-ray radiation flux from the implanted ^7Be sources was relatively weak, we had to use a closer counting geometry (<18 cm) to obtain reasonable statistics. In this mode the finite sizes of both source and Ge(Li) detector are important parameters, and must be taken into account. By using a model⁴ which reduces both the source and the detector to geometric points, we were able to make the geometric corrections.

Table 26. Comparison of quoted and measured absolute gamma-ray strengths for selected standard sources.

Nuclide	Vendor identification No.	Quoted source strength ^a (10^6 dis/min)	Measured source strength ^b (10^6 dis/min)	Difference (%)	Vendor ^c
System No. 1^d					
^{133}Ba	3432	2.597 (0.2%)	2.612 (1.5%)	-0.5	LMR
^{137}Cs	1901	2.424 (1.5%)	2.476 (1.1%)	-2.1	LMR
^{137}Cs	070	22.80 (1.3%)	22.85 (1.0%)	-0.2	IAEA
^{137}Cs	74-092	22.64 (1.7%)	22.61 (1.0%)	-0.1	OMH
^{137}Cs	3962	24.80 (1.3%)	25.29 (2.6%)	+2.0	AMR
System No. 2^d					
^{133}Ba	3432	2.597 (0.2%)	2.610 (1.4%)	-0.5	LMR
^{134}Cs	336-80	23.64 (1.0%)	23.13 (1.0%)	+1.7	PTB
^{137}Cs	1901	2.424 (1.5%)	2.450 (1.0%)	-1.1	LMR
^{137}Cs	070	22.80 (1.3%)	22.43 (1.0%)	+1.6	IAEA
^{137}Cs	74-092	22.64 (1.7%)	22.31 (1.0%)	+1.5	AMR

^a Uncertainties in parentheses are quoted by the vendor.

^b Uncertainties in parentheses are statistical only.

^c PTB, Physikalisch-Technische Bundesanstalt, Berlin; IAEA, International Atomic Energy Agency, Vienna; OMH, National Office of Measures, Budapest; AMR, Amersham International Limited, Amersham, U.K.; LMR, Laboratoire de Metrologie des Rayonnements Ionisants, Saclay.

^d The Ge(Li) detectors associated with system Nos. 1 and 2 had coaxial geometry, with active volumes of 46 and 57 cm³, respectively.

and estimate reliably the uncertainties due to these corrections. We checked the geometric correction by using a small quantity of chemically separated ^7Be deposited on a silicon detector. The source material was deposited as a thin film (<0.0015 cm thick) over an area about 0.5 cm in diameter, a reasonable approximation of the spot size of the implanted ^7Be sources. The gamma radiation from the chemically separated source was subsequently measured in each spectrometer system at precisely known positions (± 0.01 cm relative to the detector housing), and the geometric correction was applied. Table 27 summarizes these data for one of the spectrometer systems (No. 2). The data show that for source-to-detector separations between about 18 and 2 cm, the model yields a constant source strength with an uncertainty of better than $\pm 0.5\%$. This indicates the accuracy to which the geometric correction can be made.

Counting times for each implanted ^7Be source ranged from 1 to 14 d. Before each counting experiment, a ^{57}Co radiation source was placed near the detector housing to yield a counting rate of about 80/s for the 121-keV decay line. This source remained fixed through all counts for calibration and background as well as through the count of the implanted ^7Be detectors. During periods of extended counting, we checked the accumulation rate in the 121-keV photopeak periodically for any deviations in the counting rate due to system instabilities. An additional check was done by measuring the decay rate of a standard ^{137}Cs source several times before and after each long count. These data showed no detectable changes ($>0.5\%$) in detector efficiency during the extended counts. The overall system dead time was determined primarily by the decay of the ^{57}Co monitor source, and was less than 0.5% for all measurements of ^7Be activity and system backgrounds.

We checked the background in the Ge(Li) spectrometer systems with a count about 14 h long before and after each extended count of the implanted ^7Be detectors. Observed background rates ranged from <0.1 (count/h)/channel (No. 1) to <0.2 (count/h)/channel (No. 2), and produced no distinguishable peak structures in the vicinity of the 477.6-keV photopeak. Figure 67 shows a composite plot of the peak and background spectrum. Finally, any extraneous ^7Be activity that might have been deposited outside the sensitive area of the detector was estimated by counting the various mechanical components that supported the silicon detector

Table 27. Measured source-strength reproducibility as a function of source-detector separation for a chemically separated ^7Be source.

Count No.	Measured source strength ^a (10^6 dis/min)	Source-to-detector distance ^b (cm)
1	3.196	17.783
2	3.198	14.783
3	3.174	12.783
4	3.187	10.783
5	3.159	8.783
6	3.163	6.783
7	3.174	5.783
8	3.155	4.783
9	3.190	3.783
10	3.195	3.283
11	3.161	1.783
Average		3.177 ± 0.016 (0.5%)

^a Statistical uncertainty for each value is about 0.5%.

^b Estimated position uncertainty is ± 0.01 cm.

Figure 67. Typical Ge(Li) spectrum, showing the prominent 477.6-keV gamma-ray line. The total counting time for this spectrum was 13.31 d at a source-to-detector distance of 6.35 cm. The lower portion of the figure shows the background observed during a 14-h count immediately preceding the photopeak measurement.

during the implantation runs. Counting times ranging from 6 to 20 h on selected components showed no unexpected ^7Be activity above background.

We combined the results of the gamma-ray counting measurements, after appropriate corrections for radioactive decay, with the measurements of the number of ^7Be ions deposited on each detector to determine the electron-capture branching ratio. The results from the three separate runs and their respective gamma-ray counts are summarized in Table 25. Also summarized are the sources of uncertainty

associated with each measurement. From these results we obtain a weighted average of $10.7 \pm 0.2\%$ for the branching ratio. This value, together with other measurements, gives a weighted average and standard deviation of $10.45 \pm 0.05\%$. We conclude that the solar neutrino problem and the discrepancies in the $^3\text{He}(\alpha, \gamma)^7\text{Be}$ cross section are not caused by an error in the previous measurement² of the ^7Be branching ratio.

This work was supported by the LLNL Nuclear Test Program.

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A Search for Superheavy Elements Using the $^{48}\text{Ca} + ^{254}\text{Es}$ Reaction

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We measured upper limits for the production of superheavy elements (SHEs), using the compound-nucleus reaction $^{48}\text{Ca} + ^{254}\text{Es}$. This combination permits the closest approach of any practical fusion reaction to the predicted island of stability at the 184-neutron shell. We used aqueous and gas-phase chemistry to isolate SHE fractions within 1 h from the end of bombardment. The fractions were counted for spontaneous fission activity and energy. In these preliminary experiments, we measured cross-section upper limits of about $5 \times 10^{-32} \text{ cm}^2$ for half-lives from fractions of a day to a few weeks.

Theoretical calculations by Randrup *et al.*³ and Fiset and Nix⁴ predict that the maximum stability against spontaneous fission (SF) is at the closed 184-neutron shell for the superheavy elements (SHEs). Half-lives for SF differ by 10^9 between these calculations. The calculations of Randrup *et al.* show that the fission-barrier heights decrease more rapidly below 184 neutrons, becoming only 3 to 4 MeV at 178 neutrons, with corresponding SF half-lives of 10^{-16} y as compared with years or more at 184 neutrons. The half-lives near 184 neutrons are thus dominated by alpha or beta decay.

This sensitivity of SF half-life to neutron number for the SHEs dictates a very small choice of nuclear reactions that yield a product as close as possible to the 184-neutron shell. The much favored and studied $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction would produce element 116 with 178 neutrons after emission of two neutrons from the compound nucleus. The experimental cross-section limits of 10^{-35} cm^2 for this reaction indicate either a half-life shorter by a factor of 10^{13} than the 130 y predicted by Fiset and Nix, or a drastically lower production cross section. If the predictions of Randrup *et al.* are reasonable, the SF half-life of this nuclide would be less than 10^{-16} y and the fission barrier only 4 MeV. A fission barrier below the neutron binding energy would result in such a reduction in the reaction cross section by leaving the nucleus

with an excitation energy greater than the fission barrier but less than the neutron binding energy. This phenomenon is estimated to lower the cross section by more than 10^9 because of fission during de-excitation of the compound nucleus. The consequence of such an effect makes it imperative to choose a reaction which comes as close as possible to the 184-neutron shell, where the fission barriers are highest. From this standpoint, the $^{48}\text{Ca} + ^{254}\text{Es}$ reaction is the most favorable compound-nucleus reaction currently available. Randrup *et al.* predict fission barriers of greater than 6 MeV for the 2n out-product of the einsteinium reaction, with SF half-lives comparable to the alpha half-lives. A large additional gain in half-life would be made because the 2n out-product from this reaction is an odd-odd nucleus.

We bombarded a $5\text{-}\mu\text{g}/\text{cm}^2$ ^{254}Es target with ^{48}Ca at the SuperHILAC accelerator at the Lawrence Berkeley Laboratory (Berkeley, CA). (Cross sections for actinide transfer-reaction nuclides produced by this reaction are given in "Transfer Cross Sections from the Reaction of ^{48}Ca and ^{254}Es ," in this section.) There were four bombardments, each lasting several hours, for a total of 7.7×10^{15} ^{48}Ca particles. Two ^{48}Ca bombarding energies were used: 238 and 246 MeV. These energies are 2 MeV under and 6 MeV over the Coulomb barrier, respectively. They were chosen to minimize excitation energy

in the compound nucleus in order to minimize fission and the number of neutrons emitted during de-excitation.

We collected recoils from the einsteinium target on metal foils, which were then chemically processed to yield SHE fractions for SF counting. Two types of chemistry were performed, based on the predicted properties of SHEs.⁵⁻⁷ These chemistries were designed to recover elements 110 through 118. The primary fusion product, element 119, was predicted to decay rapidly³⁴ by alpha emission into that range of elements and would thus be detected by the SF or possibly alpha decay of its descendants. We performed a solution-chemistry procedure using sulfide precipitations and cation-exchange chromatography within a 45-min period from the end of bombardment (EOB) to the beginning of counting. Chemical yields of the naturally occurring elements platinum through polonium were used as substitutes for elements 110 (eka-Pt) through 116 (eka-Po). Table 28 shows the chemical yields, which were fairly consistent except for lead. The yield of lead was apparently low because of slow stripping from the cation-exchange column. Because eka-Pb is predicted to be more strongly complexed in HCl, its chemical yield should be higher.

We performed gas-phase chemistry on the recoil products from two of the bombardments. This chemistry is based on the predicted increase in volatility and in noble character of elements 112 through 117 in their elemental state, relative to their naturally occurring homologs. Noble gases (element 118) were also collected with this procedure. The recoil catcher foil was heated in a quartz apparatus using hydrogen carrier gas. The SHEs were subsequently condensed on palladium-covered nickel foils, which were counted for SF and alpha activity. The noble gases were collected within 20 min after EOB

Table 28. Percent chemical yields for SHE homologs, obtained from the two solution-chemistry procedures.

Bombardment	Element						
	110 Pt	111 Au	112 Hg	113 Tl	114 Pb	115 Bi	116 Po
A	40	44	48	47	18	54	43
B	47	53	55	53	21	61	59

with about an 80% chemical yield. Two additional fractions were collected which included elements that condense in the ranges 5 to 400°C and 400 to 1000°C. The collection times for these fractions were 2 to 3 h and the chemical yields were about 25 and 60%, respectively. Both types of chemistry provided excellent decontamination from the actinide elements, whose isotopes interfere by also decaying by SF.

In over two months of counting, we observed no SF events in samples from either of the chemistries. By assuming the detection of two SF events and a 50% chemical yield, we determine that the cross section for SHE production is $\leq 5 \times 10^{-32}$ cm² for half-lives of a few days for all bombardments. Figure 68 shows the half-life vs cross-section limits we obtained for the two solution-chemistry experiments only.

We believe that the $^{48}\text{Ca} + ^{254}\text{Es}$ reaction deserves further attention as a pathway to SHEs. The proposed production of 40 µg of ^{254}Es at Oak Ridge National Laboratory (Oak Ridge, TN) could produce a target 100 times larger than that used in our experiments. With a larger target and increased bombardment times, cross sections of about 10^{-35} cm² are achievable.

It is also important to increase the sensitivity for short lifetimes. The calculations of Randrup *et al* suggest that we cannot expect a long-lived SHE to result from alpha and beta decay of element 119, the primary fusion product. Instead, one or two alpha decays, several tens of milliseconds apart, would be followed by SF decay. Such millisecond half-lives are accessible by the SHIP velocity filter at GSI (Darmstadt, FRG), or perhaps by a fast helium-jet system.

This research was supported by the LLNL Nuclear Chemistry Research Program

Figure 68. Cross-section limits for the production of SHEs as a function of half-life, from solution-chemistry experiments.

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Transfer Cross Sections from the Reaction of ^{48}Ca and ^{254}gEs

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We measured transfer cross sections for the production of isotopes of einsteinium, fermium, mendelevium, and nobelium from the bombardment of ^{254}gEs with 266-MeV ^{48}Ca ions. Cross sections for the fermium isotopes are similar to those measured previously in bombardments with ^{18}O and ^{22}Ne , but the mendelevium isotopic cross sections are about one order of magnitude lower. In the case of mendelevium, the bombarding energy of ^{48}Ca , a projectile with a high binding energy, was not sufficient to entirely overcome the very negative Q values of the reactions.

^{254}gEs is the most neutron-rich isotope that can be obtained in quantities sufficient to provide adequately thick targets (about $100\ \mu\text{g}/\text{cm}^2$) for possible superheavy-element (SHE) production. The aim of our work is to investigate the feasibility of using ^{254}gEs for such a purpose by measuring cross sections for the production of known heavy isotopes and, subsequently, by using the derived cross-section systematics to search for hitherto unknown neutron-rich isotopes with $Z = 101$ up to the SHEs. The discovery and characterization of these unknown sub-SHE isotopes improves the possibility of producing SHEs both by allowing us to predict the optimum experimental conditions for a search and by reducing the length of the extrapolations necessary to estimate the SHE half-lives and cross sections.

We bombarded a $4.8\text{-}\mu\text{g}/\text{cm}^2$ ^{254}gEs target, electroplated on a thin beryllium foil, with 266-MeV ^{48}Ca ions from the SuperHILAC at Lawrence Berkeley Laboratory (Berkeley, CA). The laboratory bombarding energy of 266 MeV is about 11% above the Coulomb barrier; this amount of energy over the barrier generally provides the maximum transfer cross sections. We had previously investigated transfer yields from heavy-ion bombardments of ^{254}gEs by using as projectiles ^{18}O (98 MeV), ^{22}Ne (121 and 126 MeV), and ^{136}Xe (826 and 888 MeV).³

The reaction products recoiling from the einsteinium target were collected on a copper

Figure 69. Measured cross sections for isotopes of einsteinium, fermium, mendelevium, and nobelium, produced in the bombardment of ^{254}gEs with 266-MeV ^{48}Ca ions.

catcher foil 6.3 mg/cm² thick. Following each of two short (~2 h) bombardments, the catcher foil was transported to a laboratory for separation of the actinide products before isotopic analysis by alpha spectroscopy with surface-barrier detectors.

Figure 69 shows the measured cross sections for the actinide products. Figure 70 compares the cross sections for fermium, mendelevium, and nobelium isotopes to those obtained in the ²⁵⁴Es bombardments with ¹⁸O and ²²Ne. Although the fermium cross sections from all the bombardments are comparable, the mendelevium cross sections are more than one order of magnitude lower for the ⁴⁸Ca bombardment. It has been suggested by Artukh *et al.*⁴ and Bondorf *et al.*⁵ that an observed correlation between the cross section and the ground-state Q value (Q_{gg}) for a given transfer reaction is due to the statistical nature of the transfer mechanism. In a simple phase-space argument, the transfer probability (and, hence, the cross section) is determined by the level density of the final system, which is proportional

to $\exp [(Q_{gg} + \Delta E_C)/T]$. Here, ΔE_C is the difference in Coulomb energy between the entrance and exit channels, and T is the equilibrium temperature of the dinuclear intermediate system. However, this approach is not considered reliable when the bombarding energy is close to the Coulomb barrier, as is the case here. In addition, there is no systematic difference in Q_{gg} values for producing isotopes of a given element, whether the projectile is ¹⁸O, ²²Ne, or ⁴⁸Ca.

If, instead, we take the level density of the final system to be related to the product excitation energy E^* , as do Hoffman and Hoffman,⁶ we gain a better qualitative understanding of why the mendelevium isotopic cross sections are lower for the ⁴⁸Ca bombardment. Hoffman and Hoffman⁶ compared cross sections measured by Lee *et al.*⁷ for the production of isotopes of berkelium through fermium from bombardments of ²⁴⁸Cm with ¹⁶O, ¹⁸O, ²⁰Ne, and ²²Ne. They found that the reaction cross section for each element is

Figure 70. (a) Comparison of cross sections for the production of fermium isotopes in the bombardment of ²⁵⁴Es with 98-MeV ¹⁸O, 121-MeV ²²Ne, 126-MeV ²²Ne, and 266-MeV ⁴⁸Ca. (b) Production of mendelevium and nobelium isotopes.

Table 29. Comparison of calculated excitation energies for mendelevium isotopes from the bombardment of ^{254}Es with ^{18}O , ^{22}Ne , and ^{48}Ca .

Mendelevium isotope	E^* (MeV)			
	^{18}O (98 MeV)	^{22}Ne (121 MeV)	^{22}Ne (126 MeV)	^{48}Ca (266 MeV)
^{254}Md	-6.29	-0.83	-0.83	-4.76
^{255}Md	+3.72	+0.46	+0.62	-3.77
^{256}Md	+0.68	+2.55	+2.52	+2.25
^{257}Md	+2.53	+0.90	+1.34	-3.31
^{258}Md	+6.10	+1.77	+2.39	-3.01

roughly proportional to $\exp(cE^*)$, where c is a positive constant representative of each heavy element product. In cases where E^* is negative, i.e., the available product mass falls below the ground-state mass surface, the energy deficit acts as a barrier to the reaction. Since reactions between a given target and projectile that yield the same products vary in impact parameter and energy transferred, E^* is not a single-valued parameter but a representation of the weighted average value of an energy distribution for a given reaction. A reaction with a negative E^* can still proceed to some extent, being driven by the high-energy tail of the excitation-energy distribution. Table 29 shows the calculated (average) excitation energies for mendelevium isotopes from our bombardments of ^{254}Es with ^{18}O , ^{22}Ne , and ^{48}Ca . The energies are based on the equation $E^* = E_{\text{CM}} + Q_{\text{gg}} - E_{\text{B}}(f) - KE(f)$,⁶ where E_{CM} is the center-of-mass bombarding energy, $E_{\text{B}}(f)$ is the Coulomb barrier of the product nuclei, and $KE(f)$ is the center-of-mass kinetic energy of these products.

We see in Table 29 that the excitation energies for mendelevium isotopes from ^{48}Ca bombardment are considerably lower than those from the other projectiles. A ratio of cross sections, $\sigma_{\text{Ca}}/\sigma_{\text{X}}$, in which the production of a given mendelevium isotope from ^{48}Ca bombardment is compared with that from another heavy-ion projectile, would be proportional to $\exp[c(E_{\text{Ca}}^* - E_{\text{X}}^*)]$. For cases in which $E_{\text{Ca}}^* < E_{\text{X}}^*$, the cross-section ratio is less than 1.

Our measured ^{259}No cross section from the ^{48}Ca bombardment is comparable to cross sections from the other bombardments. The treatment of excitation energy above, however, suggests that it should be considerably lower. The ^{259}No cross section from the ^{48}Ca bombardment should be taken as an upper limit, since the number of events upon which it is based is very small, and no spontaneous-fission events were observed from the electron-capture daughter, ^{259}Md .

We conclude that ^{48}Ca produces transfer products with yields similar to those obtained with oxygen and neon ions. The conclusion comes from this experiment, from analysis of the results of bombarding of ^{254}Es with ^{18}O and ^{22}Ne ions, and from the results of bombarding ^{248}Cm with ions of O and Ne (Ref. 7), and ^{48}Ca (Refs. 8 and 9). In all cases, the maximum attainable cross sections are on the order of a few millibarns, and for the transfer of a given number of nucleons, the cross sections are similar. The low mendelevium cross sections in this experiment demonstrate that a projectile bombarding energy must be chosen to yield product excitation energies large enough to permit sampling of the available ground-state mass surface and to provide the highest possible cross sections. At the same time, the bombarding energy must not excite the heavy product to the point that it is largely destroyed by fission.

This work was supported by the LLNL Nuclear Chemistry Research Program.

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Dynamics of Fission and Superheavy Nuclei

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We used a new microscopic dynamic model to recalculate the fission half-lives of superheavy nuclei. We also calculated alpha- and beta-decay half-lives with standard methods. We find that the previous estimates of fission half-lives for the isotopes of $Z = 114$ were too optimistic. Our calculation suggests that the $Z = 126$ isotopes are more stable against fission than the $Z = 114$ isotopes, but are subject to alpha decay.

The predictions of the half-lives of nuclei in the unknown charge-mass regions are complicated by the large uncertainty in the ground-state mass extrapolations and by the lack of a reasonable solution of the dynamic Hamiltonian associated with nuclear fission. In view of the enormous effort already spent and planned in search of the previously predicted island of relatively stable superheavy nuclei,^{2,3} we recalculated fission half-lives of superheavy nuclei with a new microscopic dynamic model. We also calculated alpha- and beta-decay half-lives with standard methods.

The main differences between our study and the previous best calculation are as follows:

1. Instead of assuming *a priori* that a nucleus is spherical or deformed, axial or nonaxial, we let the collective Schrödinger equation govern the shape of the nucleus.

2. We calculated the energy of zero-point motion exactly by solving the collective Schrödinger equation of nuclear deformations. The calculated value varies from 1.4 MeV for ²⁴⁰Pu to 21.8 MeV for ²⁰⁸Pb. This is in marked contrast to the previously assumed value of 0.5 MeV for all heavy nuclei.

3. We calculated the three shape-dependent mass parameter functions microscopically. Previously, a single function extrapolated from the asymptotic form and fitted to the observed lifetime values was used.

4. We used the shell-correction method to calculate the deformation energy surface, as previously. We matched this potential, however,

to the correct asymptotic tail at large fragment separations. In addition to the fragment masses and the exact Coulomb interaction between them, this tail depends on a nuclear interaction of the Yukawa type whose two parameters are deduced from a fit to two known fission half-lives.

5. We assumed that the strengths of the spin-orbit and ℓ^2 terms of the single-particle Hamiltonian are shell-dependent (i.e., N -dependent, where N is the principal quantum number), rather than Z - and A -dependent. This contrasts with the Nilsson model,⁴ in which these strengths are taken to be Z - and A -dependent. We determined the values for the lower shells ($N = 0$ to 2) by fitting the spectra of nuclei near closed shells, as is also done in the Nilsson prescription. But the values for higher shells were determined by means of a stationary condition^{5,6} imposed on the Strutinsky shell correction⁷ for all doubly magic nuclei (N or $Z = 2, 8, 20, 50, 82, 126, \text{ and } 184$). That is,

$$\frac{\partial(\delta U)}{\partial \nu_{\ell s}} = 0 = \frac{\partial(\delta U)}{\partial \nu_{\ell \ell}}, \quad (1)$$

where δU is the Strutinsky shell correction, and $\nu_{\ell s}$ and $\nu_{\ell \ell}$ are the spin-orbit and ℓ^2 strengths, respectively. The same values were used for protons and neutrons, except for an overall scaling factor for the energy levels.

We based our calculations on the dynamic deformation model⁸ (DDM), which has been shown to reproduce the low-energy spectra of

selected even-even nuclei from ^{12}C to ^{240}Pu without any parameter adjustments. We extended the model to the large deformations expected in the fission of transuranic and superheavy nuclei.⁹

The collective Hamiltonian is given by⁸:

$$H_{\text{DDM}} = 1/2 [B_{\beta\beta}(\beta, \gamma) \dot{\beta}^2 + 2B_{\beta\gamma}(\beta, \gamma) \dot{\beta} \dot{\gamma} + B_{\gamma\gamma}(\beta, \gamma) \dot{\gamma}^2] + 1/2 \sum_{k=1}^{k=3} \theta_k(\beta, \gamma) \omega_k^2 + V_{\text{DDM}}(\beta, \gamma) , \quad (2)$$

where the Hill-Wheeler¹⁰ parameters β and γ are given by

$$\begin{aligned} R_1 &= R_0 \exp \left[\sqrt{\frac{5}{4\pi}} \beta \cos \left(\gamma - \frac{2}{3}\pi \right) \right] \\ R_2 &= R_0 \exp \left[\sqrt{\frac{5}{4\pi}} \beta \cos \left(\gamma + \frac{2}{3}\pi \right) \right] \\ R_3 &= R_0 \exp \left[\sqrt{\frac{5}{4\pi}} \beta \cos \gamma \right] , \end{aligned} \quad (3)$$

where R_1 , R_2 , and R_3 are the semi-axes of an ellipsoid and R_0 is the radius of the sphere with the same volume. The vibrational mass parameters are identified by $B_{\beta\beta}$, $B_{\beta\gamma}$, and $B_{\gamma\gamma}$, and the rotational moments of inertia are θ_k ($k = 1, 2, 3$), with ω_k the rotational frequencies. We require that the Hamiltonian H_{DDM} be invariant under rotations, time reversal, and reflection across any plane passing through the origin.

The potential $V_{\text{DDM}}(\beta, \gamma)$ is related to the “true” potential for the fission model as

$$\begin{aligned} V &= V_{\text{DDM}}(\beta, \gamma) - \kappa_2(\rho - 3/4)^2 \quad \text{for } \rho < \rho_t \\ &= M_1 c^2 + M_2 c^2 + \frac{C_t}{\rho} - \frac{C_A}{\rho} \exp \left[-\frac{\rho - \rho_t}{\rho_0} \right] \\ &\quad \text{for } \rho \geq \rho_t . \end{aligned} \quad (4)$$

The quantity ρ is a dimensionless variable and is related to the separation distance r between fragment centers by $\rho = r/R_0$. The distance ρ_t is given by the equation $(R_1 + R_2)/R_0 = (A_1^{1/3} + A_2^{1/3})/A^{1/3}$, where R_1 and R_2 are the radii of the two fission fragments with mass numbers A_1 and A_2 , respectively. The masses $M_1 c^2$ and $M_2 c^2$ are

taken from the droplet model for given (Z_1, A_1) and (Z_2, A_2) combinations.¹¹ The constants C_t and C_A are given by

$$C_t = \frac{Z_1 Z_2 e^2}{R_0} , \quad C_A = 84.17 \rho_0 (A_1 A_2)^{1/5} ,$$

where

$$\rho_0 = \frac{\alpha_0}{R_0} , \quad \text{and} \quad \alpha_0 = \frac{\hbar c}{m_\pi c^2} = 1.438 \text{ fm} . \quad (5)$$

The quantity C_A was obtained from fits to the spontaneous-fission half-life $t_{1/2}^{\text{SF}}$ of ^{240}Pu and $^{260}\text{104}$. The parameter κ_2 is chosen so that $V(\rho < \rho_t) = V(\rho > \rho_t)$ at $\rho = \rho_t$. Also, we require that the potential [Eq. (4)] and its derivative be continuous at $\rho = 3/4$.

