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INMM

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Editorials

Dr. Curtis G. Chezem

MBA-99

'EXCELLENT SUPPORT'

This is the first issue of the second year of the Journal of the INMM. We should wish ourselves happy birthday. We can summarize the year nicely. We did what we said we would and the credit goes to our managing editor, Tom Gerdis. Tom defined what we could do and then set about to do it with excellent support from the Executive Committee. Congratulations to all. The one bothersome thing to me is the lack of suggestions for improvement from the membership. Perhaps this is an additional salute to Tom's know-how as a professional journalist and student of administration.

The most significant development during the last year has to be the upheaval in the Atomic Energy Commission. Out of this appears to be developing a progressive dialog between INMM Chairman Harley Toy and Jim Powers, Chief of the Materials Protection Standards Branch of Regulatory. AEC/ INMM discussions of the role of the INMM certification program are absolutely necessary.

It would be shortsighted of the AEC or their successors to fail to recognize and exploit the self-improvement effort of the INMM. Our Institute leaders would be equally derelict not to force the issue. As we see it now, both parties are moving into the proper posture of mutual respect and recognition. This leaves your editor without a "soap box" to generate a cause celebre . . . at least temporarily.

What about manpower availability in Nuclear Materials Control? To the best of my knowledge, Nuclear Materials Control does not have a textbook to the extent that having studied and worked examples in the book a student should be able to pass a certification exam. Manny Kanter at Argonne and Walt Meyer at the University of Missouri have assembled what should pass as a fine self-study course. It was designed as a three-semester-hour self-paced senior or graduate level course. I hope these two devotees can whip it into a neat pedagogical package. We need it, now!

'MORE THAN . . . WE ARE . . .'

A letter from Howard Freitag published elsewhere in this issue, has started the thinking process moving again. It seems presumptuous for anyone less than an "old-timer" to be charged, at least honorarily, with the editorial communications function, but we try. Howard's letter has contributed a bit of "folk-lore" to the permanent record of the Institute. We had hoped that by introducing an "Out of Context" column we would stimulate memories of the long, yet short, history of the group. We would welcome some folksy vignettes from our readers . . . little tales told which would help record the heritage of the Institute.

This editor believes that we are more than ledgers, counters, statistics, accounts, manometers, scales, regulations, and so on . . . we are human beings bringing a heritage of accuracy, responsibility

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THE CHAIRMAN SPEAKS

Harley L. Toy

UPDATE—SPRING '73

It hardly seems possible but in a matter of weeks we will be heading West for San Diego to open our 14th Annual Meeting. It seems we just left Boston. I am sure your Program Committee would agree as they are in the process of putting together the final pieces for this year's meeting. Armand Soucy our Vice Chairman in charge of the Annual Meeting along with his Program Chairman, Roy Cardwell, have been hard at it since leaving Boston.

The Program Committee is definitely not a one or two man operation. Armand and Roy are assisted by the recognized talents of Fred Forscher, Doug George, and Shelly Kops. With a team like that you can once again count on an exceptional meeting. This year's program will consist of some twenty-five papers and a government-industry panel discussion. The panel will take an in-depth look at the presently proposed rule changes to 10 CFR Parts 50, 70, and 73. Recognizing the fact that by meeting time certain of the proposed rule changes may be effective, the Program Committee feels there is much to be gained by such a panel discussion, especially in the area of implementation of the changes.

I am happy to report that Mr. Les Rogers, Director of the Directorate of Regulatory Standards has accepted our invitation to deliver the luncheon address on June 21. I am sure Mr. Rogers will provide us direct insight into the role and objectives of the Standards Directorate and discuss specific standards areas of mutual concern.

Preregistration forms with the program agenda along with other details of the meeting will be mailed in early May.

I had the opportunity to attend the recent AIF Workshop on Physical Protection sponsored by the Forum's Committee on Nuclear Materials Safeguards. Some fifteen Institute members were on hand to participate in the discussions aimed at the proposed AEC rule changes to 10 CFR Parts 50, 70, and 73.

Dr. John E. VanHoomissen, a member of your Executive Committee, provided one of industry's views in a formal presentation **Industrial Guidelines to Materials Protection.** John will be actively participating in our Government-Industry Panel at San Diego and will share his views on the "up-in-the-air" situation in the proposed physical protection regulations. The reference to "up-in-the-air" situation stems from my observations during the workshop sessions. Throughout the formal papers, questions, and discussions regarding the proposed physical protection rule changes there appeared to be a common-thread of incompatibility and reservation. At this stage the proposed rule changes to Part 73 just don't "hang-together" which leads me to believe that the proposed changes as they now stand will not fly.