If one knows how to solve Eqs. (2) and (4), a matter discussed below, then the spontaneous-fission half-life, $t_{1/2}^{\text{SF}}$, can be calculated by using the following expressions:

$$\begin{aligned} t_{1/2}^{\text{SF}} &= \frac{(\ln 2) \hbar}{\Gamma_f} , \quad \Gamma_f = \sum_{A_1} \sum_{Z_1} \Gamma_f(Z_1, A_1) , \\ \Gamma_f(Z_1, A_1) &= \frac{\hbar \omega P_f(Z_1, A_1)}{2\pi} \\ P_f(Z_1, A_1) &= \frac{1}{1 + e^{A_f}} \end{aligned} \quad (6)$$

$$A_f(Z_1, A_1) = 2 \int_{s_s} \sqrt{2B_{ss}(V_s - E)} ds .$$

The fission path was chosen by minimization of the action A_f for each charge-mass split $(Z_1, A_1; Z_2, A_2)$. The total energy of the fissioning nucleus is E , and for spontaneous fission it is the zero-point energy, calculated by Eq. (2). We calculated the alpha-decay half-life t_α from the following expression:

$$\log(t_\alpha/s) = A_Z(Q_\alpha/\text{MeV})^{-1/2} + B_Z , \quad (7)$$

where $Q_\alpha(Z, N) = M(Z, N) - M(Z - 2, N - 2) - M(2, 2)$, and M denotes the atomic mass excess of the nucleus in question. $M(Z, N)$ and $M(Z - 2, N - 2)$ were taken from the DDM calculations. We obtained the parameters A_Z and B_Z from the systematics of alpha decay as given by Viola and Seaborg¹²:

$$A_Z = 2.11329Z - 48.9879$$

$$B_Z = -0.39004Z - 16.9543 \quad (8)$$

For beta-decay calculations we used the method employed by Fiset and Nix.¹³

The Hamiltonian H_{DDM} is solved numerically for 92 β, γ mesh points; β ranges from 0 to 0.97 and γ from 0° to 60°. Figure 71 shows the main steps leading to the solution. Step 1 is similar to the Nilsson model in that a large configuration space ($N = 0$ to 10 major shells in our case) is employed. In the Nilsson model, however, the $\Delta N = 2$ mixing is included only approximately by using the cylindrical basis; in contrast, we use the spherical-oscillator basis and include the $\Delta N = 2$ mixing exactly by diagonalizing the triaxial-oscillator Hamiltonian. Also, instead of the Bohr definition¹⁴ of deformation employed in the Nilsson model,⁴ we employ the Hill-Wheeler¹⁰ definition. The latter allows us to include small amounts of hexadecapole deformations (not only β_{40} but also β_{42} and β_{44}) without having to introduce them as extra degrees of freedom. The spin-orbit and ℓ^2 strengths are parameterized in such a way that the single-particle level energies and matrix elements are independent of Z and A , except for an isospin-dependent energy scaling factor defined in terms of the oscillator energy constants:

$$\hbar\omega_p = (41 \text{ MeV})A^{-1/3} \left[1 - \frac{N-Z}{3A} \right]$$

$$\hbar\omega_n = (41 \text{ MeV})A^{-1/3} \left[1 + \frac{N-Z}{3A} \right] \quad (9)$$

The constant 41 MeV comes from the requirement that the nuclear radius be given approximately by $R = (1.2 \text{ fm}) A^{1/3}$. The $N - Z$ dependence comes from the requirement that protons and neutrons have approximately the same radii. Thus, even though we start with the same single-particle basis for protons and neutrons, the final single-particle levels are different for protons and neutrons.

In step 2, the pairing effects are included by performing the Bogoliubov¹⁵ transformation on the deformed quasi-particle basis. In contrast to the usual BCS method, in which only the particle-particle channel of the pairing interaction is included, we also include the particle-hole channel. This turns out to be particularly important for removing the possibility of divergence in the expressions for the mass parameters and moments of inertia.

In step 3, the shell-correction method of Strutinsky⁷ is employed in order to calculate the potential energy of deformation (β and γ). Instead of the usual liquid-drop model, we employ the droplet model¹¹ for the uniform part of the potential energy.

We use the cranking-generator coordinate method¹⁰ in step 4 in order to calculate the three moment-of-inertia functions and the three vibrational mass-parameter functions (for $\beta\beta$, $\beta\gamma$, and $\gamma\gamma$ vibrations). These six functions, the potential function, and the electromagnetic moment functions are calculated microscopically for each point of a β - γ mesh consisting of 92 mesh points. The pair-fluctuation corrections are included for each of these points. We then quantize the collective Hamiltonian, a generalized version of the Bohr Hamiltonian for five-dimensional quadrupole vibrations,¹⁴ and solve the corresponding collective Schrödinger equation numerically.¹⁶

Table 30 shows our preliminary predictions of fission barriers and half-lives of selected transuranic and superheavy nuclei. We find that the previous best estimate of the fission half-life of the nucleus $Z/A = 114/298$ was too optimistic. Instead, we find that the nucleus $Z/A = 126/310$ is quite stable against fission. According to alpha-decay systematics, however,

Figure 71. Main steps of the dynamic deformation model.

the predicted alpha-decay half-life of this nucleus is only 33 μ s. In general, our calculation suggests that the $Z = 126$ isotopes are more stable against fission than the $Z = 114$ isotopes, but are subject to alpha decay.¹⁹ We suggest that

the search for superheavy nuclei be extended to the $Z = 126$ region and focused on the properties of alpha decay.

This work was supported by the LLNL Nuclear Chemistry Research Program.

Table 30. Fission barriers and lifetimes of selected transuranic and superheavy nuclei.

Nucleus A/Z	Fission barrier ^a (MeV)		Spontaneous fission		Half-life		Total (experiment)
	Theory	Experiment	Theory	Experiment	α (theory)	β (theory)	
²⁴⁰ Pu	4.6	6.0 (Ref. 17) 5.6 $\pm$ 0.2 (Ref. 18)	1.15×10^{11} y	1.32×10^{11} y	—	—	6570 y
²⁵⁶ Fm	3.3	—	3.13 h	2.86 h	—	—	2.63 h
²⁵⁸ 104	3.1	—	15 s	—	—	—	11 ms ^b
²⁶⁰ 104	3.0	—	72 ms	76 ms	—	—	20–80 ms
²⁹⁸ 114	3.4	—	0.24 ns	—	1.5×10^{14} y	Stable	—
³¹⁰ 126	9.6	—	7.2×10^{23} y	—	3.3×10^{-5} s	18.8 s	—
³²⁶ 126	6.5	—	2.9×10^8 y	—	1.4×10^{-6} s	10.1 h	—
³⁴⁰ 126	3.3	—	0.020 ns	—	4.0×10^{-3} s	Stable	—
³⁵⁴ 126	0.0	—	1.2×10^{-22} s	—	9.8×10^{-7} s	Stable	—

^a Outer barrier in ²⁴⁰Pu. For others, there is only one barrier.

^b GE Chart of Nuclides, 1977.

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Spontaneous Fission in Neutron-Rich Transfermium Nuclei

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Measurements of fragment energy and mass that we made for ^{259}Md and $^{260}[104]$ indicate a major and abrupt transition in the fission behavior of neutron-rich transfermium nuclei. Fission-fragment measurements for ^{258}No , though made on a very small number of events, tend to support the same conclusion.

In the last 15 years we have learned that nuclear shells have a very broad and pervasive impact on the fission process. In the first few decades after the discovery of nuclear fission, the nucleus was treated as a drop of liquid with smoothly varying attractive and repulsive forces. Although this model still forms the underlying basis for fission, we also observe large effects from the superimposition of shell corrections derived from coupling the quantum states of individual nucleons, as seen in the calculated barriers in Fig. 72. The consequences of single-particle coupling on the fission process can be striking, and often overshadow those originating from the intrinsic liquid-drop component. In past reports, we pointed out several major features attributable to shell effects in the spontaneous fission (SF) of the lighter actinides, the sudden transition to symmetric fission in the fermium isotopes, and finally, new experimental information indicating another transition in the SF of transfermium nuclides due to the disappearance of shell perturbations.¹ In each transition, the abruptness is surprising, and for the moment, such rapid changes in fission behavior lack a theoretical rationale.

There are well-known phenomena associated with shell structure in the fissioning parent, but other notable effects are explained by emerging shells in the fragments. Among these are the fragment-mass and kinetic-energy distributions from spontaneous and neutron-induced fission. While the liquid-drop model of fission allows only a symmetric mass division, fragment shells can induce an asymmetric distribution, which is

Figure 72. Fission barriers calculated with single-particle corrections to the liquid-drop barrier.

a well-known characteristic of fission in the lighter actinides. In the SF of nuclides of these elements, the total kinetic energy (TKE) increases slowly from about 160 MeV to about 200 MeV as Z of the fissioning nuclide increases. A sharp transition to symmetric mass division occurs at ^{258}Fm and ^{259}Fm , and the TKE rises abruptly to about 240 MeV.^{2,3} Both the asymmetric and symmetric mass distributions are qualitatively understood from models in which the static deformation-energy surfaces show that the minimum energy path toward scission is strongly influenced by shells in the nascent fragments and the fissioning nucleus.⁴ We can probably assume that both fragment and fissioning nucleus shells are important since the influence of closed fragment shells is already felt at deformations only slightly greater than the first saddle. However, if an adiabatic equilibrium is maintained in the descent from saddle to scission, the emerging fragment shells will dominate and govern the flow of mass to the final decision point at scission. In this model, the fragment shells steer and direct the mass distribution toward fission products with strong shells. Thus, the very high TKEs and mass symmetry in the fission of ^{258}Fm and ^{259}Fm can be attributed to the preferred formation of fragments near the doubly closed ^{132}Sn nucleus.⁵

Until recently, the mass and kinetic-energy distributions have been measured for only two transfermium nuclides, namely ^{252}No ,^{6,7} and $^{262}[105]$.⁸ We recently completed measurements on the SF properties of 103-min ^{259}Md ,⁹ the third transfermium nuclide to be studied. The mass distribution is highly symmetric and only slightly broader than those of ^{258}Fm and ^{259}Fm . The TKE distribution for ^{259}Md , however, is considerably broader than those of ^{258}Fm and ^{259}Fm , and peaks at 201 MeV, about 35 to 40 MeV lower in energy. The proximity of ^{259}Md to ^{258}Fm and ^{259}Fm (the difference is only one proton or one neutron and proton) strongly suggests that all three nuclides should have similar fission properties. The unusually low fragment energies from ^{259}Md indicate another sharp change in fission behavior after the one undergone in the transition from the light to heavy fermium isotopes. We understand the very high kinetic energies associated with the fission of ^{258}Fm and ^{259}Fm as being due to the high Coulomb energy arising from the compact scission shape of two spherical tin-like fragments. Why ^{259}Md fissions mass-symmetrically into fragments very similar to

those of ^{258}Fm and ^{259}Fm and still releases 35 to 40 MeV less kinetic energy is still very much an unresolved question.

Last year, we reported our preliminary results and tentative conclusions from fragment-mass and energy distributions for the spontaneous fission of 20-ms $^{260}[104]$.¹⁰ Final results¹ showed that the mass distribution was symmetric, but the TKE was only 199 MeV, or some 43 MeV lower than the TKE we had determined for ^{259}Fm , which also fissions mass-symmetrically. Based on the ^{259}Md and early $^{260}[104]$ fragment measurements, we concluded that nuclear fission in this region of heavy nuclei was governed more by the macroscopic features of a liquid drop than by the effects attributed to single-particle couplings, as strongly displayed in the fission of ^{258}Fm and ^{259}Fm . To determine whether these measurements represent another major transition in fission properties rather than the deviate behavior of only two nuclides, we needed additional measurements from other neutron-rich transfermium isotopes. Thus, we recently began measuring the mass and TKE

Figure 73. Schematic of the SWAMI instrument, used to measure coincident fragment energies from the spontaneous fission of millisecond nuclides produced by heavy-ion reactions with actinide-target isotopes. A portion of the recoil nuclei from the nuclear reaction is stopped in 100- $\mu\text{g}/\text{cm}^2$ aluminum foils surrounding the rotating wheel, where they are moved between opposing pairs of surface-barrier detectors.

distributions from fragments emitted in the SF of 1.2-ms ^{258}No .

The Spinning Wheel Analyzer for Millisecond Isotopes (SWAMI), shown in Fig. 73, was specifically designed for these difficult experiments and was first used in obtaining the fission properties of 20-ms $^{260}\text{[104]}$. We are now employing SWAMI and a ^{248}Cm target bombarded with 69.2-MeV ^{13}C ions to produce ^{258}No . The wheel velocity has been increased to 3800 rpm in order to rapidly move the 1.2-ms ^{258}No to and between the opposing Si(Au) detectors. As a result of the much higher wheel velocity, several experimental problems have arisen that are still of some concern. Nevertheless, we observed about 32 fission events, of which 11 were due to the spontaneous fission of 2.6-h ^{256}Fm . The remaining 21 events decayed with a half-life between 1 and 1.5 ms and were produced with an approximately 10-nb cross section. This cross section is at least ten

times lower than that estimated from the Jackson-Sikkeland model for complete fusion reactions, and thus it gives us some uneasiness about the certainty of the original nuclide identification for ^{258}No .¹¹ However, the recoil energy we roughly measured from ranges in aluminum for the 1.0- to 1.5-ms activity corresponds to that calculated for a compound-nucleus product. Based on the net spontaneous-fission events, our first and tentative conclusion is that their most probable TKE is slightly below 200 MeV, and the mass distribution, as estimated from the narrow half-width, is most likely symmetric. If we are observing fission from ^{258}No , then it is quite clear that fission in the transfermium region has shifted abruptly from strong fragment-controlled fission in the heaviest fermium isotopes to fission governed by the macroscopic forces of the nucleus.

This research was supported by the LLNL Nuclear Chemistry Research Program.

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Energy Dependence of Fission-Fragment Angular Distribution for Reactions Induced by ^{19}F , ^{24}Mg , ^{28}Si , and ^{32}S on ^{208}Pb

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We measured the energy dependence of fission-fragment angular distributions for reactions induced by ^{19}F , ^{24}Mg , ^{28}Si , and ^{32}S on ^{208}Pb over the range of incident energies from $E/A = 5.6$ to 10 MeV/amu. For all four systems, the angular distributions are inconsistent with the saddle-point deformations of the rotating-liquid-drop model.

During the past year, we continued to explore the properties of fissioning nuclei at high angular momentum and excitation energy.³ In particular, we measured the angular distributions of the fission fragments from reactions induced by the heavy ions ^{19}F , ^{24}Mg , ^{28}Si , and ^{32}S on ^{208}Pb . These experiments were carried out to investigate the range of validity of the standard transition state theory for fission⁴ and of the equilibrium shapes predicted by the rotating liquid-drop model (RLDM),⁵ and to distinguish fast fission from normal compound nucleus fission. The term fast fission is used to designate fission occurring from values of angular momentum greater than the critical angular momentum $J_{Bf} = 0$ at which the fission barrier disappears. $J_{Bf} = 0$ is usually taken from the RLDM.

The angular distribution data was analyzed using the transition state theory for nuclear fission to obtain information on the moments of inertia or shape of the fissioning nucleus. These experimentally deduced shapes were compared with predictions of models such as the RLDM. In recent measurements of ^{32}S induced reactions on heavy targets,⁶ the angular distributions indicated symmetric fission of composite nuclei that were highly deformed. The RLDM, on the other hand, predicts that in the same mass range, the fissioning nucleus is almost spherical at the saddle point. Earlier studies of ^{16}O -induced reactions⁷ on similar targets showed qualitative agreement with the predictions of the

RLDM. In order to explain this discrepancy, it has been suggested⁸ that fast fission may be occurring in the ^{32}S induced reactions but not in the ^{16}O induced reactions. Our experiments were designed to explore this possibility.

We made our measurements at the superconducting linear postaccelerator at Argonne National Laboratory (Argonne, IL) over the range of incident energies from $E/A = 5.6$ to 10 MeV/amu.^{9,10} In these reactions, we sampled fissioning systems formed with up to $120\hbar$ units of angular momentum and with excitation energies of 50 to 120 MeV. Cross sections of fission fragments and elastic scattering were obtained at laboratory angles corresponding to a center-of-mass angular range of 20° to 176° . Contributions from sequential fission following deeply inelastic reactions were eliminated by requiring kinematic coincidences of the two fission fragments. The absolute normalization of the angle integrated cross sections is believed to be accurate to $\pm 7\%$. We analyzed the data to determine the range of angular momenta contributing to the fission process and to deduce information on shape for comparison with the predictions of existing models of rotating nuclei.

The angle-integrated fission cross section for each energy was used to calculate the distribution of angular momenta leading to fission at that energy. We did this by assuming that all entrance channel partial waves ℓ leading to fusion result in fission of a compound system with $J = \ell$. This is a good approximation because

the fission barriers in this mass region are considerably lower than the neutron binding energy, even at low J . In the sharp-cutoff (SCO) model, which is commonly used in these analyses, it is assumed that all ℓ waves of less than a critical value ℓ_{crit} lead to fusion and hence to fission, and that all partial waves with $\ell > \ell_{\text{crit}}$ do not. Thus the partial cross section σ_ℓ for each ℓ can be written as

$$\begin{aligned} \sigma_\ell &= \pi\lambda^2 (2\ell + 1) \quad \text{for } \ell \leq \ell_{\text{crit}} \\ \sigma_\ell &= 0 \quad \text{for } \ell > \ell_{\text{crit}} \end{aligned} \quad (1)$$

where $\lambda^2 = \hbar^2/2mE_b$ and E_b is the bombarding energy. Then the total fission cross section σ_f is given by

$$\sigma_f = \pi\lambda^2 \sum_{\ell=0}^{\ell_{\text{crit}}} (2\ell + 1) = \pi\lambda^2 (\ell_{\text{crit}} + 1)^2 \quad (2)$$

Using this approximation, we obtain values of ℓ_{crit} for each system that range from well below (at the lowest energies) to well above (at the highest energies) the value of $J_{B_f} = 0$ predicted from the RLDM. For energies corresponding to $\ell_{\text{crit}} < J_{B_f} = 0$, we expect our data to be characteristic of compound-nucleus fission.³

In order to examine the uncertainties introduced by using the SCO approximation, we also did the analysis using the optical model with parameters chosen to fit the elastic-scattering data. This model, in contrast to the SCO model, gives a very broad distribution of partial cross sections with the same total fission yield. The actual distribution is probably

Figure 74. Partial-wave capture cross sections obtained from the sharp-cutoff model and optical model for $^{32}\text{S} + ^{208}\text{Pb}$ at 180 MeV.

somewhere between these two extremes.

Figure 74 shows ℓ distributions obtained from the SCO model and the optical model for $^{32}\text{S} + ^{208}\text{Pb}$ at 180 MeV.

With the angular-momentum distribution leading to fission given by either of these two models, we analyzed the angular distributions using transition-state theory.¹¹ Within the framework of the transition-state theory of fission, the direction of fission is fixed by $\mathbf{K}$, the projection of the angular-momentum vector $\mathbf{J}$ on the nuclear symmetry axis. The quantum number K is assumed to be conserved in the passage of the nucleus from its transition state to the configuration of the separated fragments. The angular distribution is obtained by summing over the distributions of J and K , giving the following expression for the angular distribution $W(\theta)$:

$$W(\theta) \propto \sum_J T_\ell(\epsilon) \sum_K \rho_t(E_t, K) P_f^K(\theta) \quad (3)$$

where $T_\ell(\epsilon)$ is the transmission coefficient for formation of the transition state with energy ϵ in the fission mode and entrance-channel angular momentum ℓ ; $\rho_t(E_t, K)$ is the density of levels at the transition state with energy $E_t = E^* - B_f - E_{\text{rot}}$ (where E^* is the excitation energy of the composite system, B_f is the barrier height, and E_{rot} is the energy tied up in rotation); and $P_f^K(\theta)$ is the probability distribution for fission at angle θ . Using a constant temperature approximation, the level density at the saddle point is given by $\rho_t(E_t, K) \propto e^{E_t/T}$. The nuclear temperature T is defined by $E_t = aT^2$, where a is a constant. With this simplifying assumption, the quantum number K is described by a Gaussian distribution of width K_0^2 . This width can be related to the shape of the nucleus at the transition state by

$$K_0^2 = \mathcal{J}_{\text{eff}} T / \hbar^2 \quad (4)$$

where $\mathcal{J}_{\text{eff}}$ is the effective moment of inertia, defined by

$$\mathcal{J}_{\text{eff}}^{-1} = \mathcal{J}_{\parallel}^{-1} - \mathcal{J}_{\perp}^{-1} \quad (5)$$

$\mathcal{J}_{\parallel}$ and $\mathcal{J}_{\perp}$ are the moments of inertia parallel and perpendicular to the symmetry axis, respectively. The probability distribution $P_f^K(\theta)$ can be calculated exactly in a quantum treatment, or it can be treated classically for high angular momenta. We used the quantum treatment.

Figure 75 shows the measured angular distributions of fission fragments. All angular distributions exhibit a deviation from the $1/\sin\theta$ behavior which would be expected if the angular-momentum vector were aligned exactly perpendicular to the beam axis ($K = 0$). Moreover, they do not show the isotropic behavior that would be expected if the angular-momentum vector could lie with equal probability in any direction. Since the deviation from the $1/\sin\theta$ behavior is evident only at angles near forward or backward beam

directions, it is convenient to describe the shape of the angular distribution in terms of an anisotropy parameter, the ratio of the cross section $W(\theta)$ at 0° to that at 90° .

We observed the following qualitative trends in the data:

1. For all systems, the anisotropy increases with increasing bombarding energy.
2. The anisotropy increases with the mass of the projectile.

The first trend is in qualitative agreement with the predictions of the RLDM; the second trend,

Figure 75. Fission-fragment angular distributions in the center-of-mass system, measured for reactions induced by ^{19}F , ^{24}Mg , and ^{28}Si on ^{208}Pb . Solid curves are obtained from global fits of Eq. (6). Dashed lines show the classical limit of the angular distributions corresponding to $d\sigma/d\Omega = 1/\sin\theta$.

however, is contrary to expectations, based on the RLDM, that the saddle-point shape becomes more spherical with increasing mass.

It has been customary to fit data on angular distributions using a constant value of K_0 . This procedure has been used extensively for light-ion-induced reactions, where the composite system has low angular momentum and excitation energy. It can be seen from Eq. (4) that a constant K_0 implies that both T and $\mathcal{J}_{\text{eff}}$ are independent of J . It is predicted by the RLDM, however, that the fission barrier B_f , the rotational energy E_{rot} , and the effective moment of inertia $\mathcal{J}_{\text{eff}}$ are all functions of the angular momentum. Qualitatively, as the fission barrier approaches zero, the rotating nucleus becomes increasingly spherical, and $\mathcal{J}_{\text{eff}}$ and K_0 approach infinity.

We modified the fitting process by choosing a functional form for $K_0(J)$ to fit the angular distribution at each energy for a given system. Since $T(J)$ varies slowly, we assumed a constant value for each energy. Then we chose a form for

$\mathcal{J}_0/\mathcal{J}_{\text{eff}}$, where $\mathcal{J}_0$ is the rigid-body moment of inertia of an equivalent sphere, such that:

$$\begin{aligned} \mathcal{J}_0/\mathcal{J}_{\text{eff}} &= a - bJ^2 \quad \text{for } J^2 \leq (a - c)/b \\ &= c \quad \text{for } J^2 > (a - c)/b \end{aligned} \quad (6)$$

The coefficients a , b , and c were determined for each nucleus to give the best fit to the angular-distribution data. These fits are shown by the solid curves in Fig. 75. Figure 76 shows the best-fit values of $\mathcal{J}_0/\mathcal{J}_{\text{eff}}$. The extracted values of $\mathcal{J}_0/\mathcal{J}_{\text{eff}}$ are in clear disagreement with the saddle-point shapes of the RLDM.

Although we have not completed our analysis of the results, we can make the following tentative conclusions:

1. At low angular momenta, the moments of inertia increase with increasing angular momentum. This is in qualitative agreement with the RLDM. Quantitative agreement was not obtained in any of the systems, however. The agreement is improved at low energies by using

Figure 76. Dependence of $\mathcal{J}_0/\mathcal{J}_{\text{eff}}$ on angular momentum J . $\mathcal{J}_0/\mathcal{J}_{\text{eff}}$ is extracted from analysis of the angular distributions. Dashed and dotted lines result from the global fits of Eq. (6), for the optical model and sharp-cutoff model, respectively. Bars correspond to fits with a constant value for K_0 at each energy; the estimated errors for these fits are indicated by the vertical bars, and the lengths of the horizontal bars correspond to the range of angular momenta contributing to the fission cross sections. Solid curves show the effective moments of inertia predicted by the rotating-liquid-drop model (RLDM).

ℓ distributions obtained from the optical model instead of the SCO. However, because the moments of inertia extracted from the low-energy data are so sensitive to the choice of ℓ distribution (extracted K_0 is 15% larger when the ℓ distribution from the optical model rather than the SCO model is used), the quantitative results at the lowest energies are not reliable.

2. At high angular momenta, the effective moments of inertia reach asymptotic values greater than 0.6. This indicates that the fissioning systems are considerably deformed.

3. The moments of inertia extracted from the angular distributions are relatively insensitive to the mass of the fissioning system. This is contrary to the predictions of the RLDM.

4. The moments of inertia extracted from the data are not in agreement with the predictions of the RLDM, even at low energy. These deviations must be understood before we can determine whether we are seeing the onset of fast fission as we raise the energy in these systems.

The interpretation of the large deformations obtained for the heavy fissioning nuclei at high angular momenta is at present open. It is not possible through our set of experiments to determine whether the large deformations result from an entrance-channel effect or from a property of the composite system. If the transition state at which the K distribution for fission is frozen is not reached until some point

on the path between the saddle point and the scission point, then the angular distributions might still be understood within the statistical transition-state theory. One test of this possibility would be to measure angular distributions from the fission of a heavy compound system formed by different entrance channels.

Another open question is the interpretation of the deviations from the liquid-drop shapes at low energy. Recent gamma-ray measurements of $^{16}\text{O} + ^{154}\text{Sm}$ at energies close to the Coulomb barrier¹² implied a broad ℓ distribution similar to that obtained from the optical model. If this is the case, one would expect a better agreement with the shapes predicted by the RLDM. Discrepancies would still exist, however. It is also possible, at the lowest energies, if shell effects are not completely washed out, that the theoretical saddle point would be more deformed than the RLDM prediction.

In the future we plan to investigate further the systematics of the energy and angular-momentum dependence of the extracted moments of inertia. In addition, we plan to refine the global fits of Fig. 76 to include the effects of temperature and of possible prefission evaporation of neutrons.

This work was supported by the LLNL Nuclear Test Program.

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Fission of Hot Rotating Nuclei: Implications of Heavy-Ion Experiments

M. A. McMahan and M. G. Mustafa

We compared moments of inertia extracted from fission-fragment angular-distribution data with the predictions of macroscopic fission models that include angular momentum and temperature. The energy dependence of the extracted moments of inertia is linear and relatively constant for composite systems in the range $Z^2/A = 33$ to 40. This dependence cannot be explained by existing macroscopic models, which calculate shapes at either the saddle point or the scission point.

In order to further our understanding of the roles of temperature and angular momentum in fission, we deduced moments of inertia and their energy dependence from fission-fragment angular-distribution data for a variety of heavy-ion reactions, and compared these results with the predictions of macroscopic fission models that include angular momentum and temperature. We studied the reactions of ^{11}B , ^{12}C , ^{14}N , and ^{16}O on ^{209}Bi , and ^{19}F , ^{24}Mg , ^{28}Si , and ^{32}S on ^{208}Pb at various incident energies. For the lighter projectiles in this series ($A < 14$), experimentally deduced moments of inertia reported in the literature¹⁻³ were consistent with a liquid-drop picture of fission.⁴ For the heavier projectiles, however, decided discrepancies from the rotating-liquid-drop model (RLDM)⁴ have been reported.^{5,6} By comparing these systematic results with a modified rotating-liquid-drop model (MRLDM)⁷ and a scission model^{8,9} of fission, we hope to establish the extent to which the observed fission properties of heavy systems at high angular momenta and temperatures can be explained in terms of macroscopic models of fission.

The extraction of moments of inertia from the measured fission-fragment angular distributions is done by invoking the transition-state theory as applied to fission by Bohr and Wheeler.¹⁰ According to transition-state theory,¹¹ there exists an equilibrated complex described by the quantum numbers J , the angular momentum, and K , the projection of the angular momentum on the symmetry axis of the composite system. The quantum number K is assumed to be conserved from this transition state through

scission, and the subsequent fission process can be described statistically in terms of J and K at the transition point. It is often further assumed that (1) the transition state corresponds to the saddle point for fission, and (2) the K distribution at the transition point is Gaussian. Then, knowing or assuming the distribution of J leading to the compound system, the measured angular distributions give a value for the width, K_0^2 , of the K distribution. K_0^2 depends on the shape and temperature of the fissioning system at its saddle point through the expression

$$K_0^2 = \mathcal{J}_{\text{eff}} T / h^2, \quad (1)$$

where

$$1/\mathcal{J}_{\text{eff}} = 1/\mathcal{J}_{\parallel} - 1/\mathcal{J}_{\perp}. \quad (2)$$

The details of the data analysis are given in Ref. 12. For the present study, we incorporated the following prescription for obtaining values of K_0^2 from the experimental data.

1. For all cases, we assumed a sharp cutoff for the J distribution leading to fusion. For the cases of ^{19}F -, ^{24}Mg -, ^{28}Si -, and ^{32}S -induced reactions, this was calculated from the integrated total fission cross section. For the other cases, we used the values given in Ref. 13.

2. For the lighter systems (^{11}B , ^{12}C), we assumed a minimum angular momentum for fission based on estimates of evaporation-residue cross sections.¹³

3. Knowing $J_{\min}$ and $J_{\max}$, we calculated $\langle J \rangle$ and thus obtained $\langle K_0^2 \rangle$, the width of the K distribution averaged over J . A more consistent

method of analysis would fit the angular distributions to a distribution $K_0(J)$. This was done for the heavier projectiles in this series^{14,15} and had only a small effect on the extracted values of $\langle K_0^2 \rangle$. Our results are shown as a function of the thermal energy available for fission at the transition state, E_t , given by

$$E_t = E^* - B_f(\langle J \rangle) - E_{\text{rot}}(\langle J \rangle) \quad (3)$$

where E^* is the excitation energy, $B_f(\langle J \rangle)$, and $E_{\text{rot}}(\langle J \rangle)$ are the fission barrier and rotational energy, respectively, at the average angular momentum of the fissioning system. B_f and E_{rot} are calculated with the MRLDM. The temperature T at the saddle point is calculated using the relation

$$E_t = a_f T^2 \quad (4)$$

where a_f is taken to be $1.1A/8.5$. We ignore neutron evaporation from the composite system, which lowers the temperature of the fissioning system. This was investigated for the case of $^{19}\text{F} + ^{208}\text{Pb}$ and found not to be an important factor.

Figure 77. K_0^2 deduced from experimental fission-fragment angular distributions for the reactions ^{11}B , ^{12}C , ^{14}N , and ^{16}O on ^{209}Bi .^{2,3} Black lines are a linear fit to the data, excluding points within 10% of the Coulomb barrier. Orange and green lines are predictions of the MRLDM, with and without J dependence of the moments of inertia, respectively.

Figures 77 and 78 show as points the values of K_0^2 obtained with the above prescription. Several qualitative features can be noted in the figure about the experimental results:

1. For light systems, K_0^2 is a linear function of the available energy E_t . The solid lines in the figure show the best fit to a straight line. This already indicates a partial breakdown of transition-state theory, in which K_0^2 is a linear function of T , or $\sqrt{E_t}$.

2. For ^{32}S , K_0^2 approaches an asymptotic value at high energies.

3. The range of values of K_0^2 is approximately constant for the whole series of projectiles.

4. The data taken at energies less than about 10% above the Coulomb barrier show K_0^2 values lower than expected from the systematics.

Can these features of the systematic results be explained within the framework of existing macroscopic rotating nuclear models for fission? To answer this question, we looked at two possible models of the fissioning system: the MRLDM (the rotating-liquid-drop model with finite range⁷), and a scission model.⁸ We did not do any calculations using the RLDM because of the lack of availability of the program. It is believed, however, that the MRLDM is a more realistic model.^{7,16,17}

In the MRLDM, the total energy is calculated as the sum of Coulomb, E_C , surface, E_s , and rotational, E_{rot} , energies, and the minimum path on the potential-energy surface (PES) determines the saddle-point configuration. We can analytically calculate $\mathcal{J}_{\parallel}$ and $\mathcal{J}_{\perp}$ for the shape at the saddle point. This model was used in the past primarily to look at fission-barrier heights. The calculation of shapes, on the other hand, is quite sensitive to the curvature of the PES. Because of the flatness of the PES, the errors in the calculated $\mathcal{J}_0/\mathcal{J}_{\text{eff}}$ are about 10%. Also, because of the effect of finite range, this model calculates values of $\mathcal{J}_0/\mathcal{J}_{\text{eff}}$ about 10% smaller than the standard RLDM.

Figure 77 shows the comparison between the experimentally deduced K_0^2 and the predictions of the MRLDM for the reactions ^{11}B , ^{12}C , ^{14}N , and ^{16}O on ^{209}Bi . Theoretical predictions are included for $\mathcal{J}_0/\mathcal{J}_{\text{eff}}$ independent of J (green) and $\mathcal{J}_0/\mathcal{J}_{\text{eff}}(J)$ (orange). The first condition, i.e., $\mathcal{J}_0/\mathcal{J}_{\text{eff}}(J) = \mathcal{J}_0/\mathcal{J}_{\text{eff}}(0)$, was included because this was the way most previous analyses were done. With this assumption, the calculated values of K_0^2 are a linear function of T (or $E_t^{1/2}$). For the more realistic assumption that $\mathcal{J}_0/\mathcal{J}_{\text{eff}}$ is a function of the angular momentum, K_0^2 increases rapidly to infinity at high angular momenta. For the lighter systems of Fig. 77, K_0^2 remains approximately

linear, and using $\mathcal{J}_0/\mathcal{J}_{\text{eff}}(J)$ gives a reasonable fit at all energies except those close to the Coulomb barrier.

Figure 78 shows similar comparisons for the heavier systems ^{19}F , ^{24}Mg , ^{28}Si , and ^{32}S on ^{208}Pb . Here the breakdown of the theoretical predictions is clearly demonstrated. The data simply do not follow the trend predicted by theory, for as Z^2/A increases, the shapes become more compact.

It is clear that the theory, with or without the effects of angular momentum on shapes, is not adequate to explain the trends of the data. We looked into possible modifications within the liquid-drop model that might explain the effects. One such possibility is the effect of temperature on the fissioning system. To explore this effect, we utilized the formalism of Hasse and Stocker¹⁸ to include correction terms to the Coulomb, surface, and rotational energies, and calculated $B_f(J,T)$ and $\mathcal{J}_0/\mathcal{J}_{\text{eff}}(J,T)$ for the nucleus ^{240}Cf ($^{32}\text{S} + ^{208}\text{Pb}$) at T from 0 to 2 MeV. We note that the effective deformation energy at a given temperature T is the free energy. The qualitative effect of temperature is to decrease both the surface and Coulomb energies, with the overall result of decreasing the fission barrier and moving the saddle point to more compact shapes. Over the range of temperatures calculated, the fission barrier and $\mathcal{J}_0/\mathcal{J}_{\text{eff}}$ were both lowered by about 50%. Because this effect is in the same direction as angular-momentum effects, it does not improve the fit with experiment.

It has been suggested^{8,19} that at high energies, fission occurs fast enough that the K distribution is not frozen until further along the path to scission. If that is the case, we could expect the data to be bracketed by the shape at the saddle point and the shape at the scission point. To explore this possibility, we calculated the shape at the scission point for the cases ^{19}F , ^{24}Mg , ^{28}Si , and ^{32}S on ^{208}Pb . The scission point is modeled as two symmetric touching ellipsoids, and the shapes of the ellipsoids are obtained by minimizing the total energy. We calculated moments of inertia from these shapes and obtained K_0^2 using T at the scission point. These values of K_0^2 are shown as the blue lines in Fig. 78. It can be seen that this provides a lower limit to the data, but again, it cannot explain the energy dependence.

The general conclusion of our study is that the macroscopic rotating-nucleus models for fission

are not adequate to explain the angular distributions of fission fragments and their energy dependence. It has, however, been suggested that by studying the deviations of angular-distribution data from RLDM predictions at high energies, insight could be gained on the fission process at angular momenta for which there is no longer a fission barrier. Our results show that this is not feasible because the systematics are not explained by the RLDM, even at angular momenta where the model is thought to be valid. In plots of K_0^2 vs E_t for these systems, there are no discontinuities evident which might indicate that a new process is becoming important for either high angular momenta or heavy systems (except possibly in the asymptotic behavior of $^{32}\text{S} + ^{208}\text{Pb}$ at high energies).

We should add, however, that the basic assumption of the transition-state theory may be in question here. It is possible that axial symmetry is broken in the motion from saddle

Figure 78. K_0^2 deduced from experimental fission-fragment angular distributions for the reactions ^{19}F , ^{24}Mg , ^{28}Si , and ^{32}S on ^{208}Pb .^{14,15} Orange and green lines are predictions of the MRLDM, with and without J dependence of the moments of inertia, respectively. Blue lines indicate predictions of a scission model for fission.

to scission, leaving K undefined. It may also be the case that the angular distributions are determined by entrance-channel effects. These questions are still to be answered.

We are presently studying the role of shell effects on rotating nuclear shapes as a function of the nuclear temperature T . We hope this

will provide some insight into the deviations of the angular distributions from the predictions of the MRLDM at energies close to the Coulomb barrier.

This research was supported by the LLNL Nuclear Chemistry Research Program.

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Fission-Product Mass Yield, Thermal-Neutron Fission Cross Section, and $\bar{\nu}$ of the Long-Lived ^{236}Np Isomer

M. Lindner and D. Seegmiller¹

We measured the fission-product distribution in long-lived ^{236}Np irradiated with thermal neutrons in the LLNL Pool-Type Reactor, both by absolute gamma-ray counting and by comparison with the same fission product produced simultaneously in ^{235}U . The asymmetric distribution is similar to that of ^{239}Pu , especially for the heavy-mass peak. The average number of neutrons emitted per fission ($\bar{\nu}$) was 2.4 ± 0.8 . The thermal-fission cross section was 1.90 ± 0.14 kb when measured by comparison with thermal ^{235}U fission, and 1.74 ± 0.05 kb by comparison with ^{237}Np (n, γ) ^{238}Np .

The thermal-fission cross section of long-lived ^{236}Np has long been known to be several kilobarns.² Until recently, however, nothing was reported on the distribution of yields of the thermal-neutron fission products. In 1980 Gindler *et al.*³ published such a distribution, obtained by performing beta- and gamma-ray counting on chemically separated samples after thermal-neutron irradiation of a neptunium sample containing 20% ^{236}Np .

We performed a similar irradiation,⁴ but our measurements were made quite differently. Consequently, our results, though similar, differ markedly in details, partly due to our much smaller uncertainties, and partly to the approach to the measurements.

In 1975 we irradiated 4 ng of ^{236}Np ($^{236}\text{Np}/^{237}\text{Np} \approx 4$) in the LLNL Pool-Type Reactor for 7 h, together with 60 ng of $>99\%$ ^{235}U . Each fissionable isotope was mounted between two 5-mil wafers of ultrapure graphite to minimize background effects. Several graphite wafers containing no target material were also irradiated so that background effects could be measured. Immediately after irradiation, gamma-ray spectra were accumulated from all three targets on a Ge(Li) detector whose output (0–1 MeV) was stored in a 4096-channel pulse analyzer. The data were analyzed with the gamma-ray photopeak shape-fitting code of Gunnink and Niday.⁵ About 30 spectra were

obtained for each target at selected time intervals for a period of about 1 year.

Using that technique, we were able to identify the gamma-ray photopeak of a specified fission product with its proper half-life. From decay-curve analysis, we found that interferences

Figure 79. R values for thermal-neutron fission.

among fission products varied from nonexistent to severe for a selected gamma-ray photopeak. Background interferences were trivial. From the above analyses, the R values for 30 fission products were calculated from the expression

$$R = \left(\frac{X}{\text{Mo}} \right)_{\text{Np}} / \left(\frac{X}{\text{Mo}} \right)_{\text{U}_{\text{th}}} , \quad (1)$$

where X is the zero-time activity of a specified photopeak measured on a specified detector, for both ^{236}Np and ^{235}U ; Mo is the analogous quantity for the reference isotope ^{99}Mo (arbitrarily chosen); and U_{th} pertains to fission of ^{235}U with thermal neutrons. We then calculated the *relative* fission yield from the expression

$$\text{Rel. yld. } (X)_{\text{Np}} = R[Y_f(X)/Y_f(\text{Mo})]_{\text{U}_{\text{th}}} . \quad (2)$$

The absolute fission yields Y_f for U_{th} were taken from the recent compilation of Rider.⁶ The absolute fission-chain yields for ^{236}Np fission products were obtained from the relationship

$$Y_f(X)_{\text{Np}} = \frac{\text{Rel. yld. } (X)_{\text{Np}}}{\sum_i \text{Rel. ylds. } (X_i)_{\text{Np}}} , \quad (3)$$

in which the denominator is summed over all mass numbers, including interpolated as well as observed values.

Figure 79 shows the mass dependence of the observed R values. For purposes of comparison, the R values for ^{239}Pu calculated from Rider⁶ are also shown. Since the R values may be

Figure 80. ^{236}Np fission-product mass-yield distribution.

interpreted as "shape factors," it is clear that the heavy mass peaks for ^{236}Np and ^{239}Pu are nearly identical.

Figure 80 shows the absolute fission-chain yields derived from Eq. (3). Multiple values at a given mass number are usually the result of determining more than one gamma-ray photopeak, but occasionally they result from adjacent isobar measurements. Also shown in Fig. 80 are the absolute fission yields derived from interpretation of the absolute counting of the gamma-ray photopeaks, together with published data on decay schemes.⁷⁻⁹ While the agreement between the two methods is generally good, there are instances of disagreement attributable to the absolute-counting measurements.

One measure of confidence in the "asymmetrical" distribution in Fig. 80 is the extent to which the areas under the light- and heavy-mass peaks agree. We found that these areas agreed within less than 1% when the R -value method was used.

We attempted to derive, from the mass-yield distribution of Fig. 80, a value for the true average number of neutrons, $\bar{\nu}$, emitted per fission of the compound nucleus ^{237}Np :

$$\bar{\nu} = 237 - [\langle M \rangle_L + \langle M \rangle_H] = 2.4 \pm 0.8 , \quad (4)$$

where the average mass $\langle M \rangle_L$ is given by

$$\langle M \rangle_L = \left[\sum_{M_L} (Y_f)_L M_L \right] / \sum_{M_L} (Y_f)_L . \quad (5)$$

Here, M_L is the mass of the light-mass product and $(Y_f)_L$ is its fission yield. A similar expression holds for M_H , the heavy mass. We attribute the large uncertainty in this value to conservatism in assigning uncertainties to the interpolated values of unmeasured fission products.

We also calculated the thermal-fission cross section for long-lived ^{236}Np by two methods.

Table 31. Thermal-fission cross section of ^{236}Np .

Monitor reaction	Thermal cross section (10^{-21} cm^2)	
	Monitor	$^{236}\text{Np}(n,f)$
$^{235}\text{U}(n,f)$	0.480	1.90 ± 0.14
$^{237}\text{Np}(n,\gamma)$	0.180	1.74 ± 0.05

The first compares the fissions of ^{236}Np with those of the "external monitor" ^{235}U , while the second compares the ^{236}Np fissions with the capture reaction $^{237}\text{Np}(n,\gamma)^{238}\text{Np}$ from the

"internal monitor" ^{237}Np in the sample. Table 31 summarizes the results.

This research was supported by the LLNL Nuclear Chemistry Research Program.

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Measurement of the ^{242m}Am Neutron-Induced Cross Section

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We measured the fission cross section of ^{242m}Am from 0.005 eV to 20 MeV using time-of-flight techniques at the Oak Ridge Electron Linear Accelerator. To detect fission events, we used a hemispherical-plate ionization chamber of original design that contained 507 μg of ^{242m}Am , 168 μg of ^{235}U as a cross-section standard, and ^{252}Cf as a monitor of chamber performance. In addition to the ^{235}U , which served as a cross-section standard above 101 keV, the $^6\text{Li}(n,\alpha)$ reaction served as a shape standard below 101 keV. We obtained about 360 h of data at a flight-path distance of 9.1 m, primarily with 40-ns bursts. Particular attention was paid to correcting for the effect of neutrons scattered by the materials of the ionization-chamber stack. We compare the results with other measurements of the ^{242m}Am fission cross section and resonance integral.

The fission cross section of ^{242m}Am , an isomer with $t_{1/2} = 141 \pm 2 \text{ y}$,² is of interest in reactor design, in studies of plutonium recycling, and in the design of spent-fuel reprocessing and storage facilities. Measurements of the fission cross section have been difficult because of the rareness of the isotope. Due to its large thermal-neutron fission cross section (7000 b), this nuclide rapidly reaches an equilibrium level relative to its ^{241}Am progenitor when in a thermalized reactor neutron flux; the equilibrium atom ratio, $^{242m}\text{Am}/^{241}\text{Am}$, is 0.01. Thus, isotopic enrichment is required in order to obtain samples of sufficient purity for use in precise cross-section determinations. Enriched samples of several hundred micrograms, and containing more than 99% ^{242m}Am , such as were used in this measurement, have been available since 1976. Another problem in making cross-section measurements is caused by the rapid growth of the ^{242}Cm beta-decay daughter ($t_{1/2} = 162.8 \text{ d}$), which decays by alpha emission and spontaneous fission. This nuclide can be removed by chemical separation, and care must be taken to proceed with the measurement as rapidly as possible after the separation.

Earlier determinations of the fission cross section of ^{242m}Am included measurements with samples consisting of 20% ^{242m}Am ; the neutron

source was either a U. S. nuclear explosion³ or the LLNL Electron Linear Accelerator.⁴ More recently, with samples available consisting of more than 99% ^{242m}Am , the fission cross-section measurement was repeated at the LLNL Electron Linear Accelerator.⁵ A Russian measurement with an underground nuclear explosion covered the neutron energy range 0.04 to 4.52 MeV.⁶

We measured the fission cross section of ^{242m}Am from 0.005 eV to 20 MeV using time-of-flight techniques at the Electron Linear Accelerator at Oak Ridge National Laboratory (ORNL), Oak Ridge, TN. A new type of fission-ionization chamber, one that involves the use of hemispherical plates, was developed at ORNL for the specific purpose of permitting fission measurements on intensely alpha-active isotopes.⁷ Figure 81 shows a cross section of one unit in the ionization-chamber stack. The chamber provides for good discrimination between fission pulses and interference from alpha-pulse pileup, on the basis of a "limited transverse range" principle. The upper hemishell stops particles that originate in the target deposit and travel at large angles to the surface normal. All or a large fraction of the ionization from a fission fragment is deposited in the gas of the ionization chamber, whereas only a small fraction of the range of ionization of an alpha

Figure 81. Cross-sectional view of the hemispherical fission-chamber plates. All dimensions are in centimetres. The ratio l_2/l_1 is constant and equal to 3 in this design. The chamber consists of a stack of five pairs of plates.

particle is deposited in the gas. The major part of the alpha-particle energy is deposited when these particles intercept the upper stainless steel hemishell.

The ionization chamber consisted of a stack of five pairs of plates and contained three deposits totaling 507 μg of $^{242\text{m}}\text{Am}$, one deposit of 168 μg of ^{235}U , and a "weightless" deposit of ^{252}Cf , which served as a monitor of chamber performance. The chamber was mounted 9.1 m from the source of the pulsed neutron beam, i.e., from the center of the tantalum electron target. The fission of ^{235}U served as the cross-section standard for energies above 101 eV, while the $^6\text{Li}(n,\alpha)$ reaction cross section, normalized to ^{235}U fission in the 7.8- to 11.0-eV interval, served as a shape standard below 101 keV. Approximately 360 h of data were obtained, primarily with 40-ns bursts.

We paid particular attention to the correction of backgrounds, especially from neutrons scattered by the materials of the ionization chamber, e.g., chamber gas and windows, and target materials and backings. Since the stack of scatterers and targets was rather compact, neutrons scattered from components both ahead of and behind the target had a nontrivial probability of striking the target and causing a fission event. In order to make these corrections,

we performed detailed calculations to model neutron-scattering effects. Reference 8 contains a detailed description of this experiment and the results.

Figure 82 summarizes the results, and for comparison, it also shows the ENDF/B-V fission cross section for $^{242\text{m}}\text{Am}$ (Ref. 9) and the calculated background from scattered neutrons (shown as an equivalent cross section). A technique of variable data averaging was used to produce the plotted data, i.e., all points were shown over peaks, whereas data were averaged over many points in a relatively smooth region. Above 101 keV, where our data were based on the ENDF/B-V values for ^{235}U (Ref. 9), agreement in shape between our measurement and that of Browne *et al.*⁵ is good. Table 32 compares averaged cross sections with those from ENDF/B-V and the recent Russian work.⁶ We do not confirm a reported dip in cross section near 2 MeV.⁶ Below 101 keV, where our data are based on the shape of the $^6\text{Li}(n,\alpha)$ cross section, good agreement with previous measurements is found. In particular, agreement with the accepted thermal-fission cross section is good, and agreement in shape below 10 eV with the results of Browne *et al.*⁵ is excellent.

Our measurement spans the energy range of all previous work and confirms the

Figure 82. ^{242m}Am fission cross section as a function of neutron energy.

Table 32. Comparison of cross-section data for ^{242m}Am .

Measured quantity	Energy	Present measurement (b)	Previous measurements (b)	Difference (%)	Reference, previous work
σ_{nf}	0.0253 eV	6950 ± 250	6600 ± 300	-5	4
σ_{nf}	0-10 eV	(Fig. 82)	(Fig. 82)	-7	5
σ_{nf}	0.1-20 MeV	(Fig. 82)	(Fig. 82)	-15	5
σ_{nf}	1-20 MeV	2.567	2.441	-5	3,4,9
σ_{nf}	1-4.5 MeV	2.183	1.606	-26	6
I_f^a	0.5 eV to ∞	1800 ± 65	1570 ± 110	-13	11
I_f	0.52 eV to ∞	1750 ± 65	2260 ± 200	+29	12

^a I_f is the fission resonance integral.

measurements reported by Browne *et al.*,⁵ although a detailed comparison of the two data sets awaits the availability of the final results from Ref. 5. A file containing our experimental data (in EXFOR format) has been deposited with the National Nuclear Data Center.¹³

This work was supported by the LLNL Nuclear Chemistry Research and Nuclear Test Programs.

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Measurement of the Electrochemical Reduction of the Element Nobelium

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Using radiocoulometry, we determined the half-wave reduction and amalgamation potential of divalent nobelium in the presence of both weak (acetate) and strong (citrate) complexing ions. A comparison with the reduction potentials of the heavier actinides and lanthanides shows that the divalent oxidation state becomes increasingly stable as the atomic number increases.

One of the most important features of the heavier actinides is the increasing stability of the divalent oxidation state with increasing atomic number. The reduction potential of the trivalent to divalent oxidation state drops from +1.55 V for einsteinium to -1.4 V for nobelium, as shown in Table 33. The stable divalent state of nobelium in aqueous solution is unique among the actinide and lanthanide elements.

As a continuation of our studies of reduction potentials of the heavier actinides,^{3,4} we measured the half-wave reduction and amalgamation potential of ^{259}No in the presence of both weak (acetate) and strong (citrate) complexing ions, for comparison with the preceding members of the actinide series. We used a controlled-potential radiocoulometry cell, shown in Fig. 83, to determine the equilibrium distribution of nobelium between the aqueous and amalgam phases as a function of the applied potential. The ratio of the amount of nobelium actually reduced to the limiting amount, A/A_l , was used to determine the half-wave potential for the chosen complexing environment.

We produced ^{259}No by bombardment of a ^{248}Cm target with ^{18}O ions at the 88-in. cyclotron at the Lawrence Berkeley Laboratory (Berkeley, CA). The $^{248}\text{Cm}(^{18}\text{O}, \alpha 3n)^{259}\text{No}$ reaction was used. Nobelium atoms recoiling from the target were collected on a gold foil during a 2-h irradiation period, a process which yielded a few hundred atoms of ^{259}No for each radiocoulometric experiment. After dissolution with aqua regia, the gold was chemically separated using an anion-exchange column.

Nobelium was separated from other actinide contaminants by elution from a column of bis(2-ethylhexyl)phosphoric acid (HDEHP) on an inert support.

Tracer amounts of ^{249}Cf and ^{254}Es were combined with the nobelium and the appropriate complexing solution. This solution was then added to the coulometric cell and assayed before the start of radiocoulometry. During the reduction period, an agitator mixed the aqueous phase. After a potential was applied for a period suitable for the reduction reaction to reach

Figure 83. Controlled-potential radiocoulometry apparatus.

equilibrium (as determined by a preliminary series of experiments), aliquots were taken and counting samples prepared to determine the distributions of alpha and fission activities in the aqueous and amalgam phases. Alpha activities of ^{259}No , ^{249}Cf , and ^{254}Es were measured by alpha spectroscopy. We also followed the growth of spontaneous fission activity from ^{259}Md , the electron-capture daughter of ^{259}No , in order to determine the distribution of nobelium. A total of 18 irradiations were done to measure the two half-wave potentials reported.

Figure 84 shows the results of our measurements with a 0.05M acetate buffer solution. We measured a half-wave reduction and amalgamation potential of -1.716 ± 0.010 V relative to a saturated KCl calomel electrode (sce). This value is considerably higher than the value of -1.85 ± 0.1 V estimated by Meyer *et al.*⁵ They used a modified radiopolarographic technique with 223-s ^{255}No . The short half-life of ^{255}No and the small number of atoms produced accounts for their larger reported error. The plot of the logarithmically transformed wave, $\log A/(A_1 - A)$, shows the reciprocal of the slope to be 30 mV. This is in agreement with the value of 30 mV for a reversible reduction process involving two electrons. Correcting for the sce potential relative to the standard hydrogen potential of +0.242 V, and an estimated amalgamation potential of 0.90 V, we obtain a $\text{II} \rightarrow 0$ reduction potential of -2.37 V relative to the standard hydrogen electrode E_0 .

Similarly, we determined the half-wave reduction and amalgamation potential of nobelium in ammonium citrate to be -1.772 ± 0.015 V. This indicates a shift in the half-wave potential of $+0.056 \pm 0.018$ V from the noncomplexing acetate to the complexing citrate

solution. A similar shift was seen for fermium [~ 0.094 V (Ref. 3), and 0.100 ± 0.009 V (Ref. 6)], and mendelevium [~ 0.045 V (Ref. 3), and 0.110 ± 0.014 V (Ref. 6)]. These values are close to the shift of 60 to 90 mV observed for divalent alkaline earth ions, and much less than the 200 mV shift observed for trivalent actinides, where the larger ionic charge produces stronger bonding of complexing anions. Table 33 summarizes the reduction potentials of the heavy actinides,⁷ including the $\text{II} \rightarrow 0$ value for nobelium found in this work. The table also shows values for the comparable heavy lanthanides.⁸

Figure 84. Equilibrium distribution of nobelium between aqueous and mercury amalgam phases, A/A_1 , as a function of applied voltage. The logarithmically transformed wave $A/(A_1 - A)$ (upper line) shows an inverse slope of 30 mV (sce is saturated calomel electrode).

Table 33. Reduction potentials of the heavier actinides and lanthanides, reported as $-E_0$ vs the standard hydrogen electrode.

Half-reaction	Reduction potential (V)				
	Einsteinium	Fermium	Mendelevium	Nobelium	Lawrencium
$\text{III} \rightarrow \text{II}$	1.55	1.15	0.2	-1.4	—
$\text{II} \rightarrow 0$	2.20	2.37	2.4	2.37	—
$\text{III} \rightarrow 0$	1.98	1.95	1.7	1.1	2.06
	Holmium	Erbium	Thulium	Ytterbium	Lutetium
$\text{III} \rightarrow \text{II}$	2.9 (calc)	3.1 (calc)	2.3	1.15	—
$\text{II} \rightarrow 0$	4.12	—	—	2.77	—
$\text{III} \rightarrow 0$	2.32	2.30	2.26	2.27	2.26

In conclusion, we determined that the $\text{II} \rightarrow 0$ reduction potential of nobelium relative to the standard hydrogen potential is -2.37 V. The increasing stability of the divalent oxidation state in the heavy actinides is clearly shown in Table 33 by the change in the values of the $\text{III} \rightarrow \text{II}$ potential for einsteinium, $+1.55$ V, to nobelium, -1.4 V. This behavior supports the

conclusion that compared to analogous 4f electrons, the 5f electrons are more tightly bound in the latter part of the series. Thus there is a lowering of the 5f energy levels with respect to the Fermi level as the atomic number increases.

This work was supported by the LLNL Nuclear Chemistry Research Program.

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Observation of an E0 Isomeric Transition from the ^{238}U Shape Isomer

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We confirmed the existence of the gamma branch in the decay of the ^{238}U shape isomer by the observation of an E0 transition between the isomer and the ground state. The data are consistent with an excitation energy of 2558 keV, a spin of 0^+ , and a half-life of about 200 ns for the isomer.

The double-humped potential barrier has been accepted generally as an explanation for a number of phenomena observed in actinide nuclei, including the existence of fissioning shape isomers. Plausible evidence for a gamma-ray branch in the decay of these isomers, involving the penetration of the inner barrier back to the ground-state well, was presented by Russo *et al.*² for the case of 200-ns $^{238\text{m}}\text{U}$. There has been a strong need, however, to obtain more conclusive evidence for the existence of such a branch, since the $^{238\text{m}}\text{U}$ results² have not been confirmed and since several attempts³ to find gamma rays in the decay of a parallel case of $^{236\text{m}}\text{U}$ have failed.

The gamma-ray techniques used in previous shape-isomer investigations have several obvious limitations: (1) definite assignments of observed gamma rays to specific nuclei are not possible unless several transitions fitting into a well-known level scheme are observed; (2) gamma-ray spectrometry alone can hardly give conclusive information for the spin and parity of an isomeric state, nor can it firmly distinguish between spin and shape isomers; and (3) sources of background radiation can produce a large number of gamma rays, many of which are due to neutrons and exhibit a variety of confusing apparent half-lives.

In the present work, we used special internal-conversion-electron spectroscopic techniques to search for the expected isomeric transitions from $^{238\text{m}}\text{U}$. This method, complementary to the gamma-ray techniques, is capable in principle of overcoming most of the problems associated with the latter. For example, the observation of K

and L conversion lines in an even-even nucleus could fix both Z and A as well as the shape character and the spin of the isomer.

The ^{238}U isomer was produced by means of the $^{238}\text{U}(\text{d,pn})$ reaction with a pulsed 18-MeV deuteron beam from the tandem Van de Graaff accelerator at the Los Alamos National Laboratory (Los Alamos, NM). Internal-conversion electrons were detected at 90° to the beam with a superconducting solenoidal electron spectrometer⁴ operated in a two-loop lens mode. In this instrument, the source region visible to the Si(Li) detector is contained in a sphere with a radius of about 1 cm. We used a thin target with its plane oriented parallel or nearly parallel to the beam direction, thus increasing by a "tilting factor," n , the effective target thickness seen by the beam. Hence fission products were able to escape from the target and the background was significantly reduced. The target was depleted metallic (partly oxidized) uranium, $310\text{ }\mu\text{g}/\text{cm}^2$ thick, deposited on a $10\text{-}\mu\text{g}/\text{cm}^2$ carbon backing. The target foil was 12 mm wide by 20 mm tall and was supported along the top and bottom edges by a frame made of 0.5-mm-diam tungsten wire. A comparison of the observed and expected^{5,6} beta backgrounds shows that about 96% of the fission fragments escape from this target. Other sources of beta background were negligibly small.

The electron spectrometer was calibrated for energy and efficiency using ^{152}Eu and ^{90}Nb sources having accurately known absolute activities. These sources were also used, along with a pulser, to monitor the stability of the system. For absolute cross-section

determinations, we used a silicon fission-fragment detector with an accurately known solid angle defined by a collimating aperture. The needed fission cross-section and fragment angular-distribution data were taken from Ref. 7.

We centered the momentum-acceptance window of the spectrometer near 2.4 MeV in order to observe the K conversion line of the 2514-keV gamma ray reported in Ref. 2. With this setting, the total energy efficiency [i.e., the transmission of the solenoid times the intrinsic full-energy-peak efficiency of the Si(Li) detector], in the energy range 2.1 to 2.8 MeV, is more than 1% and reaches a maximum of 2.8% at 2.4 MeV. (We measured the efficiency at 2.301 MeV directly by using the conversion-electron line from the 2.319-MeV E5 transition in the decay of the ^{90}Nb calibration source.) After a six-day data-collection period, corresponding to almost 10^{12} fissions produced in the target, the E2 K conversion line was not observed. The upper limit for the production cross section of such a line (corresponding to 2 std dev) was 80 nb, assuming a half-life of 200 ns. This corresponds to an upper limit of $50 \mu\text{b}$ in the gamma-ray production cross section, which is clearly smaller than the value of $90 \pm 25 \mu\text{b}$ reported by Russo *et al.*³ On the other hand, we observed a

prominent line (Fig. 85) with a half-life consistent with 200 ns at $2442.4 \pm 2.0 \text{ keV}$, which, if converted in uranium, corresponds to a transition energy of $2558 \pm 2 \text{ keV}$, in excellent agreement with the excitation energy of 2559 keV of the shape isomer proposed in Ref. 2. Thus, our energy determination agrees well with that of Russo *et al.*, but there is a disagreement in the cross sections.

In view of this discrepancy, we attempted to make an independent estimate of the isomer gamma-ray production cross sections on the basis of the fission-product photopeak intensities listed in Ref. 2. The approach was to compare the 2514- and 1879-keV intensities to that of the 1280-keV line due to $^{134\text{m}}\text{Te}$, in a manner similar to that used in Ref. 8, and we obtained about $35 \mu\text{b}$, which is consistent with our experimental upper limit. Thus, one can conclude that the cross sections given in Ref. 2 are too large, perhaps by a factor of 3.

Having re-evaluated the gamma-ray intensity scale of Refs. 3 and 4, we may estimate the lower limit for the K conversion coefficient α_K corresponding to the peak found at 2442.4 keV. Our measured cross section for production of this electron line is $230 \pm 50 \text{ nb}$, assuming a half-life of 200 ns. If the line is associated with a

Figure 85. Electron spectrum from 2.3 to 2.5 MeV, obtained during the time interval 44 to 357 ns after the beam pulses of 18-MeV deuterons. A long-lived background, obtained in the delayed-time window from 1.08 to 6.05 μs , has been subtracted. A least-squares fit of a second-order polynomial to the background points is shown. The position of the K-electron peak from the 2514-keV E2 transition expected on the basis of Ref. 1 is indicated by the arrow at 2397 keV. The inset shows the decay of the 2442.4-keV peak due to the K electrons from the 2558-keV E0 transition.

gamma-ray transition in the uranium region, we obtain $\alpha_K > 0.02$; for a fission-fragment transition, after appropriate corrections for fragment escape, the limit would be $\alpha_K > 0.2$. In either case, the multipolarity of the transition must be E0. In view of the excellent energy agreement mentioned, it can be concluded that this line, which is the only line observed in the energy range 2.1 to 2.8 MeV, represents the ground-state E0 transition from the ^{238}U shape isomer at 2558 keV. It would be most unlikely for such a transition to occur in any of the neighboring nuclei in the uranium region; it would be equally improbable for an isomeric state to decay via a strong, high-energy E0 transition in any of the fission products.

Our results, including the re-evaluated data of Ref. 2, are summarized in Fig. 86. The E0 intensity given represents the total intensity, including L, M, . . . conversion and internal-pair formation contributions of 16 and 15%, respectively. The deduced value of the monopole strength parameter ρ^2 is 1.7×10^{-9} , the smallest known; the value of about 0.4 for $X(\text{E0}/\text{E2})$ is typical of the actinide region.

This research was supported by the LLNL Nuclear Chemistry Research Program.

Figure 86. Partial decay scheme (not drawn to scale) of the ^{238}U shape isomer, based on re-evaluated data of Ref. 2 and on our results. The accuracy of the estimated intensities may not be better than a factor of 2.

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Electron Spectroscopy in Coincidence with Protons from the (t,p) Reaction

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We operated an electron spectrometer in coincidence with a thin annular Si(Au) proton detector to obtain conversion-electron spectra associated with the (t,p) reaction. Electrons are transported from the target to a Si(Li) detector through a solenoidal magnetic field in a two-loop baffle arrangement. We obtained proton detection efficiencies of up to 14% with a ⁹⁸Mo target by optimizing the forward-angle position of the Si(Au) detector.

The importance and feasibility of performing gamma-ray spectroscopy in coincidence with protons from the (t,p) reaction was pointed out recently.³ Since *Q* values are positive by several MeV for the (t,p) reaction, and stopping powers are considerably larger for tritons than for protons, a suitable absorber in front of the proton detector can stop all the scattered tritons while transmitting virtually all the protons. Therefore the necessary particle discrimination is much more easily accomplished than, for example, in the case of the (p,t) reaction.⁴

With the (t,p) reaction, some of the most neutron-rich isotopes become available for study. It would be desirable to combine conversion-electron spectroscopy with gamma-ray data in order to improve the determinations of multipolarities and level assignments, especially in the case of 0^+ states and $E0$ transitions. Effective electron spectroscopy in this case generally requires electron-proton coincidence experiments to sort out the electron peaks associated with the (t,p) reaction from the much more complex and intense spectrum of peaks associated with the (t,xn) reactions.⁵ High detection efficiency is important in both detectors, especially since internal conversion coefficients are small (typically about 1% or less). There is no need for good energy resolution in the proton detector, however, since its primary purpose is to provide a coincidence gate for background reduction.

We developed an electron-proton coincidence system optimized for use with in-beam electron

spectroscopy and the (t,p) reaction. The target, in the source position of an electron spectrometer, was bombarded with tritons from the Van de Graaff accelerator at the Los Alamos National Laboratory (Los Alamos, NM). An annular Si(Au) detector located behind the target on the triton beam line detected protons from the (t,p) reaction.

The electron spectrometer⁶ consists of a solenoidal magnetic field with a two-loop baffle system to transport electrons from the target to a Si(Li) energy-dispersive detector. The solenoid axis is perpendicular to the beam line. With a 16-mm-diam detector, the momentum-transmission window has a width of about 20% and a maximum transmission of about 7%. The maximum efficiency for electron detection in the full-energy peak of the detector is smaller than this by a factor of up to 2 because of backscatter and edge effects at the detector.

Figure 87 shows the arrangement of the target and the annular Si(Au) particle detector. The graphite absorber of 160 mg/cm² is sufficient to stop tritons with energies up to 18 MeV. Protons from the (t,p) reaction, with energies generally exceeding 16 MeV for triton-beam energies of 16 MeV, lose 5 MeV or less in this absorber.

The annular particle detector is 300 μ m thick (70 mg/cm²), with an outer diameter of 23 mm and a hole diameter of 13 mm. Protons with energies above about 6.0 MeV are not stopped by this detector, so it generally functions more as a dE/dx detector than as a full-energy detector. The lack of resolution is not a serious deficiency,

Figure 87. Si(Au) particle-detector assembly and target foil. The triton beam line coincides with the centerline (axis) of the annular detector assembly.

however, and an important advantage of the thin detector is its low sensitivity to the background radiation of gamma rays, electrons, and neutrons.

The distance between the target and the detector is adjustable. Both the solid angle subtended by the detector and the range of angles contained within the solid angle are functions of that distance. The maximum proton-detection efficiency is obtained by adjusting the distance in order to achieve the optimum overlap between the solid angle of the detector and the principal maxima of the proton angular distribution.

Targets were thin strips, typically 5 mm wide and several hundred $\mu\text{g}/\text{cm}^2$ thick. They were supported at both ends by a wire frame that allowed the target to be placed in the triton beam with the target plane parallel to the beam direction and perpendicular to the spectrometer axis. This arrangement maximized the effective target thickness (≥ 5 times the foil thickness) encountered by the triton beam, and allowed the target foils to be sufficiently thin ($< 1 \text{ mg}/\text{cm}^2$) to permit good electron energy resolution.

Targets were bombarded with 16-MeV tritons in a continuous beam of typically 5 to 10 nA. Single-parameter pulse-height data (electron energies) as well as two-parameter coincidence data, involving either time distributions or spectra from the particle detector in coincidence with the electron spectrum, were accumulated in the megaword memory of the LANL computer-based data system.

We evaluated the performance of the proton detector from prompt and random coincidence spectra obtained with test targets of ^{98}Mo and ^{204}Pb bombarded with a 16-MeV triton beam. The random coincidence spectra are equivalent to the ungated (singles) spectrum of pulses from the particle detector. Comparison with the prompt coincidence spectra showed that most of the particle-detector pulses were due to protons from the (t,p) reaction. This is in contrast to the results shown in Ref. 3 for a detector that was five times as thick as ours (1500 vs 300 μm). The thicker detector produced a much more intense low-energy tail of background pulses.

The ratio of the coincidence counting rate to the singles counting rate for an electron line

gives the detection efficiency of the proton detector. This efficiency reflects not only the solid angle subtended by the detector, but also how well the angles within the solid angle overlap the major peaks in the angular distribution of protons. Therefore, the optimized proton-detection efficiency varies, depending on the L value of the two-neutron transfer and hence on the spin of the final state. The proton energy spectrum may be broad, but we expect the detection efficiency to be quite independent of proton energy, since all protons of interest have sufficient energy to penetrate the absorber and traverse the detector. Optimum proton-detection efficiencies ranged from about 8% for protons with less favorable angular distributions to about 14% in the best case (protons that feed the $0_2^+ \rightarrow 0_1^+$ E0 transition in ^{100}Mo).

Figure 88 shows electron spectra obtained during a run of 6 h with a 25-nA beam of 16-MeV tritons on a ^{98}Mo target. The target was

enriched to 97.0% ^{98}Mo and had a thickness of 1.0 mg/cm^2 . This is a good test case because the $^{98}\text{Mo}(t,p)^{100}\text{Mo}$ reaction populates the well known low-lying 0^+ , 2^+ , and 4^+ states relatively strongly. Prominent peaks associated with the (t,p) reaction and with other reactions are labeled in Fig. 88. The E0 decay of the inelastically excited 0_2^+ state at 734 keV in ^{98}Mo appears strongly in the ungated electron spectrum. In the proton-gated spectrum, peaks due to transitions in ^{100}Mo are seen to be enhanced by two orders of magnitude or more compared to those due to competing reactions.

The inset in Fig. 88 shows the time distribution of coincidences between proton pulses and the K electrons from E0 decay of the 0_2^+ state at 694 keV in ^{100}Mo . These data yield an improved value of $3.0 \pm 0.1 \text{ ns}$ for the half-life of the 0_2^+ state.

This research was supported by the LLNL Nuclear Chemistry Research Program.

Figure 88. Electron spectra from a ^{98}Mo target bombarded with 16-MeV tritons. The top spectrum is the complete electron spectrum with no coincidence gating, and the bottom spectrum is of electrons in coincidence with pulses from the particle detector. Peaks are labeled according to the reaction (top), or the transition and its energy in keV (bottom). The inset shows the time distribution of coincidences between protons and the K electrons from the 694-keV E0 transition in ^{100}Mo . The indicated half-life of the 0_2^+ state is $3.0 \pm 0.1 \text{ ns}$.

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Measurement of E0 Transitions in ^{100}Mo and ^{102}Mo

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We made measurements of the E0 transitions in ^{100}Mo and ^{102}Mo , and determined the ρ_{21}^2 and X_{211} values for these nuclei. The systematics of the values obtained are discussed qualitatively in terms of nuclear deformation and the interacting boson model.

The transition from a spherical to a deformed shape in nuclei as a function of Z and N provides a significant test of nuclear models. The molybdenum nuclei manifest such a transition as neutrons are added to the closed neutron shell nucleus ^{92}Mo . The rapid lowering of the 2_1^+ level energy in the heavier molybdenum nuclei is an easily observed indication of this transition. The heaviest nuclei, ^{104}Mo and ^{106}Mo , possess ground-state bands similar to the rotational bands in rare earth nuclei.³

We are studying E0 decays in ^{100}Mo and ^{102}Mo to obtain additional experimental data on these transitional nuclei. The properties of 0^+ levels and of the nonradiative E0 transitions are associated with nuclear properties such as nuclear radius and deformation. For example, Rasmussen⁴ showed that the ratio of reduced transition rates [the X value, equal to $B(E0)/B(E2)$, as shown in Eq. (2)] is proportional to the square of the deformation parameter β for the beta-vibrational transitions in spheroidal nuclei. It can also be shown that when a monopole transition occurs between two states of mixed spherical and deformed configurations, the square of the monopole strength parameter ρ^2 is proportional to β^4 (Ref. 5). For the 0_2^+ state of a spherical quadrupole-phonon model, the X value equals β_{rms}^2 , where β_{rms} is a measure of the dynamic deformation, not a static deformation. These and other theoretical results indicate that the E0 transitions can provide significant information about changes in nuclear shape.

We measured the conversion-electron decay of the 0_2^+ levels in ^{100}Mo and ^{102}Mo , using the techniques described in this report (see "Electron Spectroscopy in Coincidence with Protons from

the (t,p) Reaction," in this section). The 0_2^+ levels had been identified in (t,p) reaction studies.^{6,7} The E0 transition in ^{100}Mo had been measured in (p,p'e) experiments,^{8,9} while the half-lives of the ^{100}Mo and ^{102}Mo 0_2^+ levels had been determined by Coulomb excitation,¹⁰ by the recoil-distance Doppler-shift method,¹¹ and, in the case of ^{100}Mo , by the delay-coincidence method.⁹

Table 34 shows our results for the $I_K(E0)/I_K(E2)$ electron ratios and the half-life of the ^{100}Mo 0_2^+ level. The results of previous measurements are included for comparison. From these data one can calculate, for comparison with theory, the values of the monopole strength parameter ρ^2 (Ref. 12), and the X value (Ref. 4). ρ^2 is determined from conversion-electron measurements by

$$\rho_{if}^2(E0; 0_i^+ \rightarrow 0_f^+) = \frac{I_K(E0; 0_i^+ \rightarrow 0_f^+)}{I_K(E2; 0_i^+ \rightarrow 2_j^+)} \times \frac{\alpha_K(E2; 0_i^+ \rightarrow 2_j^+)}{\Omega_K(E0; 0_i^+ \rightarrow 0_f^+)} \times W_\gamma(E2; 0_i^+ \rightarrow 2_j^+) \quad (1)$$

where

$$W_\gamma(E2; 0_i^+ \rightarrow 2_j^+) = 0.0122$$

$$\times E_\gamma^5(E2) \times B(E2; 0_i^+ \rightarrow 2_j^+) \quad ,$$

$$B(E2; 0_i^+ \rightarrow 2_j^+) = 56.4 [t_{1/2}(E2) \times E_\gamma^5(E2)]^{-1} e^2 b^2 \quad .$$

$I_K(E0)/I_K(E2)$ is the measured conversion-electron ratio, α_K is the usual conversion coefficient, Ω_K is the electronic factor (the factor for nonradiative

transitions, which is analogous to the conversion coefficient), $E_\gamma(E2)$ is gamma-ray energy in keV, and $t_{1/2}(E2)$ is the E2 gamma-ray partial half-life in seconds. The X value is defined by

$$X_{ijk} = \frac{B(E0; 0_1^+ \rightarrow 0_j^+)}{B(E2; 0_1^+ \rightarrow 2_k^+)} \times \frac{e^2 R^4 \rho^2}{B(E2)}$$

$$= 2.56 \times 10^{-6} \times A^{4/3}$$

$$\times E_\gamma^5(E2) \times \frac{I_K(E0)}{I_K(E2)} \times \frac{\alpha_K(E2)}{\Omega_K(E0)}, \quad (2)$$

where A is the atomic mass.

Figure 89 shows the values of ρ^2 and X_{211} for the even-mass molybdenum nuclei from ^{96}Mo to ^{102}Mo . The data for ^{96}Mo are unpublished data cited in Ref. 13, the values for ^{98}Mo are calculated from data in Refs. 14 and 15, and the 0_2^+ level half-life for ^{102}Mo is from Ref. 11. The large uncertainty in the ρ^2 value for ^{102}Mo is due mainly to the uncertainty in the half-life for the 0_2^+ level.

Sheline *et al.*¹⁶ speculated that the low-lying 0_2^+ and 2_2^+ levels in ^{100}Mo are the band heads of bands which have a deformed shape, while the ground state is spherical. They suggested that compared to ^{100}Mo , the deformed states in ^{102}Mo had lower energies, and the 0^+ ground state in ^{102}Mo is deformed while the 0_2^+ state is the spherical state. Bohn *et al.*,¹¹ on the other hand, suggested that since fast E2 transitions connect the 0_2^+ and 2_1^+ levels in both ^{100}Mo and ^{102}Mo , the 0_2^+ states may be beta-vibrational band

heads in these nuclei, but that additional experimental data are necessary to decide the nature of these levels. If we use Rasmussen's model, $X_{\beta 11} \propto \beta^2$, where $X_{\beta 11}$ indicates the X value for a beta-vibrational band head, we find that the deformation parameter β for ^{102}Mo is 1.7 times that of ^{100}Mo . This ratio is somewhat larger than the value of 1.48 for the deformation-parameter ratio obtained by Bohn *et al.*¹¹ for these nuclei. Our result indicates that the 0_2^+ level in ^{102}Mo can be best characterized as a beta-vibrational band head, while the 0_2^+ level in ^{100}Mo is more vibrational in character. This idea is supported by the increase in ρ^2 from ^{100}Mo to ^{102}Mo . The large value of ρ^2 for ^{98}Mo , compared to the values for ^{96}Mo and ^{100}Mo , may indicate that in this nucleus the 0_1^+ and 0_2^+ levels are highly mixed states made up of spherical and deformed configurations (in this case, $\rho^2 \approx \beta^4$, as discussed in Ref. 4). The $N = 56$ closed subshell may have some influence here by providing highly spherical configurations. Slight evidence for the influence of the $N = 56$ subshell closure is seen in the 2_1^+ energies for these nuclei, as the 2_1^+ level is a little higher in ^{98}Mo than in ^{96}Mo . The smaller value of ρ^2 for ^{100}Mo may signify a return to a more vibration-like structure for this nucleus.

Recently, calculations were made using the interacting boson model with $Z = 40$ as the proton closed shell (1-proton particle boson, $N_\pi = 1$),⁷ and with $Z = 50$ as the proton closed shell (4-proton hole bosons, $N_\pi = 4$).^{3,7} These calculations failed to reproduce the behavior of the 0_2^+ levels in the molybdenum nuclei.

Table 34. Summary of measurements on E0 transitions in ^{100}Mo and ^{102}Mo .

Nuclide	Parameter	Our measurement	Previous measurement
^{100}Mo	$I_K(E0)/I_K(E2)$	0.0458 ± 0.023^a	0.759 ± 0.045^b 0.62 ± 0.5^c
	$t_{1/2}(\text{ns})$	3.1 ± 0.1	2.4 ± 0.3^d $2.9 \pm 0.5^{e,f}$ 1.46 ± 0.5^g
^{102}Mo	$I_K(E0)/I_K(E2)$	0.31 ± 0.05^a	—
	$t_{1/2}(\text{ps})$	—	28 ± 11^f

^a An uncertainty of 5%, due to the uncertainty of the conversion-electron sweeping efficiency, should be added in quadrature.

^b From Ref. 8.

^c Calculated by the authors from data in Ref. 9.

^d From Ref. 9.

^e From Ref. 10.

^f The authors of Ref. 11 point out some possible errors in determining this value.

^g From Ref. 11.

Sambataro and Molnar¹⁷ used the interacting-boson-model framework to calculate many energy levels and relative transition rates for the molybdenum nuclei. They used $Z = 40$ as a shell closure and did two calculations, one with $N_\pi = 1$ and one with $N_\pi = 3$, where a proton

Figure 89. ρ^2_{21} and X_{211} values for molybdenum nuclei from $A = 96$ to 102.

pair is promoted across the $Z = 40$ shell closure. Here the proton-hole boson and the 2-proton-particle bosons are treated equivalently. The two calculations were mixed to give the final result. Sambataro and Molnar indicate that the configuration $N_\pi = 1$ gives states that appear to be vibrational in character, while the configuration $N_\pi = 3$ gives states that appear to be gamma-unstable in character. They point out that in their calculation for ^{102}Mo the 0^+_1 and 0^+_2 levels belong to the $N_\pi = 3$ configuration (which is consistent with our experimental results), while the 0^+_3 level belongs to the $N_\pi = 1$ configuration. The experimental determination of ρ^2 and X for the 0^+_3 level of ^{102}Mo will be difficult, but interesting from a modeling standpoint.

Our discussion has necessarily been qualitative; detailed calculation of the properties of 0^+ states and E0 transitions could further refine nuclear models for this interesting region.

This program was supported by the LLNL Nuclear Chemistry Research Program.

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Studies of Proton Pickup of the Low-Lying States in ^{96}Nb

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We studied the nuclear structure of ^{96}Nb with the (t,α) reaction at 17 MeV on an isotopically enriched target of ^{97}Mo , using a Q3D spectrometer. To assign ℓ transfer values, measured angular distributions were compared with calculations done using the distorted-wave Born approximation. We draw conclusions about the spins and parities of the observed levels by combining the results with published data and shell-model considerations.

We used the $^{97}\text{Mo}(t,\alpha)^{96}\text{Nb}$ reaction to investigate the level structure of ^{96}Nb . The target nucleus, $^{97}_{42}\text{Mo}_{55}$, has a ground-state spin and parity of $5/2^+$ which arises from the $\nu d_{5/2}^{-1}$ neutron-hole configuration. In the (t,α) reaction, the valence neutrons act as a spectator group, and proton pickup is therefore expected to give relatively clear information about the proton components making up the low-lying levels of ^{96}Nb . Previous investigations¹⁻⁴ have successfully interpreted other data on the low-lying structure of ^{96}Nb in terms of j-j coupling in the nuclear shell model. At low energy one expects states involving the $\pi g_{9/2}^{-1}\nu d_{5/2}^{-1}$ and $\pi p_{1/2}^{-1}\nu d_{5/2}^{-1}$ configurations, which give rise respectively to the following spin-parity multiplets: 2^+ , 3^+ , 4^+ , 5^+ , 6^+ , 7^+ ; and 2^- , 3^- . These states are expected to be strongly and selectively populated in a (t,α) reaction.

The $^{97}\text{Mo}(t,\alpha)^{96}\text{Nb}$ reaction was obtained with 17-MeV tritons from the tandem Van de Graaff accelerator at the Los Alamos National Laboratory (Los Alamos, NM). The target was prepared by vacuum evaporation of a sample of molybdenum oxide enriched to 94.24% ^{97}Mo . The target had a thickness of about $85 \mu\text{g}/\text{cm}^2$ and was mounted on a carbon substrate of about $50 \mu\text{g}/\text{cm}^2$. The alpha particles from the (t,α) reaction were momentum-analyzed in the Los Alamos Q3D spectrometer, and detected by a 1-m helical-wire proportional counter mounted in the focal plane of the magnet. Alpha-particle spectra were recorded in 1024 energy channels at spectrograph angles of 10, 12, 15, 20, 22, 25, 28, 34, 40, 45, 50 and 55 degrees relative to the incident beam. Figure 90 shows a representative

spectrum. The resolution is approximately 18 keV FWHM.

Figure 90. Alpha-particle spectrum of the $^{97}\text{Mo}(t,\alpha)^{96}\text{Nb}$ reaction, at a laboratory angle of 50° and a beam energy of 17 MeV. Peak numbers correspond to the labeled peaks in Table 35.

We determined peak areas and centroids by a least squares method which fits Gaussian line shapes to the peaks. To determine excitation energies, the alpha particle spectra were calibrated with the known energies from the $^{92}\text{Zr}(t, \alpha)^{91}\text{Y}$ reaction. Table 35 shows the excitation energies of the states in ^{96}Nb found in this experiment along with the results of previous studies. The energies determined in the experiment agree reasonably well with results from other reports.

We measured the elastic scattered triton yield with a solid state detector in the scattering chamber at an angle of 30° to the triton beam.

Figure 91 Angular distributions of alpha particles, from low-lying states observed in the $^{97}\text{Mo}(t, \alpha)^{96}\text{Nb}$ reaction that exhibit $\ell = 4$. Solid curves are from DWBA calculations that have been renormalized to fit the data (cm is center of mass).

The absolute differential cross sections were calculated by normalizing the yield for each peak in the alpha spectra to the elastic-scattered triton yield. The computer code DWUCK was used to calculate the theoretical angular distributions.

Figure 91 shows the angular distributions for the states populated in the $^{97}\text{Mo}(t, \alpha)^{96}\text{Nb}$ reaction that have been assigned by Comfort *et al.*² as the six members of the $\pi g_{9/2}^{-1} \nu d_{5/2}^{-1}$ multiplet. The solid curves are the results of calculations done by distorted wave Born approximation (DWBA). Each of the six states exhibits an $\ell = 4$ angular distribution, which is consistent with proton pickup from the $g_{9/2}$ shell model orbital and supports the work of Comfort *et al.*^{2,3}

The 2^- and 3^- states of the $\pi p_{1/2}^{-1} \nu d_{5/2}^{-1}$ configuration were previously assigned energies of 506 keV and 687 keV,⁴ respectively, as shown in Table 35. We observed the 506-keV level in the (t, α) reaction. There is, however, no evidence of any state at 687 keV in the proton pickup data from our measurements. Since the (t, α) reaction should strongly populate both states of the $\pi p_{1/2}^{-1} \nu d_{5/2}^{-1}$ configuration, the 687-keV level observed in other experiments is probably not the 3^- member of this doublet.

We observed two low-lying states that have $\ell = 1$ angular distributions in the (t, α) reaction. Figure 92 shows these angular distributions and the results of DWBA calculations. The ratio of the cross section of the 876-keV level to that of the 521-keV level is 1.4. This value is exactly what is expected from simple shell-model considerations if the 876- and 521-keV levels are

Table 35. Excitation energies of states in ^{96}Nb . Peak numbers correspond to the labeled peaks in the spectrum in Fig. 90. Also listed are the spins and parities (J^π) assigned to these states.

Peak number	Excitation energy (keV)		J^π
	This work	Previous reports	
0	0	0	6^+
1	40	43	5^+
2	147	142	4^+
3	192	180	3^+
4	230	233	7^+
5	521	506	2^-
6	644	630	2^-
		687	3^-
7	876		3^-

Figure 92. Angular distributions of alpha particles observed in the $^{97}\text{Mo}(t,\alpha)^{96}\text{Nb}$ reaction that exhibit $\ell = 1$. Solid curves are from DWBA calculations that have been renormalized to fit the data. (cm is center of mass.)

the 3^- and 2^- states of the $\pi p_{1/2}^1 \nu d_{5/2}^{-1}$ configuration. The $\ell = 1$ angular distributions are consistent with proton pickup from the $1p_{1/2}$ proton orbital.

Arvay and co-workers⁵ have reported a gamma-ray transition of 356 keV in the $^{96}\text{Zr}(p,n\gamma e^-)^{96}\text{Nb}$ reaction which has M1 multipolarity determined from the measured internal conversion coefficient. They made no assignment of this gamma-ray transition to the ^{96}Nb level scheme. It can be assigned to a transition from a 3^- state at 876 keV to a 2^- state at 521 keV.

In summary, the selectivity of the (t,α) reaction and the results reported by other research groups have allowed us to assign the 876-keV level as the 3^- state of the $\pi p_{1/2}^1 \nu d_{5/2}^{-1}$ configuration. Our results are consistent with the previously proposed level scheme.^{2,4}

This research was supported by the LLNL Nuclear Chemistry Research Program.

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Two-Particle Four-Hole Intruder-Band Members in ^{110}Cd and Their Modification of the Yrast Structure

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We combined detailed beta-decay studies with $(\alpha, n\gamma)$ measurements that utilized Compton-suppression and other standard techniques to study ^{110}Cd . We were able to identify coexisting two-particle four-hole intruder-band members up to $J^\pi = 8^+$, ground-state band members to 14^+ , and two-quasi-particle members to 14^+ . Their interactions were found to cause nonuniform behavior of the yrast and yrare sequences. We found multiple de-excitation paths for the 10^+ backband state and the lowest 5^- state.

Although the Compton-suppression technique has been used in heavy-ion in-beam studies to explore the region of highest spin states, its use in light-ion in-beam studies has not been exploited. Such use, in combination with detailed studies by complementary techniques involving, for example, beta decay or neutron-capture gamma-ray spectroscopy, offers an important opportunity to explore questions of structure at the backband and initial yrare regions.

The yrast sequence of ^{110}Cd was first identified by Lumpkin *et al.*³ using the $^{96}\text{Zr}(^{18}\text{O}, 4n\gamma)^{110}\text{Cd}$ reaction. The yrast sequence, when compared with that of other cadmium nuclei, exhibits a nonsystematic behavior. A possible mechanism for that behavior is the interaction between coexisting two-particle four-hole (2p4h) intruder-band (I band) members and the ground-state (GS) band members near and above the first break in the regular energy sequence of the yrast band (backband). Before the work described here, there was little evidence for the actual existence of such band members. However, for the even-even cadmium nuclei, the proton 2p4h 0^+ band heads were observed in $(^3\text{He}, n)$ reaction studies⁴ to decrease in energy from 3000 keV in ^{106}Cd , where I-band members should not perturb the low-spin collective structure, to 1473 and 1250 keV in ^{110}Cd and ^{112}Cd , respectively, where if I-band members coexist, they could modify the structure and properties of the GS and yrast bands. The difficulty with the concept of band

modification is that other than preliminary reports from beta-decay studies,⁵ no evidence has been found for coexisting GS- and I-band members.

The existence of intruder states and rotation-like bands built on them in odd-mass nuclei has been reviewed recently.⁶ They have been shown to be a common feature of nuclei with one and three nucleons beyond a single closed shell. Recently, attempts were made to study even-even cadmium nuclei where bands of a similar nature were suggested to exist in ^{110}Cd .⁵ While no evidence is found for band members in ^{106}Cd , both ^{112}Cd and ^{114}Cd show some evidence of possessing 2p4h intruder 2^+ rotational members built on the known 0^+ band head. Unfortunately, the experimental techniques used allowed one to study only the 0^+ and 2^+ members. Julin *et al.*⁷ showed that these states were mixed, and to date, in-beam studies have not identified higher-spin members.^{3,8} The question has arisen as to whether the GS and I bands dissolve into one band. For example, in the tin nuclei, where equivalent 2p2h band heads were identified, only a single band up to $J^\pi = 12^+$ could be observed in ^{112}Sn to ^{118}Sn .⁹ For the ^{110}Cd isotone ^{112}Sn , the 6^+ yrast band member possesses a multiple de-excitation.¹⁰

As Fig. 93 shows, we can identify coexisting GS- and I-band members up to $J^\pi = 12^+$ and 8^+ , respectively. These data are the result of combined Compton-suppression and standard

in-beam studies using the $(\alpha, xn\gamma)$ reaction at the Swiss Institute for Nuclear Research (Villigen, Switzerland), combined with detailed beta-decay studies using Compton-suppression, coincidence, and singles spectroscopy.

Heyde *et al.*¹⁰ investigated the general description of the 2p4h band head and its first excited state and their interaction with the GS band. They demonstrated the equivalence of using either a 2p4h particle-core coupling model or a configuration-mixing approach utilizing IBA-2 type calculations. We extended their IBA-2 calculations to ^{110}Cd . In doing so we chose the parameters used in the previous ^{112}Cd and ^{114}Cd IBA-2 calculations, and varied only those

Figure 93. Levels identified in ^{110}Cd using in-beam and beta-decay Compton-suppression and standard spectroscopy techniques, compared with levels from mixed-configuration IBA-2 calculations. Only selected levels above 3 MeV are shown, and levels on the extreme right of the experimental-results part are interconnected by low-energy transitions, with few transitions to positive-parity levels. The solid circles to the left of certain members of that group signify candidates for a possible octupole band. Also, the members of the GS and I bands are staggered from the main sequence for easier identification.

required by the change in underlying characteristics of ^{110}Cd over ^{112}Cd and ^{114}Cd . As shown on the right side of Fig. 93, there is remarkably good agreement. Two important features should be recognized. First, our combined data can identify all the collective states below the pairing gap at approximately 2700 keV with an almost one-to-one correspondence between observed and calculated levels. Second, the GS- and I-band members are well reproduced, including the staggering at $J^\pi = 6^+$ and 8^+ . As Table 36 shows, the branching ratios between band members are reproduced well with few exceptions. One difference is the stronger measured cross-band strength for the $J^\pi = 8^+$ level, which suggests a greater degree of $J^\pi = 8^+$ band-member mixing than expected from the simple GS- and I-band mixing. As we discuss below, this may be due to the mixing with a third 8^+ level that occurs between the GS- and I-band members.

We find that a third $J^\pi = 8^+$ level occurs at 3440 keV, intermediate in energy between the 3275- and 3792-keV 8^+ GS- and I-band members, respectively. Its interaction with the latter two levels could result from its possessing a $\pi(g_{9/2}^{-3}g_{7/2}^{+1})$ configuration as its main component. Such an assignment is consistent

Table 36. Comparison between IBA-2-calculated $B(E2)$ values and observed transitions.

$E_i (J_i^\pi)$	to	$E_f (J_f^\pi)$	$B(E2)^a$	
			Theory	Experiment
658 (2_g^+)		0 (0_g^+)	100	100
1476 (2_2^+)		0 (0_g^+)	0.004	0.049
		658 (2_g^+)	1.31	1.30
1542 (4_g^+)		658 (2_g^+)	1.66	1.53
1784 (2_1^+)		1473 (0_1^+)	100	100
		658 (2_g^+)	0.013	(13) ^b
		0 (0_g^+)	0.41	0.45
2250 (4_1^+)		1783 (2_1^+)	100	100
		1476 (2_2^+)	2.6	2.3
		658 (2_g^+)	0.49	0.19
		1542 (4_g^+)	3.2	(149) ^b
2877 (6_1^+)		2250 (4_1^+)	100	100
		1542 (4_g^+)	1.37	1.49
3792 (8_1^+)		2837 (6_1^+)	100	100
		2480 (6_g^+)	1.10	11.4 ^c

^a Absolute values are taken from *Nuclear Data Sheets*.¹¹ Where absolute rates are not measured, we normalize the intraband $B(E2)$ to a value of 100.

^b Assuming pure E2.

^c Possible third-band mixing (see text).

We find further evidence for multiple-path low-energy de-excitation of several other levels originally believed to de-excite by a single transition path. Of particular interest is the 2539-keV 5^- level. We place 33% of its de-excitation by a hitherto unidentified E2 transition to the known 3^- octupole state at 2078 keV. This, and its population by the sole de-excitation of a 3185-keV level, could suggest an octupole-state band. We find a number of levels at approximately 3000 keV that are consistent with the possession of negative parity and which exhibit the nature of a low-energy de-excitation path. Some of these levels are listed on the right side of Fig. 94. Their general properties appear consistent with the negative-parity broken-pair excitations found in associated even-even nuclei with neutron numbers 58 to 68.

This research was supported by the LLNL Nuclear Chemistry Research Program.

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The Anomalous $K^\pi = 0^-$ Band in ^{172}Tm

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A study of the level structure of ^{172}Tm showed that the properties of the $K^\pi = 0^-$ and $K^\pi = 1^-$ bands associated with the $[\pi 1/2[411]; \nu 1/2[521]]$ configuration are anomalous. We suggest that quasi-particle-phonon coupling may cause the strange properties observed.

We obtained information on the nuclear level structure of the odd-odd nucleus ^{172}Tm by a series of measurements of gamma radiation following the beta decay of chemically purified samples of ^{172}Er . Figure 95 shows the level scheme obtained from these data. Most of the levels observed were identified as band heads of specific two-quasi-particle structures or as associated rotational states. The energies of the individual neutron and proton one-quasi-particle structures are well known² for the adjacent isotopes and isotones of ^{172}Tm . These energies were combined with evaluated two-quasi-particle splitting energies³ to calculate the band-head energies expected in ^{172}Tm . Figure 96 compares the calculated band heads with those identified experimentally.

Of particular interest in the ^{172}Tm level scheme are the levels associated with the $\{\pi 1/2[411]; \nu 1/2[521]\}$ configuration. This is emphasized in Fig. 97, where we compare the level properties of the bands formed from the $\{\pi 1/2[411]; \nu 5/2[512]\}$ and $\{\pi 1/2[411]; \nu 1/2[521]\}$ configurations in both ^{170}Tm and ^{172}Tm . We note first that for the former configuration, the $K^\pi = 2^-$ and $K^\pi = 3^-$ bands (formed from the antiparallel and parallel coupling, respectively, of the individual neutron and proton orbitals) are separated by about 240 keV in both nuclei. The effect of the neutron-proton residual interaction, which causes the degeneracy of the two couplings to be split, should not change significantly between the two nuclei. Moreover, although small changes in single-particle wave

Figure 95. Level scheme of ^{172}Tm .

functions can and do occur from nucleus to nucleus, we expect no significant change in the value of the residual-interaction matrix element calculated for either nucleus. This is because the average potential in which the wave functions are evaluated should not be altered significantly by changes of a few nucleons. Also, it is rigorously true that the particle or hole nature of the wave functions has no effect on the value of the diagonal residual-interaction matrix element.³ Finally, Coriolis or other perturbations can affect the observed splittings differently in different nuclei, but these are generally small and in any case (with respect to the $\{\pi 1/2[411]; \nu 5/2[512]\}$ configuration) they are almost certainly absent in both ^{170}Tm and ^{172}Tm . For example, the energy of the $K^\pi, I = 2^-, 3$ rotational state can be perturbed by Coriolis interaction with the $K^\pi, I = 3^-, 3$ band head, but the effect is calculated to be very small (~ 1 keV). This, and the fact that the $\Delta K = 1$ off-diagonal matrix element associated with Coriolis coupling either to the $K^\pi, I = 0^-, 3$ or $1^-, 3$ rotational states of the $\{\pi 1/2[411]; \nu 1/2[521]\}$ configuration is expected to be zero, suggest that the relative energy of the $K^\pi, I =$

Figure 96. Comparison of the calculated and observed band structure in ^{172}Tm . The zeroth-order two-quasi-particle energies are determined from experimental data available on the isotopes and isotones adjacent to ^{172}Tm .

$2^-, 3$ rotational state should be similar in both nuclei. That is what is observed (66.1 keV in ^{170}Tm vs 62.5 keV in ^{172}Tm).

From these considerations, we conclude that based on obvious and simple physics arguments, the observed properties of the $\{\pi 1/2[411]; \nu 5/2[512]\}$ configuration in both ^{172}Tm and ^{170}Tm are virtually identical and totally in accord with our understanding of the nuclear physics. For these same reasons, we expected to see little or no difference in the comparison of the properties of the $\{\pi 1/2[411]; \nu 1/2[521]\}$ configuration in ^{172}Tm and ^{170}Tm . We did, in fact, find two significant and important differences.

The right-hand side of Fig. 97 compares the properties of the $\{\pi 1/2[411]; \nu 1/2[521]\}$ configuration in both thulium nuclei. There is a significant difference in the derived residual-interaction⁴ matrix elements and in the moment-of-inertia parameter for the $K^\pi = 0^-$ bands. A

Figure 97. Band structure associated with the lowest-lying two-quasi-particle configurations observed in ^{170}Tm and ^{172}Tm . The energies (keV) are relative to the lowest-lying band head of each configuration. The circled numbers (keV) are the experimentally determined residual-interaction matrix elements. Positive values are associated with the splitting of the two possible couplings of the component orbitals, while negative values are associated with an energy shift of the odd- and even-spin members of the $K^\pi = 0^-$ bands. The rotational parameter A (keV) is also noted for each band.

clear understanding of this anomalous behavior cannot be determined conclusively from the available information. A plausible explanation, however, may involve different degrees of quasi-particle-phonon mixing in the $K^\pi = 0^-$ bands in both thulium nuclei. Strong phonon admixtures in one-quasi-particle states involving both the proton $1/2[411]$ and neutron $1/2[521]$ orbitals are particularly distinctive characteristics of nuclei in this general mass region.² Virtually nothing is known about phonon admixtures in odd-odd

nuclei, but these must certainly play an important role in determining the detailed properties of ^{170}Tm and ^{172}Tm , and may well cause the particular anomaly noted here. Future experimental and theoretical work focussing on the vibrational aspects of low-lying two-quasi-particle states in this mass region would be particularly illuminating.

This research was supported by the LLNL Nuclear Chemistry Research Program.

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residual-interaction matrix elements. Whatever the magnitude of this effect, one suspects that it is similar in both nuclei because a comparison of the $K^\pi = 1^-$ rotational patterns shows no difference. Thus, although including these effects would alter the absolute values of the residual-interaction matrix elements, the relative values should not change.

Decoupled Bands in ^{197}Au and ^{199}Au

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We studied the conversion-electron spectra from $\text{Pt}(t,2n\gamma)\text{Au}$ reactions for $^{197,199}\text{Au}$, and found decoupled rotational bands based on the $h_{11/2}$ proton hole. We established a $21/2^+$ state that is depopulated by an E1 transition to the $19/2^-$ member of the band for both isotopes. These findings extend a systematic trend previously found in odd- A gold isotopes from ^{185}Au to ^{195}Au .

Stephens⁴ has shown that a single-particle orbital can couple with a rotating core in a rotationally aligned scheme. In that coupling scheme, the projection of the angular momentum on the rotation axis is sharp, and the state with the maximum value of this projection lies lowest in energy. A so-called decoupled band is then formed on this state with the spin sequence $I = j, j + 2, j + 4, \dots$, and with the energy spacings corresponding to those in the adjacent even-even isotopes. Decoupled bands have been reported in the odd- A gold nuclides from ^{185}Au to ^{195}Au , and have been interpreted as resulting from the rotationally aligned coupling of the $h_{11/2}$ proton-hole state to a rotating core.⁵⁻⁸ Nail⁹ has proposed similar bands in $^{197,199}\text{Au}$ for the yrast states observed in his gamma-gamma coincidence results. We used in-beam conversion-electron data to confirm those assignments and to identify a $21/2^+$ level in both nuclei.

Excited states of $^{197,199}\text{Au}$ were populated by the $(t,2n)$ reaction on targets of $^{196,198}\text{Pt}$, with beam energies of 13 and 16 MeV. We detected electrons with a new in-beam spectrometer system. The spectrometer consists of a superconducting solenoid which transports electrons through a baffle system to a cooled Si(Li) detector. We accumulated a singles spectrum for the ^{198}Pt target by operating the instrument in a slow-sweep mode in which the momentum-acceptance window was changed by stepping the current of the transporter magnet. Figure 98 shows part of this spectrum. We obtained K-conversion coefficients for the transitions of interest by combining our data with the gamma intensities of Nail⁹ and normalizing to the theoretical value¹⁰ for the 361.7-keV E2 transition. We could not obtain

reliable intensities from a singles spectrum of ^{197}Au because of the intense electron lines produced by prompt transitions and by those associated with beta-decay products. Therefore the triton beam was pulsed for the ^{196}Pt target, and spectra were recorded as a function of delay time after the beam pulse. We could measure conversion coefficients from the delayed spectra of the decoupled band fed from a $21/2^+$ isomer.¹¹ The K-conversion coefficient for the 357.8-keV $15/2^-$ to $11/2^-$ transition was determined by positioning the maximum of the transmission window at this line, where the absolute efficiency is known. A Ge(Li) detector,

Figure 98. (a) Delayed-conversion-electron spectrum of ^{197}Au . (b) Conversion-electron spectrum of ^{199}Au .

Table 37. Internal conversion coefficients for selected lines in ^{197}Au and ^{199}Au .

E_γ (keV)	Experimental		Theoretical						Multipolarity
			E1		M1		E2		
	α_K	$\alpha_{K/L}$	α_K	$\alpha_{K/L}$	α_K	$\alpha_{K/L}$	α_K	$\alpha_{K/L}$	
^{197}Au									
357.8	0.040 ± 0.004	—	0.015	6.3	0.18	6.1	0.04	2.6	E2
429.4	0.0080 ± 0.0010	—	0.010	6.7	0.11	6.3	0.027	3.2	E1
639.8	0.010 ± 0.001	4.9 ± 0.6	0.004	6.3	0.038	6.1	0.011	4.0	E2
^{199}Au									
361.7	0.04	2.9 ± 0.4	0.015	6.3	0.18	6.1	0.04	2.6	E2
584.7	0.0030 ± 0.0004	—	0.005	6.3	0.048	6.2	0.013	3.7	E1
594.4	0.010 ± 0.001	2.7 ± 0.4	0.005	6.2	0.046	6.2	0.012	3.4	E2

Figure 99 Decoupled-band levels in ^{197}Au and ^{199}Au compared with ground-band levels in adjacent mercury nuclei

also of known efficiency, recorded gamma rays for the same measurement. By combining the intensities from the two detectors for the same delay times, we could directly determine the conversion coefficient. For the higher-energy lines we used the sweep mode to obtain the

delayed-electron spectra, and obtained a normalization from the L lines of the 357.8-keV transition. Table 37 gives the results of all the K-conversion coefficient measurements, along with K-to-L ratios where possible.

Figure 99 shows a partial level scheme for the decoupled bands with values of J^π derived from our conversion-electron data. The figure also shows the energy spacings from the neighboring even-even mercury isotopes. An additional feature of the odd- A gold level schemes is the existence of a $21/2^+$ state feeding the $19/2$ level of the decoupled band. This level has been seen in ^{185}Au (except ^{187}Au), and is isomeric in ^{193}Au . It has been described as having the three-particle configuration

$[(\nu_{13/2}, \nu_{p3/2})_5, \pi h_{11/2}]_{21/2^+}$.¹¹ The measured half-life of the isomer in ^{197}Au is 6 ± 2 ns, which gives a reduced transition probability of $9 \pm 3 \times 10^{-7} e^2 \text{ fm}^2$. Assuming that the $B(E1)$ would be the same for the analogous transition which we found in ^{199}Au , we predict a value of 2.5 ± 0.8 ns for the half-life for this state.

This work was supported by the LLNL Nuclear Chemistry Research Program.

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Measurement of the Quadrupole Moment of the 5^- Level in ^{206}Hg

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We used the perturbed-angular-distribution method to measure the quadrupole interaction frequency of the $^{206}\text{Hg } \pi(h_{11/2}^{-1}s_{1/2}^{-1}, 5^-)$ isomer in metallic mercury at 225 K. We found that $\nu_Q = 136.5 \pm 3$ MHz. This gives $e_Q(^{206}\text{Hg}, 5^-) = 65 \pm 13 \text{ e fm}^2$ and an effective charge $e_{\text{eff}}(\pi h_{11/2}^{-1})$ of $2.2 \pm 0.5 \text{ e}$.

Studies of nuclei around doubly magic ^{208}Pb have contributed much to our understanding of single-particle structure and its interplay with collective excitations in nuclei. In particular, the electromagnetic properties of single-particle states often deviate strongly from the predictions of the shell model and they frequently reveal collective admixtures. Very little is known about the electromagnetic properties of the proton-hole orbitals, however, since only a few possibilities exist for investigating neutron-rich or proton-deficient nuclei.

The use of in-beam gamma-ray spectroscopy together with reactions induced by tritons has allowed us to study proton-deficient nuclei close to doubly magic ^{208}Pb . In particular, new information has been gained on the electromagnetic properties of the $h_{11/2}$ proton hole that often is a constituent of isomeric states. The magnetic moment of the $h_{11/2}$ proton orbital has been deduced from a measurement⁴ of the g factor of the $25/2^+$ isomer in ^{205}Tl , which has the configuration $\nu p_{1/2}^{-1}i_{13/2}^{-1}\pi h_{11/2}^{-1}$. In the two-proton-hole nucleus ^{206}Hg , we found an $(h_{11/2}^{-1}s_{1/2}^{-1}, 5^-)$ isomer⁵ (Fig. 100). This isomer opens the possibility of measuring a quadrupole moment by observing the time-perturbed angular distribution of the gamma-ray decay.

The 5^- isomer in ^{206}Hg was produced by the $^{204}\text{Hg}(t,p)$ reaction at the tandem Van de Graaff accelerator facility at the Los Alamos National Laboratory (Los Alamos, NM). The 16-MeV proton beam consisted of pulses 1 ns wide separated by 12.8 μs . The target of enriched polycrystalline ^{204}Hg was thick enough to stop

the beam. It was cooled to $225 \pm 5 \text{ K}$, 10 K below the melting point. The target was viewed by two Ge(Li) detectors, one placed at 0° and the other at 90° to the direction of the incident beam. For each detector, the pulse heights and the time delays with respect to the beam pulses were recorded event-by-event on magnetic tape. The data were subsequently sorted off-line to obtain the time-perturbed angular-distribution patterns. The time scale was calibrated with an accuracy of $\pm 2\%$ by means of cables. Otherwise, the experimental techniques were as described in

Figure 100. Decay of the 5^- isomer in ^{206}Hg .

Ref. 5. Energy spectra of delayed gamma rays from the reactions of 16-MeV tritons with ^{204}Hg are presented in Ref. 6.

Figure 101 shows the perturbed-angular-distribution pattern $R(t)$ of the 1034-keV E3 transition:

$$R(t) = 1/2 \left(\frac{I(0^\circ) - B(0^\circ)}{I(90^\circ) - B(90^\circ)} \right) = 3/4 [A_2 G_{22}(t)] .$$

The quantity $R(t)$ is the ratio of the count rates I at 0° and 90° after subtracting a long-lived background B . The curve fitted to the data is the theoretical time distribution $G_{22}(t)$ for spin $I = 5$ with a polycrystalline target which has axial symmetry. This fit gives the following results:

$$\nu_0 = 2.275 \pm 0.05 \text{ MHz} ,$$

$$\Delta\nu/\nu = 11 \pm 4\% ,$$

$$A_2 = 0.32 \pm 0.04 .$$

Here ν_0 is the repetition frequency of the whole pattern, or I/ν_0 is the time between the major peaks in Fig. 101. The quadrupole interaction frequency ν_Q as defined in Ref. 7 is obtained from ν_0 by applying the appropriate spin factor:

$$\nu_Q = \nu_0 \times 4I(2I - 1)/3$$

$$= 60\nu_0 = 136.5 \pm 3 \text{ MHz for } I = 5 .$$

The measured pattern shows a damping of the amplitude with time. The fitted curve reproduces the damping by using a Lorentzian distribution of frequencies rather than a single frequency; $\Delta\nu/\nu$ is the width (full width at half-maximum) of this distribution that results in the best fit to the data. A_2 is the usual angular-distribution coefficient.

The quadrupole moment eQ can be derived from the interaction frequency $\nu_Q = e^2qQ/h$ by normalizing to another measurement, and in this

way eliminating the electric-field gradient eq for a mercury nucleus in mercury metal. Haas and Shirley⁷ measured the interaction frequency for the $5/2^-$ level of ^{199}Hg in metallic mercury: $\nu_Q = 210 \pm 20 \text{ MHz}$ at liquid-nitrogen temperature. The quadrupole moment Q of this state is $0.83 \pm 0.091 \text{ b}$.^{7,8} Since our measurement was made at 225 K, a correction has to be applied for the change of the field gradient with temperature when comparing these measurements. The temperature dependence of the field gradient for a cadmium impurity in mercury has been measured by Mahnke *et al.*,⁹ and the authors give convincing evidence from similar systems that mercury in mercury should behave very similarly. The result is

$$\nu_Q(225 \text{ K}) = 0.83 \nu_Q(77 \text{ K}) ,$$

and the error of this 17% correction can be neglected. This finally gives

$$Q(^{206}\text{Hg}, 5^-) = Q(^{199}\text{Hg}, 5/2^-) \frac{136.5}{0.83 \times 210} \\ = 0.65 \pm 0.13 \text{ b} .$$

The error of 20% is a conservative estimate of the combined uncertainties in the calibration.

Our measurement is insensitive to the sign of Q ; theory predicts $Q > 0$. The present result, $A_2 = 0.32 \pm 0.04$, agrees with $A_2 = 0.38 \pm 0.05$, found in the g factor measurement,⁵ which was done under similar experimental circumstances except that the target was liquid mercury at room temperature. The pattern for $I = 5$ (Fig. 101) reproduces the details of the measured distribution well, while a fit for $I = 4$, the spin proposed in Ref. 10 for the 2.102-MeV state, visibly deviates and gives a χ^2 that is larger by a statistically significant amount. Therefore the pattern provides independent evidence of $I = 5$ for the ^{206}Hg isomer. We did not try a fit for $I = 6$, which is excluded by the gamma-ray half-

Figure 101. Measured quadrupole pattern for the 1034-keV transition from ^{206}Hg , 5^- in solid mercury metal. The ratio of the count rate at 0° to that at 90° is shown as a function of time after the beam pulse. The fit is for $I = 5$ and polycrystalline mercury. The time calibration is 9.96 ns/channel.

life. It might be noted that the shape of the time distribution depends only on the crystalline structure and the spin of the isomeric state. The quadrupole interaction frequency gives a scale factor in time, and the A_2 coefficient of the gamma ray determines the overall amplitude. The 1068-keV E2 transition ($2^+ \rightarrow 0^+$) that also occurs in the decay of the isomer has only half the anisotropy of the E3 transition; therefore its time distribution could not be fitted. In this experiment we also observed the gamma-ray lines from the decay of the $25/2^+$ isomer in ^{205}Tl , but could not discern any time-perturbed angular distribution pattern for them.

The quadrupole moment gives directly the reduced E2 matrix element:

$$\begin{aligned}\langle 5^- || M(E2) || 5^- \rangle &= 1.38eQ(5^-) \\ &= 90 \pm 18 \text{ e fm}^2.\end{aligned}$$

From this we can deduce the matrix element $\langle h_{11/2}^{-1} || M(E2) || h_{11/2}^{-1} \rangle$, as presented in Table 38. For a pure $(h_{11/2}^{-1}s_{1/2}^{-1}, 5^-)$ configuration, only angular-momentum coupling is required since the $s_{1/2}$ particle does not contribute. In this case the result is $96 \pm 19 \text{ e fm}^2$, as seen in the first column of Table 38.

Wave functions of the 5^- level that include configuration mixing have been calculated by Herling and Kuo^{11,12} in three different approximations, and by Ma and True.¹³ We use these wave functions with the single-particle E2 matrix elements of Donahue *et al.*¹⁴ to calculate the contributions of the small components to $Q(5^-)$. In each of the four wave functions, the amplitude of the main $h_{11/2}^{-1}s_{1/2}^{-1}$ component is more than 0.95, and the contributions to the quadrupole moment of the small admixtures

partially cancel. Therefore the impurities in the 5^- wave function have only a small effect on the corrected values (Table 38, columns 1–6) for the reduced E2 matrix element of the $h_{11/2}$ proton hole.

In a very first approximation, we might compare the $h_{11/2}$ moment with the measured $\langle h_{9/2} || M(E2) || h_{9/2} \rangle$ as given in column 7 of Table 38. Both matrix elements should be of equal magnitude (and opposite sign) if the radial wave functions are identical. Evidently, this comparison makes sense within a factor of 2, but the difference exceeds the experimental errors.

A shell-model calculation¹⁵ gives $\langle h_{11/2} || (E2) || h_{11/2} \rangle = 42 \text{ e fm}^2$. The ratio of the measured moment to this value gives an effective charge $e_{\text{eff}}(h_{11/2}^{-1}) = 2.2 \pm 0.5 \text{ e}$, or a polarization charge $e_{\text{pol}} = e_{\text{eff}} - e = 1.2 \pm 0.5 \text{ e}$. Astner *et al.*¹⁶ surveyed polarization charges in the lead region for good shell-model states. They found $e_{\text{pol}}(\pi h_{9/2}) = 0.53 \pm 0.04 \text{ e}$ and $e_{\text{pol}}(\nu i_{13/2}^{-1}) = 0.96 \pm 0.4 \text{ e}$. The polarization charge includes both isoscalar and isovector terms, and the isovector term accounts for most of the difference in the two cited numbers. Therefore, again one expects the E2 properties of $\pi h_{9/2}$ and $\pi h_{11/2}$ to be similar. Our measured $h_{11/2}$ moment is closer to that of the $\nu i_{13/2}^{-1}$, but the error extends close to the $\pi h_{9/2}$ value.

Ring, Bauer, and Speth¹⁵ calculated electric quadrupole moments for nuclei around ^{208}Pb in the theory of finite Fermi systems. This calculation explicitly considers the excitations of the ^{208}Pb core, which are otherwise parameterized by the effective charge. Their result, as given in column 9 of Table 38, agrees with our measurement.

This research was supported by the LLNL Nuclear Chemistry Research Program.

Table 38. Evaluation of $\langle h_{11/2}^{-1} || M(E2) || h_{11/2}^{-1} \rangle$ (e fm²) for different 5^- wave functions, and comparison with theory (see text).

Pure	Evaluation of experiment					$h_{9/2}^c$	Theory	
	KH1 ^a	KH2 ^a	KH3 ^a	MT ^b	Adopted		Shell	RBS ^d
96 ± 19	89	92	92	92	92 ± 20	-51	42	75

^a Refs. 11 and 12.

^b Ref. 13.

^c Ref. 14.

^d Ref. 15.

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The Decay of the 3291-keV $J^\pi = 25/2^+$ Level in ^{205}Tl

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We observed a $J^\pi = 25/2^+$ isomer in ^{205}Tl using techniques of in-beam gamma-ray spectroscopy and the $^{204}\text{Hg}(t,2n)^{205}\text{Tl}$ reaction. The decay scheme firmly establishes the yrast levels: [J^π , E_x (keV)] $11/2^-$, 1484.02; $15/2^-$, 2054.57; $17/2^-$, 2394.18; $19/2^-$, 2551.56; and $25/2^+$, 3290.7. Calculations of angular-distribution coefficients for transitions with pure multipolarity are in excellent agreement with experiment. We compare level energies with shell-model calculations.

The orbital gyromagnetic ratio g_ℓ of the $1h$ proton orbital has been reported⁶ as $g_\ell = 1.115 \pm 0.02$. This value resulted from the appropriate combination of the gyromagnetic ratios of the spin-orbit partners $g(h_{9/2})$ and $g(h_{11/2})$, so it is relatively free of contributions due to both core polarization and spin magnetism. We deduced the value of $g(h_{11/2})$ from the magnetic moment of the 3291-keV level in ^{205}Tl ; this newly reported state has spin parity $J^\pi = 25/2^+$ and the major configuration $(\pi h_{11/2}^{-1}, \nu i_{13/2}^{-1} p_{1/2}^{-1})$. The measurement of the magnetic moment and the deduction of $g(\pi h_{11/2})$ and $g_\ell(1h)$ have been described in Ref. 6; here we present the evidence for the $J^\pi = 25/2^+$ assignment, and describe the gamma decay of the 3291-keV level.

States in ^{205}Tl were populated with the $^{204}\text{Hg}(t,2n)^{205}\text{Tl}$ reaction. A target of liquid ^{204}Hg ($>98.2\%$ ^{204}Hg) was bombarded with tritons accelerated by the tandem Van de Graaff generator at the Los Alamos National Laboratory (Los Alamos, NM). The target was thick enough to stop the beam at all the incident triton energies used [$10.5 \leq E_t$ (MeV) ≤ 16.5]. Beam currents generally were about 1 nA. The resulting gamma radiation was detected in germanium and silicon solid-state detectors. After electronic filtering, signals were sorted and stored using an on-line computer-based data-acquisition system.⁷

We made several measurements to establish the excitation energy, decay, and J^π of the isomeric level. The pulsed-beam technique was used to search for the isomeric level reported

previously.⁸ The incident 14.2-MeV beam was bunched into pulses about 1 ns in duration and 12.8 μs apart. For each detected gamma-ray event, both its pulse height and time relative to the beam burst were stored. Gamma-ray pulse-height distributions were constructed from these data, according to (relative) event time. The event time was parameterized in five intervals, the first corresponding to beam on target, while the other four were contiguous sequential intervals of about 1.55 μs each. Several gamma rays were observed to decay with a mean life τ of about 3 to 4 μs , and for these the ratio R of the yield in the prompt time interval to the yield in the first delayed time interval was formed. The first two columns in Table 39 show the gamma rays associated with the isomer (listed according to increasing R), together with R .

Using the results of gamma-gamma-time coincidence measurements, we confirmed that the gamma rays listed in Table 39 are the predominant decay transitions of the isomer. We made the measurements at $E_t = 16$ MeV using two germanium detectors to detect gamma rays. They were located at $\theta_\gamma = \pm 90^\circ$ with respect to the incident beam direction and about 3 cm from the target center. Figure 102 shows an example of the results: the spectrum in coincidence with the 339-keV gamma ray. The predominant decay gamma rays of the isomer, except for the 496.9-keV gamma ray, are present in this spectrum. Gamma-ray energies are included in the table.

We measured the magnetic moment of the isomeric level at $E_t = 16$ MeV using the time-differential perturbed-angular-correlation

Table 39. Summary of experimental data on the decay of the 3291-keV isomer in ^{205}Tl .

E_γ^a (keV)	R^b	I_γ^c	$L_\gamma \times (1 + \alpha_t)^d$	E_x^a (keV)	A_2^e expt; calc	A_4^e expt; calc	$f_i^\pi \rightarrow f_f^\pi$	ML; δ^f
739.16 $\pm$ 0.10	0.7 $\pm$ 0.1	100	100	3290.72 $\pm$ 0.20	+0.50 $\pm$ 0.01	+0.05 $\pm$ 0.01; +0.03 $\pm$ 0.01	25/2 ⁺ 19/2 ⁻	E3
157.38 $\pm$ 0.10	8 $\pm$ 2	24	82	2551.56 $\pm$ 0.16	-0.25 $\pm$ 0.01	-0.01 $\pm$ 0.01; 0.00	19/2 ⁻ 17/2 ⁻	M1/E2; -0.03 $\pm$ 0.01
496.94 $\pm$ 0.10	—	13	13	2551.56 $\pm$ 0.16	—	—	19/2 ⁻ 15/2 ⁻	—
339.61 $\pm$ 0.05	15 $\pm$ 1	53	66	2394.18 $\pm$ 0.13	-0.36 $\pm$ 0.01	+0.01 $\pm$ 0.01; 0.00	17/2 ⁻ 15/2 ⁻	M1/E2; -0.12 $\pm$ 0.02
570.55 $\pm$ 0.10	56 $\pm$ 5	100	99	2054.57 $\pm$ 0.12	+0.29 $\pm$ 0.01; +0.28 $\pm$ 0.01	+0.06 $\pm$ 0.01; -0.07 $\pm$ 0.01	15/2 ⁻ 11/2 ⁻	E2
54.38 $\pm$ 0.03	—	—	—	1484.02 $\pm$ 0.07	-0.20 $\pm$ 0.01; -0.19 $\pm$ 0.01	-0.01 $\pm$ 0.01; 0	11/2 ⁻ 9/2 ⁺	E1
505.75 $\pm$ 0.10	110 $\pm$ 10	62	80	1429.64 $\pm$ 0.06	-0.32 $\pm$ 0.01	-0.01 $\pm$ 0.01; 0.00	9/2 ⁺ 7/2 ⁺	M1/E2; -0.10 $\pm$ 0.02
720.09 $\pm$ 0.05	227 $\pm$ 35	83	100	923.89 $\pm$ 0.06	+0.28 $\pm$ 0.01; +0.28 $\pm$ 0.01	-0.05 $\pm$ 0.01; -0.07 $\pm$ 0.01	7/2 ⁺ 3/2 ⁺	E2
203.67 $\pm$ 0.05	217 $\pm$ 10	80	115	203.75 $\pm$ 0.04	+0.34 $\pm$ 0.01	0.00 $\pm$ 0.01; 0	3/2 ⁺ 1/2 ⁺	M1/E2; 1 < δ < 3

^a Including data from Ref. 13.^b R is defined in the text.^c Measured delayed-gamma intensities ($\pm 10\%$) normalized to $I(739 \text{ keV}) = 100$ from the g factor measurement.^d Total delayed-transition intensities ($\pm 20\%$) including electron conversion, corrected for known parallel branches.¹³ Conversion coefficients are from Ref. 9 except for $\alpha_{\text{tot}}(203) = 0.44$ (Ref. 10).^e See text, Eq. (1).^f Gamma-ray multipolarity (ML) and mixing ratio (δ).

technique. The experimental arrangement and typical data obtained are described in Ref. 6. Data for the 497-keV gamma ray were not extracted because of its low intensity. Least-squares techniques were used to fit the time distribution of gamma-ray yields $[N(t)]$ to the expression

$$N(t) = A_0 e^{-\lambda t} (1 + A_2 P_2 \cos \theta + A_4 P_4 \cos \theta) \quad (1)$$

where P_i are Legendre polynomials, with

$$\theta = \omega_1 t + \psi + \gamma \quad (2)$$

From this procedure, we deduced the decay constant λ , the Larmor frequency ω_L , possible phase shifts ψ , and the angular-distribution coefficients A_2 and A_4 . The detector angle with respect to the beam direction ($\pm 135^\circ$) is expressed by γ . Angular-distribution coefficients are summarized in Table 39.

Spins and parities of the levels above the known 11/2⁻ state at $E_x = 1484$ keV were inferred from these data. The isomeric 739-keV transition was assigned ($J \rightarrow J - 3$, E3) based upon its lifetime and angular distribution. By assuming a pure multipole transition, the

alignment of the isomer was determined from the measured coefficient A_2 (739 keV).

Attenuation coefficients were found to be $\alpha_2 = 0.71 \pm 0.02$ for A_2 (739 keV), and $\alpha_4 = 0.38 \pm 0.05$ with the assumption of a Gaussian distribution of the population of the magnetic substates.¹¹ The A_2 and A_4 coefficients for all members of the gamma-ray cascade were then calculated; these are given in Table 39.

Transitions of pure multipolarity show excellent agreement between calculation and experiment in both A_2 and A_4 coefficients for the adopted spin sequence. Multipole mixing ratios δ for M1/E2 transitions were deduced from the measured A_2 coefficients; here the values of the A_4 coefficients eliminate second solutions with $|\delta| \gg 1$.

Further experimental evidence supports the spins and parities of the states involved in the cascade. The evidence includes the following. (1) There is agreement with the 1/2⁺, 3/2⁺, 7/2⁺, and 11/2⁻ assignments, which were established before.¹² (2) The measured g factor suggests the isomer is 25/2⁺ or, much less likely, 23/2⁺. (3) Intensity balance requires a conversion coefficient of more than 2 for the 157-keV transition; therefore we assign it multipolarity

Figure 102. Spectrum in coincidence with the 339-keV gamma ray produced in the bombardment of ^{204}Hg with 16-MeV tritons.

M1. (4) Since the 339-keV transition has mixed multipolarity, it has multipolarity M1/E2 and not E1/M2.

Figure 103 shows the decay scheme. It includes some weak branches between the lowest levels. These are placed by an analysis of prompt gamma-gamma coincidences.¹³ The intensities of these weak branches and corrections for internal conversion account for the missing intensity of the main cascade.

Our results agree with previous work¹² on the levels below the 1484-keV $11/2^-$ state, and the $9/2^+$ and $11/2^-$ assignments are now firm. Levels of higher spin have been observed in $^{208}\text{Pb}(p,\alpha)$ by Smith *et al.*¹⁴ and are shown in Fig. 103 for comparison. Our present assignments are in agreement with the ℓ transfers deduced in Ref. 14, except for the 2394- and 2551-keV levels, where the earlier work indicated an $\ell = 8$

transfer instead of the $\ell = 9$ transfer required by the present data.

Calculations by Blomqvist^{8,15} predicted the $25/2^+$ and $19/2^-$ levels extremely well, as illustrated in Fig. 103. This calculation uses experimental matrix elements. A calculation with the Kuo-Herling interaction has been performed by Silvestre-Brac and Boisson.¹⁶ The resulting level energies are also shown in Fig. 103. They reproduce the $15/2^-$, $13/2^-$, and $25/2^+$ levels well. On the other hand, the $17/2^-$ state is not predicted, and the predicted $21/1^+$ state is unlikely to be correct, at least with an energy as low as 100 keV below the $25/2^+$ level, as the existence of such a state would destroy the isomerism of the $25/2^+$ level.

This work was supported by the LLNL Nuclear Chemistry Research Program.

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Figure 103. Decay scheme of the 25/2⁺ isomer compared both with levels deduced in Ref. 14 and levels predicted theoretically. The columns labeled Theory (A) and Theory (B) were produced by Blomqvist,^{8,15} and Silvestre-Brac and Boisson,¹⁶ respectively. Theoretical level energies are normalized to the experimental location of the 11/2⁻ level.

Electromagnetic Properties of Isomers in ^{210}Pb

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We measured the lifetimes and magnetic moments of the $J = 8^+$ and 6^+ isomers in ^{210}Pb by using the gamma-ray perturbed-angular-distribution technique. The levels were populated with the $^{208}\text{Pb}(t,p)^{210}\text{Pb}$ reaction. We found the g factors of the 8^+ and 6^+ states to be -0.312 ± 0.008 and -0.312 ± 0.015 , respectively, and we deduced the magnetic moment μ to be $-1.42 \pm 0.07 \mu_N$ for the $2g_{9/2}$ neutron orbital. Lifetimes are 290 ± 25 ns for the 8^+ state and 71 ± 9 ns for the 6^+ state. We found $\langle g_{9/2} || M(E2) || g_{9/2} \rangle = -39 \pm 2 \text{ e fm}^2$, and an effective charge of $0.88 \pm 0.05 \text{ e}$ for the $2g_{9/2}$ neutron orbital.

The low-lying levels of the nucleus ^{210}Pb , with just two neutrons outside the ^{208}Pb core, are well described as members of the $\nu g_{9/2}^2$ configuration. The energies of the high-spin members of this configuration are separated by less than 100 keV, with the result that the 8^+ and 6^+ levels have lifetimes of the order of 100 ns. Lifetimes of that magnitude are ideally suited for pulsed-beam gamma-ray measurements, from which accurate g factors and $B(E2)$ values can be determined. Furthermore, because these high spin states are quite pure, one can use simple additivity rules to extract from these measurements the magnetic moment and effective charge for the $2g_{9/2}$ neutron orbital.

The members of the $\nu g_{9/2}^2$ configuration in ^{210}Pb have been identified by charged-particle reactions^{3,4} and are shown in Fig. 104. Previous in-beam gamma-ray measurements, which utilized heavy-ion-induced reactions,⁵ gave mean lifetimes of 225 ± 22 and 30 ± 10 ns for the 8^+ and 6^+ levels, respectively. We used the $^{208}\text{Pb}(t,p)^{210}\text{Pb}$ reaction and the time-differential perturbed-angular-distribution (DPAD) method to determine the g factors of these isomers and to improve the accuracy of the lifetime measurements. Time distributions of the gamma rays were measured at angles of $\pm 135^\circ$ with respect to the beam direction. We obtained the DPAD pattern by forming the ratio $R(t)$ of the intensities I , as shown by the equation

$$R(t) = \frac{I(135^\circ, t) - I(-135^\circ, t)}{I(135^\circ, t) + I(-135^\circ, t)}, \quad (1)$$

which yields

$$R(t) = [3A_2/(4 + A_2)] \sin 2\omega_L t. \quad (2)$$

Figure 105 shows the ratio function for the 297-keV $4^+ \rightarrow 2^+$ transition, along with a fit to the sine function. The resulting Larmor precession frequency ω_L corresponds to a g factor of -0.311 ± 0.005 ; the value of the angular-distribution coefficient A_2 , $+0.28 \pm 0.04$, is reasonable for a stretched E2 transition. It is important to note that the same frequency is obtained for fits over different regions of the time distributions. Since these distributions have contributions both from the 6^+ and 8^+ isomers, we can conclude that the g factors of these states are the same within approximately 2%. This is predicted by the additivity principle for states with pure $(j)^n$ configurations. After correcting for the Knight shift and diamagnetic shielding, we obtain

$$g_{8^+} = -0.312 \pm 0.008.$$

We determined the lifetime of the 8^+ state by fitting the sum of the normalized time distributions from the two detectors to the function $I_0 \exp(-t/\tau)$ for $t > 300$ ns. In this range the contribution from the 6^+ state should be negligible. This results in $\tau_{8^+} = 290 \pm 25$ ns,

which is about 30% larger than the previously reported values for which the measuring range was limited to less than 500 ns. We determined the lifetime of the 6^+ state by fitting the entire time range to the function that describes successive decay through two isomers with different lifetimes. Figure 106 shows the fit with the experimental data for the 297-keV transition. We find that the mean lifetime of the 6^+ state is 71 ± 9 ns.

Assuming the 8^+ state in ^{210}Pb has only the $\nu g_{9/2}^2$ configuration, our measured g factor gives $-1.40 \pm 0.04 \mu_N$ for the magnetic moment of the $2g_{9/2}$ neutron orbital. The effects of small admixtures of other configurations can be taken into account by uncoupling the two-particle wave functions and summing over the

Figure 104. Members of the $\nu g_{9/2}^2$ configuration in ^{210}Pb . Gamma-ray energies are from the present work. The energy of the 8^+ level is taken from the (t,p) and (t,t') reaction studies described in Refs. 3 and 4.

contributions of the single-particle matrix elements. Using the wave functions of Kuo and Herling⁶ we find $\mu(\nu g_{9/2}) = -1.46 \pm 0.07 \mu_N$. This value is comparable to the value of $-1.39 \pm 0.07 \mu_N$ (Ref. 7) for $\mu(\nu g_{9/2})$, extracted from the $(\pi h_{9/2} \nu g_{9/2}, 7^-)$ isomer in ^{210}Bi . The Schmidt value for the $g_{9/2}$ neutron orbital is $-1.91 \mu_N$. The deviation between the Schmidt value and the measured value is largely due to core-polarization effects. Petrovich⁸ used an effective operator formalism to calculate the magnetic moments for several single-particle orbitals in the ^{208}Pb region. His result of $-1.42 \mu_N$ is in excellent agreement with the experimental values.

Using the lifetimes of the 8^+ and 6^+ states, we calculated the $B(E2)$ values for the two transitions. We used transition energies of 82 ± 5 and 97.9 ± 1 keV, and total conversion coefficients of 15.2 and 7.13, respectively. The resulting $B(E2)$ values were $46 \pm 5 \text{ e}^2\text{fm}^4$ for the $8^+ \rightarrow 6^+$ transition, and $158 \pm 20 \text{ e}^2\text{fm}^4$ for the $6^+ \rightarrow 4^+$ transition. The reduced E2 matrix element $\langle 6^+ \| M(E2) \| 8^+ \rangle = 28 \pm 1 \text{ e fm}^2$ can be used to extract the reduced matrix element for the $2g_{9/2}$ neutron by a recoupling procedure similar to that used for the magnetic-moment analysis. We obtain $\langle g_{9/2} \| M(E2) \| g_{9/2} \rangle = -39 \pm 2 \text{ e fm}^2$, which is in good agreement with the value of $-38 \pm 4 \text{ e fm}^2$ from ^{210}Bi . We can also use the $B(E2)$ value to determine the quadrupole moment of the $2g_{9/2}$ neutron orbital as $-29 \pm 2 \text{ fm}^2$. The additivity of effective charges suggests that this represents the quadrupole moment of the ground state of ^{209}Pb . Proetel *et al.*⁹ performed calculations of this state, allowing for

Figure 105. Ratio function for the 297-keV transition, as described in the text. The time calibration is 3.44 ns/channel.

admixture of the 2^+ and 3^- core states of ^{208}Pb . Their wave function,

$$|^{209}\text{Pb, g.s.}\rangle = 0.95|g_{9/2}\rangle + 0.13|2^+ \otimes g_{9/2}\rangle + 0.24|3^- \otimes j_{15/2}\rangle + \dots, \quad (3)$$

yields a value of $-35 \pm 3 \text{ e fm}^2$ for the reduced E2 matrix element. They used experimental values for the core states and zero charge for the $\nu g_{9/2}$ orbital. The second term in the wave function provides almost 90% of the value. Their result is in good agreement with our measurement.

Our data on the isomers of the $\nu g_{9/2}^2$ configuration in ^{210}Pb represent the first measurements of the electromagnetic properties of this single-particle neutron orbital that do not depend on measurements of proton properties. The results reinforce the applicability of the additivity of anomalous g factors and effective charges in the ^{208}Pb region. This applicability is reflected both in the magnetic-moment measurements, in which we observe that the g factors of the 6^+ and 8^+ states are equal, and in the agreement between our value for the $\nu(2g_{9/2})$ magnetic moment and that extracted from the $\pi h_{9/2}\nu g_{9/2}$ isomers in ^{210}Bi . Similarly, there is consistency between the reduced matrix

elements $\langle g_{9/2} \| M(E2) \| g_{9/2} \rangle$ of our work and those from ^{210}Bi . Furthermore, our results provide a good test of the shell-model wave functions calculated for these "two-particle" nuclei, and thereby of the nucleon-nucleon interactions used to generate them.

This work was supported by the LLNL Nuclear Chemistry Research Program.

Figure 106. Sum of the time distributions for the 297-keV transition for $+135^\circ$ and -135° . The time calibration is 3.44 ns/channel.

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Single-Particle, Cluster, and Intruder States of ^{213}Fr Studied by Gamma-Ray, Conversion-Electron, and Alpha Measurements of ^{213}Ra and $^{217\text{m}}\text{Ac}$ Decay

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We determined the levels in the $N = 126$, $Z = 87$ nucleus ^{213}Fr by heavy-ion production techniques. In-beam gamma rays and conversion electrons, in coincidence with alpha particles from $29/2^+ ^{217\text{m}}\text{Ac}$, were used to identify a $13/2^+ (i_{13/2})$ level at 1105 keV above the $9/2^- (h_{9/2})$ ground state. We also investigated the decay of 2.7-min ^{213}Ra by means of gamma-ray and conversion-electron coincidence and singles studies. ^{213}Ra electron-capture data were used to identify a 498-keV $7/2^- (f_{7/2})$ first excited state as well as higher-energy members of the $h_{9/2}^3$ states. The observed ^{213}Fr level structure compares well to shell-model calculations and demonstrates the preservation of explicit three-particle clusters. There is evidence for the presence of the $p_{3/2}$ and intruder $s_{1/2}$ and $d_{3/2}^-$ hole states.

The odd-proton nuclei beyond ^{208}Pb , with $N = 126$, fill the $h_{9/2}$ orbital with higher-lying states involving the $f_{7/2}$ and $i_{13/2}$ shell-model orbitals. For $Z = 85$, the nucleons form explicit three-particle ($h_{9/2}^3$) clusters, which yield a higher level density below 2 MeV than in ^{209}Bi with 83 protons. Few cases exist in which the preservation of the explicit three-particle $h_{9/2}$ cluster can be investigated. The excited states of ^{213}Fr offer the opportunity to extend the $N = 126$ odd-proton-nuclei systematics to five nucleons beyond ^{208}Pb . Unfortunately, the only states known to date are the $9/2^-$ ground state and high-spin states from heavy-ion in-beam gamma-ray studies.

In order to investigate the level structure of ^{213}Fr , we performed in-beam alpha-decay studies of the ^{217}Ac , $29/2^+$ isomer and electron-capture-decay studies of ^{213}Ra with J^π of $1/2^-$. These studies identified many new levels below 3 MeV in ^{213}Fr , which allowed us to compare the general level structure of ^{211}At with three-particle clusters³ to ^{213}Fr with five protons beyond a doubly closed shell and with shell-model calculations.

Figure 107 shows a schematic of the experimental arrangement for our measurements of the alpha decay of $^{217\text{m}}\text{Ac}$. We used a 75-MeV ^{12}C beam from the VICKSI facility at the Hahn-Meitner Institute (Berlin, FRG). The beam was focused on a target of ^{209}Bi about 1 mg/cm^2 thick. The recoil products were caught downstream by a catcher foil on the axis of a superconducting conversion-electron spectrometer. Two Si(Li) and alpha detectors measured electron-electron and alpha-electron coincidences.

The $^{204}\text{Pb}(^{13}\text{C},4n)$ reaction was used to produce ^{213}Ra activity. We had two separate arrangements in these experiments: one for the measurement of conversion-electron and gamma-ray coincidences, and one for gamma-ray coincidence relationships. We irradiated a ^{204}Pb target about 2 mg/cm^2 thick with a ^{13}C beam. A carbon catcher foil 0.5 mg/cm^2 thick was placed immediately behind the target to catch the recoil products. This catcher foil, one of 21 equally spaced foils, was secured to a movable belt; after an irradiation period of 200 s, the beam was shut off for 10 s while a stepping motor advanced the

Figure 107. Setup for alpha and electron measurements in the decay of $^{217\text{m}}\text{Ac}$.

foil to the front of the counting station, where it was counted for 200 s.

For the measurement of conversion-electron-gamma-ray coincidences, the foils were transported into the superconducting conversion-electron spectrometer, which was placed off-line. The superconducting solenoid was identical to the one described in Ref. 4. A germanium detector replaced one of the Si(Li) detectors of Fig. 107. For gamma-gamma coincidences, the catcher foil was transported to the center of four germanium detectors. In both configurations, we monitored the position of the catcher foil by viewing it through a port with a TV monitor.

Besides the external multigamma and conversion-electron standards, the transitions of the decay products ^{209}Ra and ^{209}At served as internal calibration points. Multiparameter data, consisting of the coincident energies, time after irradiation, and time between the events were recorded on a magnetic disk using a PDP-11 computer facility. The data were analyzed after the experiment was completed.

The strongest alpha branch from $^{217\text{m}}\text{Ac}$, with the structure $(\pi i_{13/2} \nu g_{9/2}^2, 29/2^+)$, proceeds to an

1105-keV level in ^{213}Fr , which decays with an M2 transition to the $h_{9/2}$ ground state. This is, therefore, the $i_{13/2}$ single quasi-particle state. Likewise, the beta decay gives the $f_{7/2}$ state at 498 keV and a candidate for the $p_{3/2}$ state at 2610 keV. As seen in Fig. 108, the excited single-particle states are lowered by 400 to 500 keV compared with ^{209}Bi . This trend is expected from pairing. Preliminary evaluation of the beta-decay data suggests that the 2632- and 2950-keV levels are the $s_{1/2}$ and $d_{3/2}$ single-proton-hole intruder states. Their excitation energy increases slightly with proton number.

Apart from the energies of possibly five single-particle levels, we find candidates for the $3/2^-$, $5/2^-$, $7/2^-$, and $9/2^-$ states of the $h_{9/2}^5$ configuration with seniority 3, which is now complete except for the $11/2^-$ level. This multiplet is very similar to ^{211}At .⁵⁻⁷ Also, the shell-model calculations with a surface-delta interaction, by Zwartz and Glaudemans,⁸ agree quite well, as Fig. 108 shows.

This research was supported by the LLNL Nuclear Chemistry Research Program.

Figure 108. Experimental level scheme of ^{213}Fr compared with ^{211}At , selected states in ^{209}Bi , and a shell-model calculation.

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Levels of ^{244}Cm Populated by the Beta Decay of 10-h ^{244g}Am and 26-min ^{244m}Am

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We measured gamma rays and conversion electrons for the ^{244}Am beta-decay isomers in an ^{243}Am target undergoing neutron capture in a high-flux reactor. New transitions accompanying beta decay of 26-min ^{244m}Am were observed. These included 984.92-keV and 977.80-keV E0 transitions assigned to the de-excitation of a first excited 0^+ band in ^{244}Cm with level energies of 984.91 (0^+) and 1020.76 (2^+) keV. The ratio of reduced transition probabilities $B(\text{E}0)/B(\text{E}2)$ for this 0^+ level is 1.5 ± 0.2 .

In previous studies of the beta decay of ^{244}Am isomers, no prominent gamma rays were reported for ^{244m}Am ,⁵⁻⁷ while the gamma-ray spectrum accompanying ^{244g}Am decay was well characterized.^{7,8} In this work we measured the gamma-ray and conversion-electron spectra of these ^{244}Am beta-decay isomers, and assigned new gamma transitions to the beta decay of 26-min ^{244m}Am , including a prominent E0 transition that depopulates the first excited 0^+ state of ^{244}Cm . This is the first observation of an excited 0^+ state in ^{244}Cm . The gamma transitions assigned to the beta decay of 10-h ^{244g}Am were determined with greater precision than in previous studies.

We made our experimental measurements with ^{243}Am targets undergoing neutron capture in thermal-neutron fluxes exceeding 10^{14} n/cm²s, at the High Flux Reactor of the Institut Laue-Langevin in Grenoble, France. Although we identified approximately 760 prompt secondary gamma rays, this paper is concerned only with those transitions assigned to the beta decay of the ^{244}Am isomers. We measured gamma-ray energies and intensities with two curved-crystal spectrometers, and conversion electrons with a beta spectrometer. The spectrometers, standard measuring procedures, and data-analysis methods are described in Refs. 9-11.

The decay scheme in Fig. 109 shows the results of these measurements: transition

energies, intensities, and multiplicities. Seven of these transitions, those depopulating the 6^+ level at 1040.19 keV and the cascade depopulating the 8^+ level at 501.79 keV, were already identified as transitions in ^{244}Cm following ^{244g}Am decay.⁷ The decay of this longer-lived isomer was assigned to the ground state of ^{244}Am ,⁷ a level predicted to have spin and parity $I^\pi = 6^-$ and configuration $5/2^- [523]p + 7/2^+ [624]n$.

In our recent study of $^{243}\text{Am}(n,\gamma)^{244}\text{Am}$,¹¹ we assigned spin and parity $I^\pi = 1^+$ and configuration $5/2^+ [642]p - 7/2^+ [624]n$ to the 26-min ^{244}Am isomer. This assignment, for which there exists conclusive experimental proof, contradicts the previous assignment of $I^\pi = 1^-$, thought to be the singlet state that corresponds to the $I^\pi = 6^-$ triplet ground state. We have shown experimentally that this 1^- state occurs at 173 keV in ^{244}Am .

The experimental observations for the 984.92-keV transition consisted of four electron peaks from conversion in the K, L₁, M₁, and N₁ shells of curium; no corresponding photon was observed with the curved-crystal spectrometer. Thus we interpret this transition to be of pure E0 multipolarity. The observed ratio of L₁/K conversion was 0.21 ± 0.03 , which agrees well with the theoretical values of 0.184¹² and 0.195.¹³ The most likely placement for this relatively intense transition is in the depopulation of a 0^+

Figure 109. Levels of ^{244}Cm from gamma transitions accompanying beta decay of ^{244}Am isomers. Transition intensities ($I_\gamma + I_{ce}$) per 100 neutron captures are listed following gamma-ray energies and multiplicities.

level in ^{244}Cm at 984.91 keV to the ground state. Consistent with this postulate is the observation of a 941.95-keV gamma ray that is assigned as an E2 transition depopulating this level to the 2^+ first excited state. The experimental value for the ratio of reduced probabilities of the E0 and E2 transitions depopulating this level can be expressed as $X = B(E0; 0_2^+ \rightarrow 0_1^+)/B(E2; 0_2^+ \rightarrow 2_1^+) = 1.5 \pm 0.2$.

We observed a pair of clearly defined electron peaks that are assigned to the K and L_1 conversion of a 977.796 ± 0.020 -keV transition, again with no corresponding photon detected. Since we already determined that the transition connecting the 984.91-keV 0^+ level with the ground state has a large X value, one might expect the E0 multipolarity to be a major component of the $2_2^+ \rightarrow 2_1^+$ transition. Based on that concept, the new transition defines a 2^+ state at 1020.761 keV. The rotational parameter of this excited 0^+ band is 6.0 keV, which is 17% less than that of the ground-state band.

We assign four additional transitions with energies above 1 MeV to the de-excitation of levels in ^{244}Cm following beta decay of 26-min ^{244}Am . These assignments are based upon the

observation of gamma rays and corresponding conversion-electron lines, and the relative energies of each pair. We propose that these transitions are evidence for the tentative assignment of low-spin levels at 1084.20 and 1105.91 keV. Of the possible experimentally derived spin and parity assignments indicated for these levels, one combination is consistent with assignment to the 1^- and 2^- members of a $K = 1$ rotational band, corresponding to levels observed in ^{246}Cm at 1079 and 1105 keV.

Given our experimental evidence for an excited 0^+ band in ^{244}Cm , we compare in Fig. 110 the energy and X value of the 985-keV state with those for 0_2^+ levels in other actinides. It is evident that the experimental X value for this level is larger than typical X values for 0_2^+ states in this region. Rasmussen¹⁴ has demonstrated that for collective quadrupole vibrations of deformed nuclei (beta vibrations), one expects values of $X = 4\beta_0^2 = 0.21$ for ^{244}Cm . Ragnarsson and Broglia¹⁵ have discussed 0^+ states in actinide nuclei in the framework of BCS and RPA theory. They calculated the characteristics of states termed "pairing isomers," which exist because of the inhomogeneity of

oblate and prolate levels in the vicinity of the Fermi surface. Their microscopic calculations predict a number of low-lying excited 0^+ states that are composed predominantly of proton and/or neutron pairs in oblate orbits, configurations which are not components of the ground state. In the case of excitations whose dominant components are from proton orbits, enhanced X values are predicted in the range 1.0 to 12.0. Our data indicate that the 985-keV band in ^{244}Cm has this type of structure and, in fact, the principal components in its wave function, according to our calculations, arise from the $1/2^+[400]$ and $11/2^-[505]$ proton orbits. Other 0^+ states with significant neutron amplitudes and, hence, smaller X values, are calculated to exist in curium nuclei at energies below those of states with a predominantly proton structure. The failure to observe these modes in our experimental study is consistent with their expected structure and may be ascribed to a combination of hindered beta decay and low X values.

This work was supported by the LLNL Nuclear Chemistry Research and Nuclear Test Programs.

Figure 110. Experimental energies and X values for the first excited 0^+ states in actinide nuclei.

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Nuclear Structure of ^{244}Am Excited with the (n,γ) Reaction

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We studied secondary gamma rays and conversion electrons of the $^{243}\text{Am}(n,\gamma)^{244}\text{Am}$ reaction with crystal spectrometers and an electron spectrometer, respectively, at the Institut Laue-Langevin (Grenoble, France). We also measured primary gamma rays following thermal-neutron capture in ^{243}Am . A level scheme of ^{244}Am was constructed containing 62 excited levels. We interpret the levels in terms of coupled proton and neutron Nilsson configurations. The results are in good agreement with a semi-empirical model in which excitation energies are calculated in odd-odd nuclei from those in adjacent nuclei.

Detailed information on level schemes for odd-odd nuclei, which are the most complex nuclear species, can yield valuable information both on the coupling of single-particle states for the unpaired nucleons and on the interaction between these nucleons. Therefore, many odd-odd nuclei have been studied carefully. In deformed nuclei the proton-neutron residual interaction is manifest in the Gallagher-Moszkowski splitting and the Newby shift. These effects were recently discussed and compared with theoretical calculations by Boisson, Piepenbring, and Ogle⁶ for the rare-earth region. In spite of this interest, few deformed odd-odd nuclei have been studied in detail in the actinide region. The structures of several nuclei have been characterized from alpha- and beta-decay studies, and a few nuclei have been examined with (d,p) and (d,t) reactions. Just three level schemes for actinide nuclei have been established with the (n,γ) reaction, namely, those of ^{238}Np (Refs. 7 and 8), ^{242}Am (Ref. 9), and ^{250}Bk (Ref. 10).

Experimental data for the ^{244}Am level scheme were rather sparse and not very well understood before this investigation. Spin and parity values of 6^- and 1^- had been assigned to the ground state and 69 ± 10 -keV isomer, respectively, from beta-decay experiments by Vandenbosch and Day.¹⁰ They proposed the proton and neutron Nilsson configurations $5/2^- [523] \pm 7/2^+ [624]$ for these states. A study of the $^{243}\text{Am}(d,p)^{244}\text{Am}$

reaction¹¹ (with a resolution of 15 keV) has yielded many levels and some tentative Nilsson-configuration assignments. In order to provide a precise level scheme of ^{244}Am , which is necessary for comparison to theoretical predictions, we studied the $^{243}\text{Am}(n,\gamma)^{244}\text{Am}$ reaction with high-precision gamma and electron spectrometers.

We performed all experiments with ^{243}Am targets undergoing thermal-neutron capture in the high-flux reactor at the Institut Laue-Langevin (Grenoble, France). Secondary gamma rays in the energy range 30 to 400 keV were measured with the GAMS 1 curved-crystal spectrometer, and gamma rays in the energy range 200 to 1200 keV were measured with the GAMS 2/3 curved-crystal spectrometer. The target consisted of 50 mg of $^{243}\text{AmO}_2$ (99.9% isotopic purity). We measured primary gamma rays with a pair spectrometer installed at the same beam tube and detecting gamma rays from the same target as the curved-crystal spectrometers. The conversion-electron spectrum of secondary transitions was scanned with the beta spectrometer BILL. The target for this spectrometer consisted of 4 mg of ^{243}Am . The spectrometers, standard measuring procedures, and data-analysis methods are described in Refs. 12 and 13.

We find experimental evidence that the lowest-energy low-spin band in ^{244}Am has $K^\pi = 1^+$, in contrast to previous assumptions, and that

Figure 111. Rotational bands and Nilsson configurations in ^{244}Am . Tentative levels are indicated by dashed lines. Tentative configuration assignments are given by parentheses. Inverted triangles indicate level population by primary gamma rays from thermal-neutron capture; open triangles indicate that two levels exist at the energy of the primary transition. Circles indicate possible population of a level by the (d,p) reaction.

Figure 112. Comparison of experimental (left) and predicted (right) level energies.

a $K^\pi = 1^-$ band exists at 173 keV. This hypothesis is supported by two important experimental facts: (1) complete population of the $I = 1$ to 4 levels in an 85-keV band by primary transitions (presumably E1 in character), which decay from the $I^\pi = 2^-$ or 3^- capture states, and (2) complete population of the $I^\pi = 2$ to 5 levels in a 173-keV $K^\pi = 1^-$ band by the

(d,p) reaction (denoting a $5/2[523]$ Nilsson configuration for the spectator proton). The energy of this 1^+ isomeric state has been set at 85.0 ± 3.0 keV, based on the energy of the primary gamma ray feeding this level and a neutron binding energy of 5363 ± 3 keV.¹⁴

Assuming that the spins and parities of levels in the two low-lying $K = 1$ bands were well

established, we identified additional levels by the use of the Ritz combination principle and correspondence with level energies defined by primary gamma transitions and the (d,p) reaction population. We proposed (K, I) quantum numbers for many levels by looking for the best agreement with the Alaga rule. Figure 111 shows a level scheme established on that basis, containing 62 excited levels with 287 transitions.

A prediction of the level structure of an odd-odd deformed nucleus can be obtained if the following assumptions are made:

1. Each quasi-particle excitation energy of the odd proton and odd neutron is similar in an odd-odd nucleus and in the adjacent odd-proton and odd-neutron nuclei.

2. The effective moment of inertia of each rotational band consists of the effective moment of inertia of the even-even core plus effective additional contributions from the odd proton and the odd neutron.

3. Gallagher-Moszkowski splittings and Newby shifts can be calculated by assuming a form for the proton-neutron residual interaction. The strength of this interaction is found by fitting calculated results to experimental data. More details of this model may be found in "Level-Structure Modeling of Odd-Odd Deformed Nuclei," in this section.

Figure 112 shows the results of the calculations for ^{244}Am along with the experimental level energies. Odd-nucleon quasi-particle excitations and rotational parameters in ^{243}Am , ^{245}Am , ^{243}Pu , and ^{245}Cm were taken from Ref. 15. Even-even rotational parameters were obtained from ^{244}Pu and ^{244}Cm . Piepenbring and Boisson¹⁶ calculated the Gallagher-Moszkowski splittings E_{GM} and Newby shifts E_{N} by assuming a delta force

between proton and neutron. We reduced the E_{GM} values by a scale factor of 0.6, derived from fitting other Gallagher-Moszkowski splittings in actinide nuclei. For configurations where calculated values of E_{GM} or E_{N} were not available, we assumed values of 80 or 30 keV, respectively.

We interpreted the levels in ^{244}Am in terms of coupled proton and neutron Nilsson configurations by comparing the experimental level scheme with the model prediction just discussed. The (d,p) data of Grotdal *et al.*¹¹ can be related to the present scheme if an absolute energy error of about 10 keV is attributed to the (d,p) measurement. Six Nilsson configurations can be unambiguously assigned to rotational bands with band-head energies below 400 keV, while assignments at higher energies are less certain.

Figure 112 demonstrates that the level-energy predictions obtained with the simple semi-empirical model described previously agree very well with our experimental results. The average deviation between experimental and predicted band energies and moments of inertia are 21 keV and 8%, respectively. This agreement is much better than for odd-odd nuclei in the rare-earth region. Thus, neither the quasi-particle energy nor the effective moment of inertia of an unpaired nucleon is affected appreciably when another unpaired nucleon of opposite isospin is added to the nucleus. The main reason is the rigid and well-deformed even-even core of an actinide odd-odd nucleus. It is interesting to note that even the Coriolis interaction is relatively unmodified by transfer from an odd nucleus to an odd-odd nucleus, as evidenced by the effective moment of inertia.

Table 40. Theoretical and experimental Gallagher-Moszkowski splittings E_{GM} and Newby shifts E_{N} .

Configuration	E_{GM} (keV)		E_{N} (keV)	
	Experiment	Calculation ^a	Experiment	Calculation ^a
$5/2^- [523] \pm 7/2^+ [624]$	199	189	—	—
$5/2^- [523] \pm 5/2^+ [622]$	—	87	+25	—14
$5/2^- [523] \pm 1/2^+ [631]$	69	55	—	—
$5/2^- [523] \pm 7/2^- [743]$	—	87	—	—
$5/2^+ [642] \pm 5/2^+ [622]$	—	36	—	—54
$5/2^+ [642] \pm 1/2^+ [631]$	—	63	—	—
$7/2^+ [633] \pm 7/2^+ [624]$	—	—	+32	—

^a Calculated by Piepenbring and Boisson¹⁶ and reduced by a factor of 0.6.

Since the (n,γ) reaction excites mainly levels with $I \leq 4$, few band heads with larger values for the K quantum number were identified in ^{244}Am ; thus, only two pairs of bands representing both couplings of the proton and neutron configurations were observed. Table 40 compares the extracted splittings with theoretical values. The experimental values tend to be slightly higher (5% and 25%), and the agreement is reasonably good. The Newby shift of +26 keV determined for the $5/2^- [523] -$

$5/2^+ [622]$ band is very similar to the same shift observed in ^{242}Am ,⁹ but it is in disagreement with the calculated value of -14 keV of Piepenbring and Boisson.¹⁶ The experimental value for the $7/2^+ [633] - 7/2^+ [624]$ band is +32 keV; no corresponding calculated value is available.

This work was supported by the LLNL Nuclear Chemistry Research and Nuclear Test Programs.

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Level-Structure Modeling of Odd-Odd Deformed Nuclei

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We extended the use of a technique for modeling quasi-particle excitation energies and rotational parameters in odd-odd deformed nuclei. The input parameters required for the calculation were derived from empirical data on single-particle excitations in neighboring odd-mass nuclei. Configuration-specific values for the Gallagher-Moszkowski splittings were used, a feature approximated by a single constant value in many earlier modeling efforts. We compare calculated and experimental level structures for ^{238}Np , ^{244}Am , ^{250}Bk , ^{160}Tb , ^{166}Ho , ^{170}Tm , ^{176}Lu , and ^{182}Ta . The agreement for the actinide species is excellent, with band-head energies deviating 25 keV and rotational parameters 5% on the average. Corresponding deviations for the rare-earth nuclei are 55 keV and 7%. We discuss several applications of this modeling technique.

Our approach to modeling the level structure of odd-odd deformed nuclei is based upon two simple but very useful concepts. The first is that in considering the coupling of two odd particles to a deformed core, the excitation energy of a given configuration can be described in simplest terms as the sum of the two odd-nucleon excitations. The second concept is that the effective moment of inertia for a rotational band can be expressed as the sum of three components: the moment of inertia of the even-even core plus increments from the addition of each of the odd nucleons.

The modeling technique we used was first described in detail by Struble *et al.*³ and Motz *et al.*⁴ They proposed that if the proton-neutron residual interaction energy is small compared to the energy with which the odd nucleons are bound to the core, the excitation can be calculated by a simple extension of the odd-A model, and the interaction energy can be treated later as a perturbation. This residual proton-neutron interaction destroys the degeneracy of the two states formed by parallel and antiparallel coupling of the odd-nucleon angular momenta. Precise rules for the ordering of the two states with respect to energy have been formulated and

shown to apply experimentally by Gallagher and Moszkowski.⁵ Thus, the separation between these pairs of states is designated as the Gallagher-Moszkowski splitting.

The excitation of an odd nucleon in a deformed nucleus can be treated theoretically by various versions of single-particle potential theory. For our purposes, however, we can obtain these excitations empirically from experimental data for odd-mass nuclei having the same number of nucleons as the modeled species. Thus, as input to our model calculations, we used empirical data almost exclusively, although theoretical calculations served to guide the interpretation of these basic excitations.

Another manifestation of the proton-neutron interaction in odd-odd nuclei is the appearance of an energy shift between the states of odd and even angular momentum within a given $K = 0$ band, the so-called "Newby shift."⁶ This quantity and the Gallagher-Moszkowski splitting energy are functions of the residual interaction. The form and evaluation of this residual interaction have been the subject of several articles⁷⁻¹⁰; a majority of these have evaluated matrix elements for use in the rare-earth deformed region.

The effective moment of inertia for a given rotational band, θ , can be derived by the following method, first described by Struble *et al.*³:

$$\begin{aligned}\theta_{\text{odd-odd}} &= \theta_{\text{even-even}} + \delta\theta_p + \delta\theta_n \\ &= \theta_{e-e} + (\theta_p - \theta_{e-e}) + (\theta_n - \theta_{e-e}) \quad (1) \\ &= \theta_p + \theta_n - \theta_{e-e}.\end{aligned}$$

Earlier, Prior¹¹ had considered the incremental change of the moment of inertia caused by the presence of an odd nucleon; modeling odd-odd moments of inertia was also treated by Scharff-Goldhaber and Takahashi.¹² These calculated effective moments of inertia for rotational bands in an odd-odd nucleus are affected by variations in rotational parameters of the odd-mass nuclei caused by Coriolis interactions in those cases where significant interaction occurs.

We developed a computer program which calculates the level structure of an odd-odd deformed nucleus based on the assumptions discussed above. The quasi-particle energy for a given orbital in an odd-mass nucleus is given by the expression

$$\begin{aligned}E_I &= E_{qp} + \hbar^2/2\theta [I(I+1) - K^2 \\ &\quad + \delta_{K,1/2} a(I+1/2)(-1)^{I+1/2}] \quad , \quad (2)\end{aligned}$$

where a is the decoupling parameter. The term $-K^2$ was assumed in this expression instead of the often used form that contains a $-2K^2$ term. This modification has the effect of changing the magnitude (and even the sign) of the zero-point rotational energy E_{ZPR} for a given band. The quantity E_{ZPR} is the difference between E_K [defined by substituting K for I in Eq. (2)], and E_{qp} . Thus, it can be expressed as

$$E_{ZPR} = \hbar^2/2\theta (K - a\delta_{K,1/2}) \quad . \quad (3)$$

A more precise definition of E_{ZPR} includes a term containing the expectation value of $\langle j^2 \rangle$, which is usually the major contributor and which consequently yields a larger zero-point energy than that given by Eq. (3). Nevertheless, for simplicity, many authors have chosen to incorporate this term in the expression E_{qp} and to evaluate E_{ZPR} using a simple expression, as we did. For example, Ogle *et al.*¹³ adopted the form shown in Eq. (3). They suggested that there may

be correlation effects associated with residual interactions not included in their model that would tend to reduce the expectation value of $\langle j^2 \rangle$. They adopted the use of a $-K^2$ term [as in Eq. (2)] for their data analysis and tentatively tested making the coefficient of this term -1.5 . They concluded that the absolute magnitude of this coefficient could not be chosen much larger than 1.5 without destroying the qualitative agreement between model predictions and quasi-particle level systematics. Nielsen and Bunker,¹⁴ in evaluating E_{ZPR} explicitly for several Nilsson orbitals, found that a significant reduction in the estimate of $\langle j^2 \rangle$, and hence of E_{ZPR} , improved the overall energy fit of their calculations to experimental data for the holmium isotopes. We found that there is better agreement with experiment for our selected set of odd-odd nuclides if one assumes the form that contains a $-K^2$ term in Eqs. (2) and (4).

The excitation energies of levels in the odd-odd nucleus are calculated using the expression

$$\begin{aligned}E_I &= E_{qp}^p + E_{qp}^n + \hbar^2/2\theta_{\text{odd-odd}} [I(I+1) - K^2] \\ &\quad \pm E_{GM}/2 - \delta_{K,0}(-1)^I E_N \pi \quad , \quad (4)\end{aligned}$$

where π denotes the parity of the states as introduced in Ref. 7 and is equal to ± 1 for positive or negative parity. The results of this calculation are completely predictive; no experimental information from the odd-odd nucleus itself is included, only empirical data derived from neighboring odd-mass and even-even nuclei. The E_{GM} and E_N terms in Eq. (4), which are designated as the Gallagher-Moszkowski splitting and Newby shift, respectively, are functions of the effective neutron-proton residual interaction.

Several calculations of Gallagher-Moszkowski splittings and Newby shifts have been made assuming simple central forces. It has been demonstrated⁷ that the direct or Wigner force in the effective neutron-proton residual interaction does not contribute to the E_{GM} and E_N terms. Jones *et al.*⁸⁻¹⁰ demonstrated that some improvement in fitting experimental data was obtained in going from a delta force to a Gaussian force, but the addition of a tensor force without an exchange term made only a slight improvement. Boisson *et al.*⁷ worked with empirical data from rare-earth nuclei involving 43 two-quasi-particle configurations. They determined strength and range parameters for

various types of effective interaction by fitting the calculated matrix elements to the empirical data. For actinide nuclei, on the other hand, calculated E_{GM} and E_N values, such as those listed in Table 41, have not been published previously.

Tables 42 and 43 compare the experimental and calculated band-head energies and rotational parameters of ^{250}Bk and ^{244}Am . The experimental data for these nuclei include information from recent measurements using the (n,γ) and (d,p) reactions,^{15,16} as well as data from earlier measurements.¹⁷ The information on the level schemes of neighboring nuclei was taken largely from Ref. 17. The calculated results listed in Table 42 were obtained assuming a delta force between proton and neutron and a Woods-Saxon central potential. (For a detailed discussion of the method, see Ref. 7.) The calculated E_{GM} values (listed in Table 41) were reduced by a scale factor of 0.6, derived from a global fit of Gallagher-Moszkowski splittings in the actinide region. For configurations where calculated values of E_{GM} or E_N were not available, values of 80 and +30 keV, respectively, were assumed. The uncertainties in band-head energy listed in the fourth columns of Tables 42 and 43 do not represent experimental error but are derived from the spread in the two experimental values taken from the odd-mass nuclei. For example, the excitation of a given proton orbital in ^{250}Bk relative to the ground-state orbital is derived

from observations in ^{249}Bk and ^{251}Bk . The two experimental observations are averaged and their spread (plus that from the odd-neutron observations) determines the uncertainties listed.

The agreement between experimental and calculated band-head energies for these nuclides is excellent; the average deviations are ± 25 and ± 21 keV for ^{250}Bk and ^{244}Am , respectively. Similarly, the experimental rotational parameters agree extremely well with the calculated values; the average deviations are $\pm 5\%$ and $\pm 8\%$ for ^{250}Bk and ^{244}Am , respectively. This ability to predict rotational parameters proves useful in identifying the configuration of a rotational band. One can see in ^{250}Bk , for example, a clear difference between configurations that contain the $3/2^- [521]p$ orbital and those that contain the $7/2^+ [633]p$ orbital. For the former, the calculated rotational parameters have values of 5.7 to 5.8 keV. For the latter, the proton orbital is subject to strong Coriolis interaction and, hence, displays a large effective moment of inertia, as in, for example, odd-mass berkelium isotopes. The calculated values are thus smaller, 4.4 to 4.5 keV in several cases, and even 3.3 keV when this proton is coupled with a $1/2^- [761]$ neutron.

Table 44 summarizes the results of the comparisons for ^{250}Bk and ^{244}Am ; for ^{238}Np , another actinide nucleus under investigation; and for several rare-earth nuclides whose structures have been modeled. For the last set, the modeling technique used was nearly identical to

Table 41. Comparison of experimental and calculated values of Gallagher-Moszkowski splittings E_{GM} and Newby shifts E_N in ^{244}Am and ^{250}Bk .

Proton configuration	Neutron configuration	Nucleus	E_{GM} or E_N (keV)		
			Expt	Calc	Expt/Calc
5/2 ⁻ [523]	1/2 ⁺ [631]	^{244}Am	70	91.9	0.76
	1/2 ⁺ [620]	^{244}Am	21	—	—
	5/2 ⁺ [633]	^{244}Am	-25.4 (N)	—	—
	5/2 ⁺ [622]	^{244}Am	25.7 (N)	-22.1	-1.16
	7/2 ⁺ [624]	^{244}Am	200.2	314.2	0.64
3/2 ⁻ [521]	7/2 ⁺ [613]	^{250}Bk	66.4	90.7	0.73
	1/2 ⁺ [620]	^{250}Bk	110.3	173.9	0.63
7/2 ⁺ [633]	7/2 ⁺ [624]	^{244}Am	33.1 (N)	—	—
	7/2 ⁺ [613]	^{250}Bk	135.0	70.5	1.91
		^{250}Bk	-25.0 (N)	-87.7	0.29
	1/2 ⁺ [620]	^{250}Bk	83.6	91.4	0.91
	3/2 ⁺ [622]	^{250}Bk	91.2	104.1	0.88
	1/2 ⁻ [761]	^{250}Bk	38	—	—

that for the actinides; one difference was that values for E_{GM} and E_N were obtained from Ref. 6, where these matrix elements were calculated employing a more complex force, namely, a central force with intrinsic-spin polarization and with long-range and tensor contributions that were determined from a fit to experimental data.

Although the basic technique for this model was described in the mid-1960s, ours are among the few reported results in which precisely calculated E_{GM} and E_N values were incorporated into the model; previous authors most often assumed some constant average value for E_{GM} in deriving calculated band-head energies. Because of this difference, and perhaps because of an improved technique for evaluating zero-point rotational energy, we find that our predicted band-head energies show better agreement with experiment than previously. For example, in modeling ^{166}Ho levels, Struble *et al.*³ calculated the energies of nine band heads with an average deviation from experiment of 104 keV; our calculation of the same nine band heads resulted in an average deviation of 51 keV. In another publication, however, Struble and Rasmussen¹⁸ treated the theoretical prediction of six band-head energies in ^{166}Ho with greater rigor, i.e., by assuming a finite central force with Gaussian radial dependence in calculating two-body

matrix elements, assessing configuration mixing, etc. They produced a much improved fit to the experimental band-head energies, with average deviations of about 20 keV, and reproduced the ordering of the two lowest bands, which apparently violates Gallagher-Moszkowski coupling rules. In modeling ^{238}Np levels, Ionescu *et al.*¹⁹ calculated the energies of six band heads with an average deviation from experiment of 43 keV; our average deviation for this set was 31 keV.

The data in Table 44 indicate a somewhat better fit to experimental band-head energies in the calculations for actinide nuclei than for those of the rare-earth region. Similarly, the quality of the fit to experimental rotational parameters is better for the actinides. The larger deviations for the rare-earth nuclei are not compensated for by the larger magnitude of their rotational parameters, as indicated by the percentage deviations. Perhaps this better agreement with the model can be explained by a greater rigidity of the even-even core in the actinide nuclei.

Thus, the evidence shows that this modeling technique accurately reproduces experimental band-head energies and rotational parameters for these deformed nuclei. With this method, then, we can model all of the intrinsic single-particle excitations and rotational bands built on these

Table 42. Comparison of experimental and predicted band-head energies and rotational parameters for ^{250}Bk .

K^π	Configuration	Band-head energy (keV)		Rotational parameter A (keV)	
		Expt	Calc	Expt	Calc
2^-	$\{3/2^- [521]p + 1/2^+ [620]n\}$	0	0	5.77	5.70
1^-	$\{3/2^- [521]p - 1/2^+ [620]n\}$	104	99 ± 4	5.35	5.70
4^+	$\{7/2^+ [633]p + 1/2^+ [620]n\}$	36	37 ± 18	4.31	4.37
3^+	$\{7/2^+ [633]p - 1/2^+ [620]n\}$	115	88 ± 18	4.14	4.37
5^-	$\{3/2^- [521]p + 7/2^+ [613]n\}$	97	100 ± 33	5.80	5.83
2^-	$\{3/2^- [521]p - 7/2^+ [613]n\}$	146	137 ± 33	5.59	5.83
7^+	$\{7/2^+ [633]p + 7/2^+ [613]n\}$	86	114 ± 47	4.56	4.44
0^+	$\{7/2^+ [633]p - 7/2^+ [613]n\}$	175 (1^+)	110 ± 47 (1^+)	—	4.44
2^+	$\{7/2^+ [633]p - 3/2^+ [622]n\}$	212	211 ± 33	4.18	4.50
5^+	$\{7/2^+ [633]p + 3/2^+ [622]n\}$	316	287 ± 33	4.43	4.50
6^+	$\{5/2^+ [642]p + 7/2^+ [613]n\}$	406	410 ± 89	—	5.39
3^-	$\{7/2^+ [633]p - 1/2^- [761]n\}$	526	513 ± 18	3.4	3.30
4^-	$\{7/2^+ [633]p + 1/2^- [761]n\}$	566	596 ± 18	3.9	3.30
6^+	$\{7/2^+ [633]p + 5/2^+ [622]n\}$	552	557 ± 25	—	4.41
Average deviations:		25		0.26 (4.8%)	
		(for 13 bands)		(for 11 bands)	

Table 43. Comparison of experimental and predicted band-head energies and rotational parameters for ^{244}Am .

K^π	Configuration	Band-head energy (keV)		Rotational parameter A (keV)	
		Expt	Calc	Expt	Calc
1^+	$\{5/2^+[642]\text{p} - 7/2^+[624]\text{n}\}$	85	60 ± 42	3.4	3.0
1^-	$\{5/2^- [523]\text{p} - 7/2^+[624]\text{n}\}$	173	161 ± 14	5.3	5.4
2^-	$\{3/2^- [521]\text{p} - 7/2^+[624]\text{n}\}$	259	235 ± 56	5.8	5.8
0^-	$\{5/2^- [523]\text{p} - 5/2^+[622]\text{n}\}$	286 (1^-)	319 ± 31 (1^-)	5.3	5.4
3^+	$\{1/2^+[400]\text{p} - 7/2^+[624]\text{n}\}$	345	(336)	5.2	(5.4)
0^+	$\{7/2^+[633]\text{p} - 7/2^+[624]\text{n}\}$	374 (0^+)	362 ± 69 (0^+)	6.0	6.9
2^+	$\{5/2^+[642]\text{p} - 1/2^+[631]\text{n}\}$	(416)	451 ± 56	(2.7)	3.1
2^+	$\{5/2^- [523]\text{p} - 9/2^- [734]\text{n}\}$	417	426 ± 21	4.1	4.2
3^-	$\{5/2^- [523]\text{p} + 1/2^+[631]\text{n}\}$	418	442 ± 28	(5.6)	5.7
0^+	$\{5/2^+[642]\text{p} - 5/2^+[622]\text{n}\}$	(475)(2^+)	463 ± 59 (2^+)	—	3.0
2^-	$\{5/2^- [523]\text{p} - 1/2^+[631]\text{n}\}$	482	488 ± 28	(6.6)	5.7
2^-	$\{5/2^+[642]\text{p} - 9/2^- [734]\text{n}\}$	514	538 ± 49	3.2	2.6
2^+	$\{3/2^+[651]\text{p} - 7/2^+[624]\text{n}\}$	(612)	(395)	(5.8)	(6.2)
1^+	$\{5/2^- [523]\text{p} - 7/2^- [743]\text{n}\}$	(668)	665 ± 14	—	5.8
1^-	$\{3/2^- [521]\text{p} - 5/2^+[622]\text{n}\}$	678	586 ± 74	5.1	5.7
1^-	$\{5/2^+[642]\text{p} - 7/2^- [743]\text{n}\}$		826 ± 42		3.1
Average deviations:		21 (for 9 bands)		0.5 (8.4%) (for 9 bands)	

excitations in any deformed odd-odd species for which input data are available. We have a capability to model structure in some nuclei for which little or no experimental data exist. Calculated level schemes can be extended to energies somewhat higher than the ranges given in Table 44, although we realize that other kinds of excitations in these nuclei, such as vibrational motion and quasi-particle excitations involving more than two unpaired nuclei, are neglected in our limited modeling approach.

Several applications of this modeling technique are potentially useful. Among them are the re-interpretation of existing experimental data. We developed significant new interpretations of the experimental data for level schemes of ^{242}Np and ^{246}Am . Another application involved modeling the level structure of several rare-earth nuclei in order to provide input to the calculation of cross sections for their production in neutron-induced reactions. Elsewhere in this report (see "Isomer-Ratio Calculations for Neutron-Induced Reactions on Deformed Nuclei . . ." in Section 3), results are described for the calculation of isomer ratios for (n,γ) and $(n,2n)$ reactions leading to the product nuclei ^{176}Lu , ^{174}Lu , and ^{236}Np . The use of calculated discrete-level sets for these nuclei, produced with the present modeling technique, has resulted in very significant improvement in

agreement between calculated and experimental values for isomer ratios in each case.

Finally, we compared the calculated level density in ^{166}Ho for a selected set of levels, namely those for which $I^\pi = 2^+$ to 5^+ , with experimental data taken by Stelts *et al.* using the resonance-averaged capture technique.²⁰ In their experiment, they expected to observe all levels within the selected range of spin and parity up to an excitation of 1.6 MeV because of the averaging of gamma-decay widths over the many capture states. We calculated a cumulative number of levels (140) with our model that is 58% greater than the number observed experimentally, even after corrections based on estimates for levels missed experimentally. Although the difference between calculation and experiment for the levels below 0.8 MeV is not very large, 20%, there is a significant difference for the levels in the energy bin 0.8 to 1.2 MeV, and up to the limit of 1.6 MeV. Since the discrepancy raises questions about the validity both of the modeling results and the assumptions regarding the experimental technique, further experiments are being performed to study these questions in ^{176}Lu and ^{170}Tm .

This work was supported by the LLNL Nuclear Chemistry Research Program.

Table 44. Odd-odd species in actinide and rare-earth regions: comparison of experimental and calculated band-head energies, rotational parameters, and Gallagher-Moszkowski splittings (E_{GM}).

Nucleus	Number of bands	Energy range (keV)	$\langle E_{\text{expt}} - E_{\text{calc}} \rangle$ (keV)	$\langle A_{\text{expt}} - A_{\text{calc}} \rangle$ Absolute difference (% difference)	E_{GM} (expt/calc) ^a (keV)
²³⁸ Np	9	0-345	29	0.18 keV (3.0%)	1.18,0.87,0.88
²⁴⁴ Am	16	0-680	21	0.5 keV (8.4%)	1.06,1.23
²⁵⁰ Bk	14	0-570	25	0.26 keV (4.8%)	0.95,1.41,1.14,1.27
¹⁶⁰ Tb	8	0-380	51	0.8 keV (8.2%)	1.03,1.07,1.13
¹⁶⁶ Ho	10	0-560	54	0.9 keV (8.8%)	0.80,1.08,1.31
¹⁷⁰ Tm	5	0-450	80	0.6 keV (5.2%)	2.04,0.98
¹⁷⁶ Lu	12	0-840	59	1.6 keV (10.8%)	1.14,1.30
¹⁸² Ta	7	0-270	31	0.5 keV (3.9%)	0.94,0.97,1.14

^a Ratios are listed for pairs with experimentally determined Gallagher-Moszkowski splittings.

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Structural and Electronic Properties of Platinum Oxides

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We found that platinum oxides can be separated into two distinct groups based on the valence state and coordination of platinum. One group contains platinum in planar four-fold coordination and a nonintegral oxidation state, resulting in strong metal-metal interactions and high conductivity. The other group contains tetravalent platinum in octahedral coordination, and the compounds have much lower values of electrical conductivity. Platinum oxides are an excellent example of a class of compounds in which electronic properties can be understood in terms of structural and crystal-chemical considerations.

Elucidation of the relationship between the structure and the physical and chemical properties of crystalline materials is fundamental to an understanding of the nature of solids. The identification of structural features responsible for certain characteristics can be useful in predicting the properties of an unknown or poorly understood compound. Analysis of this type is also useful for designing new materials that possess a desired set of physical or chemical properties.

The synthesis and characterization of binary and ternary platinum oxides is a field in which considerable progress has been made in the past 15 years. The interest in this area has been spurred by several factors. First, while platinum

is commonly assumed to be inert in the presence of oxygen at high temperatures, experience has shown that this is not always true. For example, platinum reacts with other metals to form oxides during experiments using platinum containers or thermocouples. An understanding of platinum oxides is thus important. Second, platinum oxides are of interest because of their catalytic and electrochemical properties.^{1,2} Finally, the discovery of high conductivity in some ternary platinum oxides containing infinite chains of platinum atoms³⁻⁶ has led to investigations of the relationships between their crystalline structures, chemistry, and physical properties.^{7,8}

I reviewed the structure, crystal chemistry, and electronic properties of all known platinum

Table 45. Electrical conductivity of selected platinum oxides.

	Pt-Pt distance (Å)	Dimensionality	Conduction type	Electrical conductivity at 298 K ($\Omega \cdot \text{cm}$) ⁻¹	Activation energy for semiconductor (eV)	Reference
Platinum in planar coordination						
$\text{Na}_x\text{Pt}_3\text{O}_4$ ($x = 1.0$)	2.84	3D	Metallic	1×10^4		5
$\text{Mn}_{1+x}\text{Pt}_{3-x}\text{O}_6$ ($x \approx 0.2$)	3.16	1D	Semicond.	1×10^2 ($\parallel c$)	0.03	6, 8
Platinum in octahedral coordination						
$\beta\text{-PtO}_2$	3.14	1D	Semicond.	2×10^{-1}	0.2	6
Ca_4PtO_6	4.78	Isolated	Insulator	10^{-9}		9

oxides, and demonstrated that some physical and chemical characteristics of these materials can be understood best in terms of fundamental structural factors. I also found that platinum combines with oxygen and a wide variety of alkali, alkaline-earth, transition, and rare-earth metals at elevated temperatures and pressures.

Table 45 shows a representative cross section of structurally characterized platinum oxides for which electrical conductivity data are available. The table shows that platinum oxides span the range from metal to insulator to low-dimensional conductor. In general, platinum oxides can be divided into two groups, based on their electrical conductivity: those in which there is direct electronic interaction between platinum atoms on adjacent sites in a structure, and those with no significant degree of interaction. This distinction in the degree of direct metal-metal interaction can be directly related to a dichotomy in the possible valence states and coordination of platinum in oxide structures.

Highly conducting platinum oxides such as $\text{Na}_x\text{Pt}_3\text{O}_4$ and $\text{Mn}_{1+x}\text{Pt}_{3-x}\text{O}_6$ contain partially oxidized platinum (Pt^{n+} , where $2 < n < 4$) in four-fold planar coordination. The strong metal-metal interactions result from the presence of linear arrays of unbridged platinum ions forming directly interacting systems. The structural feature that allows metallic bonding is short platinum-platinum distances perpendicular to the plane of the platinum site. That feature permits the free d electrons outside the plane of the ligands to interact with neighboring platinum atoms. The structural configuration is termed columnar stacking.¹⁰ Figure 113 shows an idealized example. The result is the formation of a d_{z^2} band containing collective delocalized electrons. Figure 114a shows a schematic representation in which the formation of the band structure for Pt^{2+} (d^8) is in a perfect square-planar site (D_{4h} symmetry).

The existence of the d_{z^2} band in columnar-stacked platinum oxides can also be related to the chemical nonstoichiometry often found in the highly conducting materials. In platinum oxides such as $\text{Na}_x\text{Pt}_3\text{O}_4$ and $\text{Mn}_{1+x}\text{Pt}_{3-x}\text{O}_6$, the valence state of platinum is nonintegral because of the partial oxidation of platinum atoms in the metal chain. This partial oxidation ($\text{Pt}^{2+} \rightarrow \text{Pt}^{n+}$, $2 < n < 4$) removes electrons from the d_{z^2} band (Fig. 114b), and leaves it only partially filled, promoting metallic conductivity. In $\text{Na}_x\text{Pt}_3\text{O}_4$, the sodium content x can vary from 0 to 1, and the concentration of conduction electrons varies

with composition to produce the proper charge compensation. In $\text{Mn}_{1+x}\text{Pt}_{3-x}\text{O}_6$, deviations from stoichiometry are needed to promote platinum-platinum interactions, which stabilize its structure.

$\text{Na}_1\text{Pt}_3\text{O}_4$ is the prototypical metallic platinum oxide whose conductivity increases with decreasing temperature. It contains three orthogonal sets of columnar stacks linear platinum chains, one stack parallel to each crystallographic axis of its cubic unit cell. In $\text{Mn}_{1+x}\text{Pt}_{3-x}\text{O}_6$, however, columnar stacking occurs parallel to the c axis only, so high conductivity is limited to only one dimension. One-dimensional conductivity is inherently unstable, particularly at low temperatures,¹¹ and this is demonstrated in one-dimensional platinum oxide conductors, in which conductivity decreases with decreasing temperature.

Platinum oxides with low electrical conductivity contain fully oxidized tetravalent platinum in octahedral coordination polyhedra. Structures containing octahedrally coordinated Pt^{4+} can be categorized by the type of polyhedral linkage employed, e.g., face-shared linkage, edge-shared linkage, corner-shared linkage, or structures with isolated octahedra. Naturally, a compound such as Ca_4PtO_6 , which contains platinum in isolated octahedral sites, has no platinum-platinum interactions and is an insulator. However, $\beta\text{-PtO}_2$ contains chains of edge-shared PtO_6 octahedra extending along the

Figure 113. Idealized representation of columnar stacking of planar MO_4 groups. Metal-metal interaction occurs perpendicular to the plane of the site along the chains of metal atoms.

c axis of the orthorhombic unit cell. In spite of platinum-platinum distances of 3.14 Å, comparable to $\text{Mn}_{1+x}\text{Pt}_{3-x}\text{O}_6$, the electrical conductivity of $\beta\text{-PtO}_2$ is lower by three orders of magnitude. That is because the electronic configuration of Pt^{4+} (d^6) in octahedral coordination is low-spin, with six d electrons just filling the band formed by cationic t_{2g} orbitals of the platinum atom chains. The low conductivity of $\beta\text{-PtO}_2$ confirms that adjacent Pt^{4+} cations in octahedral coordination will not promote metallic conductivity, since the Fermi level occurs in the band gap between bands of t_{2g} symmetry and those derived from e_g wave functions in an octahedral environment.^{12 13}

Platinum oxides, as a group, demonstrate that metal-metal distances are not as important as other crystal-chemical and structural considerations, e.g., polyhedral linkage and dimensionality, and platinum valence and degree of partial oxidation, in determining the strength and temperature dependence of electronic interactions and electrical conductivity. Experiments are in progress to measure the behavior of highly conducting one-dimensional platinum oxides at elevated pressures and temperatures.

This research was supported by the LLNL Nuclear Test Program.

Figure 114. (a) Schematic representation of the atomic energy levels for d^8 metals (e.g., Pt^{2+}) in square-planar coordination (D_{4h} symmetry), and the formation of a band structure for a metal atom chain. (b) Effect of partial oxidation of the d_{z^2} band, which leaves a partially filled band and promotes metallic conductivity.

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Metastable Phase Separation in Au-Fe Alloys

C. E. Violet¹ and R. J. Borg

We found that 4.2-K ^{57}Fe Mössbauer spectra of Au-Fe alloys with an iron concentration between 10.5 and 33 at.% are well described by two superposed six-line spectra. Presumably, these derive from two different chemical environments, which are identified with the $\langle 420 \rangle$ platelet and solid-solution phases.

Theories purporting to explain the magnetic behavior of spin glasses are based on the assumption that the alloy is a single-phase solid solution.² It is generally presumed that such alloys can be prepared by a fast quench from high temperature, and phase separation, when present, proceeds according to the familiar pattern of nucleation and growth. However, short-range order in Au-Fe alloys was pointed out by Crane *et al.*,³ and its nature elucidated by the diffuse-x-ray studies of Dartyge *et al.*⁴ in alloys with iron concentrations between 14 and

19 at.%. They found a two-phase system in which iron was concentrated in platelets oriented in the $\langle 420 \rangle$ planes, about two planes thick and extending 30 Å parallel to their plane.

In this report, we: (1) present Mössbauer spectroscopy data that agree with and augment those x-ray results, (2) note that such platelets are a common pre-precipitation phenomenon in certain fcc alloys, including Au-Fe,⁵ and (3) emphasize that because of the extreme rapidity of pre-precipitation, it is virtually impossible to prevent platelet formation (except

Table 46. Parameters of the 4.2-K spectra, obtained by least-squares fitting.^a

$\bar{X}$ (at.%)	A_h/A_l	Isomer shift (mm/s)		Quadrupole shift (mm/s)		Hyperfine magnetic field (kOe)	
		v_l	v_h	ϵ_l	ϵ_h	H_l	H_h
10.5	0.368	0.58	0.47	0.069	-0.148	269	316
16.8	0.529	0.58	0.51	0.045	-0.196	272	324
18.5	0.757	0.58	0.51	0.084	-0.096	276	324
20	1.079	0.59	0.50	0.097	-0.099	281	328
25	1.747	0.57	0.44	0.117	-0.089	298	338
33	0.327	0.54	-0.03	0.077	0.037	319	334

^a The symbols in the table are defined as follows:

$\bar{X}$ is bulk iron concentration,

A is total resonance absorption of lines 1 through 6,

v is center shift relative to a palladium source at room temperature,

ϵ is quadrupole shift of the ^{57}Fe magnetic levels,

H is magnetic hyperfine field,

l is low (magnetic hyperfine) field phase,

h is high (magnetic hyperfine) field phase.

Spectra calculated with these parameters are shown in Fig. 115.

in the very dilute alloys) by quenching from high temperature. Consequently, it appears that virtually all experimental work has been done on samples that were not even approximately random-solid solutions. Yet interpretations of those experiments have been based on concepts such as the infinite cluster and finite clusters, stemming from the random-alloy model. The diffuse-x-ray studies and our Mössbauer results strongly suggest that for nondilute Au-Fe, and perhaps for similar fcc alloys, the random-alloy model is inappropriate.

Alloys containing iron in concentrations of 10.5, 16.8, 18.5, 20, 25, and 33 at.% were prepared as described.⁶ All were rolled to a thickness of 5 μm or less, annealed at 900°C for 10 h, and quenched in water. Figure 115 shows the 4.2-K Mössbauer spectra. They are well described by two superposed six-line ⁵⁷Fe magnetic hyperfine spectra. Table 46 gives the parameters, which were determined by least-squares fitting.

We propose that the two distinct six-line spectra must derive from iron in two different compositional phases. This interpretation differs from that of Ridout⁷ and Window,⁸ who assumed a random-solid solution. The values of v_l , ϵ_l , and H_l are continuations of their values in dilute Au-Fe.⁶ Therefore, the low-field phase in these alloys is the solid-solution phase described in Ref. 6. Values of v_h are distinctly different from those of v_l and are shifted toward alpha-iron; H_h is significantly greater than H_l . This suggests that the iron is more concentrated in the high-field phase. The quadrupole shift for the high-field phase, ϵ_h , differs markedly from ϵ_l . About 25% of the iron in the 33-at.% alloy is in the alpha-iron phase, which is evidenced by its hyperfine-field magnitude and temperature dependence. This is consistent with the equilibrium phase diagram.⁹ The relative amounts of the two phases in the other alloys do not correspond to the phase diagram and are therefore metastable. We have elected to call the precipitated alpha-iron the high-field phase for the 33-at.% alloy. However, the possibility of a small quantity of platelets being present but not detectable in the Mössbauer spectra is not ruled out.

Eq. (3) in Ref. 6 gives the iron concentration of the low-field and high-field phases, X_l and X_h . These are given in Table 47 and plotted in Fig. 116. The fraction of iron atoms in the high-

Figure 115. Mössbauer resonance absorption spectra at 4.2 K for Au-Fe alloys with iron concentrations of 10.5, 16.8, 18.5, 20, 25, and 33 at.%.

Table 47. Iron concentrations and high-field-phase iron fractions.

$\bar{X}$ (at.%)	X_l (at.%)	X_h (at.%)	$\bar{X}_c$ (at.%)	X_c/X	f_{Fe} (%)
10.5	8.4 ± 1.4	24.8 ± 3.8	10.2 ± 1.5	0.97 ± 0.14	27 ± 1
16.8	9.1 ± 1.5	30.4 ± 4.8	12.7 ± 1.9	0.76 ± 0.11	35 ± 3
18.5	10.0 ± 1.6	30.4 ± 4.8	15.7 ± 2.1	0.85 ± 0.11	43 ± 2
20	11.4 ± 1.8	34.4 ± 5.7	17.5 ± 2.2	0.88 ± 0.11	52 ± 2
25	16.6 ± 2.6	64.1 ± 11.2	31.4 ± 3.8	1.26 ± 0.15	64 ± 2
33	26.7 ± 4.1	100.0 ^a	32.6 ± 6.2	0.99 ± 0.19	25 ± 2

^a Alpha-iron. Figure 116. Iron concentrations of the low-field phase X_l and high-field phase X_h vs bulk iron concentration $\bar{X}$. Curves for X_l vs $\bar{X}^{3/2}$ and X_h vs $\bar{X}^2$ are also shown.

field phase is given by $f_{Fe} = A_h/(A_l + A_h)$, which is plotted in Fig. 117. As shown in Fig. 116, X_h and X_l vary approximately as $\bar{X}^2$ and $\bar{X}^{3/2}$, respectively. In Fig. 117, f_{Fe} varies approximately as $\bar{X}^{3/2}$. It may be significant that

Figure 117. Fraction of iron atoms in the high-field phase, f_{Fe} vs bulk iron concentration $\bar{X}$. The curve for f_{Fe} vs $\bar{X}^{3/2}$ is also shown.

X_h and X_l increase suddenly at about 17 at. % iron. This is consistent with our earlier report of a critical composition at about 17 at. % iron.¹⁰⁻¹² The possibility of a spinodal decomposition to a metastable ordered phase, viz Au_5Fe , is a distinct but as yet undemonstrated possibility.¹³

In light of the results of Dartyge *et al.*,⁴ we identify the high-field phase with the platelets and the low-field phase with the solid-solution matrix. Dartyge *et al.* considered the platelets in their samples to be predominantly iron atoms, but our results indicate that they contain about 30 at. % iron and the matrix contains about 10 at. % iron. These Mössbauer results modify but do not negate the x-ray results.¹³

Pre-precipitation on specific crystallographic planes is a rather common phenomenon and has been found in such diverse systems as Ni-Al,

Al-Ti, and Ni-Cu-Al;¹⁴ and Cu-Ti;¹⁵ as well as many others. There is a variety of phase-separation-ordering transformation modes whenever an ordering spinodal is situated just below the phase-separation spinodal.¹⁶ Such transformations produce two distinct compositional phases rather than the continuous variation postulated by earlier theories of spinodal decomposition.¹⁷ Because the phase separation occurs on such a fine scale, with each typically but a few atomic layers thick, such reactions are extremely rapid and account for the failure of normal quenching procedures to maintain the single-phase structure.

The model we propose may account for much of the unusual magnetic behavior of Au-Fe alloys containing more than about 10 at.% iron.

These unusual forms of behavior include the double maxima in the a.c. susceptibility² and the double magnetic transition.^{18,19} In our model, the high-field phase in Au-Fe bears a definite crystallographic relationship to the matrix, and theories of magnetic behavior based on random nucleation and growth of iron-enriched clusters are not applicable. Finally, our model makes compatible the electron diffraction²⁰ and Mössbauer²¹ results obtained from Cu(Ni-Fe), which is known to undergo spinodal decomposition. It may also apply to Cu-Mn spin glasses, in which short-range order analogous to that of Au-Fe is observed.²²

This work was supported by the LLNL Office of Equal Opportunity and the LLNL Nuclear Chemistry Research Program.

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