At the moment there are a multitude of questions unresolved in the area of physical protection requirements for site installations and protection requirements for certain SNM in transit. The Commission has intimated that their objective in the area of physical protection is that of stipulating performance criteria rather than stringent, hard-fast regulations. At the workshop the Commission representatives echoed a pressing need for comments and suggestions on the proposed rules.

Along this line I would direct your attention to the recent Atomic Energy Clearing House Publication, dated April 2, 1973, Volume 19, in which Dr. Russ Wischow, President of Nuclear Audit and Testing Company, and Mr. Lawrence D. Low, former Director of Division of Compliance, commented quite comprehensively on the proposed physical protection regulations. I am sure you'll find the comments, suggestions, and proposals set forth by Wischow and Low a "down-the-line" approach with respect to the extent of the physical protection question and the Commission's proposed rule changes. Dr. Wischow's comments are also directed at 10 CFR, **Revised Material Control and Accounting Requirements for Special Nuclear Material.**

Here again some straight forward questions are brought to light relative to proposed inventory requirements and the limit of error of MUF on material balances.

Some final notes on recent Institute activities:

(1) Discussions are continuing with Mr. James Powers, Chief of Materials Protection Standards Branch of the Directorate of Regulatory Standards relative to our Certification Program. We are exploring areas whereby our Certification Program could be compatible for inclusion in the **USAEC Regulatory Guide Series.**

(2) Mr. Bernie Gessiness, former Institute Chairman, and currently heading up an Ad Hoc Committee on the Institute "Today and In the Future" recently submitted a progress report on the committee's actions to date. Bernie's report was a comprehensive, in-depth appraisal of our current activities and projected recommendations on the future course of the Institute. We shall ask Bernie to prepare a report on the Committee's recommendations for publication in the next issue of the Journal.

(3) The Nominating Committee for 1973 has presented a slate of nominees for the 1973 election of Officers and Executive Committee Members. Ballots will be mailed to the membership by April 15.

(4) Bob Delnay, Chairman of N15, continues to do an outstanding job as evidenced by recently ANSI-Approved standards. Many thanks to the numerous Institute members actively engaged in N15 work. To appreciate the efforts of these individuals one must understand that all the time and labor involved is pure gratis. The many after-hours efforts of the individuals involved in N15 have been too long overlooked. Let us hope the Industry and the Commission are aware of their contributions.

I am looking forward to our San Diego gathering.—Harley L. Toy, Chairman.

LETTERS TO THE EDITOR

Editor:

The exchange of letters between Norton and Lovett published in the January, 1973 issue addressed the question of what effect monthly inventories has on the uncertainty of an annual MUF. It should be clear that the uncertainty of a total MUF calculated over a given time period is in no way affected by the number of inventories which may have been taken during that time period. Rather, it depends only on the beginning and ending inventories for the period in question, plus removals and additions. I do not believe that one must utilize "Complex statistical techniques" to grasp this simple fact.

John L. Jaech
Staff Consultant
Exxon Nuclear
Richland, Wash.

Editor:

Please refer to NMM Vol. 1, No. 4 January 1973, page 7 OUT OF CONTEXT, the eighth item: "The INMM manual

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defines a Nuclear Materials Manager as a 'person . . . program standards . . . control. He shall posses the . . . to create or implement a Nuclear Materials control system'."—Russ Weber and Shelley Kops, 1962.

As I am the author of the definition of a Nuclear Materials Manager, I must correct the item as reported by you. I was an officer of INMM at the time my definition was selected. (I don't remember if it was while I was a committee chairman or a member of the Executive Committee.) The membership was asked to submit their definitions. They were received and reviewed by the INMM officers at a quarterly meeting, which I attended. After reviewing the definition submissions, Ralph Lumb made a motion which was seconded and passed that my submission be selected as the official definition for the INMM manual. I am sure Dr. Lumb can support this statement.

Howard R. Freitag
Kennewick, Wash.

Editor's Note: Our thanks to an old friend for clarifying the record. Since Russ and Shelley were quoting the manual, kudos to the author of the definition are implied and, if not obvious, are herewith tendered.—C.G.C.

NEWS

DOORWAY MONITORS FOR FISSILE MATERIALS

SAN DIEGO— Rad Tech has developed a new safeguards tool—a doorway monitor to detect diversion of fissile materials. The monitor will detect approximately one gram or more of ^{239}Pu within a count time of approximately one second. This allows for monitoring personnel as they proceed past the monitor at a normal walking speed.

The Rad Tech Doorway Monitor (P.O. Box 608, San Diego, CA 92138) basically consists of two 6 to 8 feet long scintillation detectors, mounted one on either side of the doorway, and a unique electronic signal processing system.

The length of the detectors provides a uniform response along the height of the doorway, and eliminates "dead spots" that are commonly found in systems utilizing small detector arrays. The system is specifically designed for high stability. Production of the new monitors has started and the first deliveries are scheduled for late May. Requests for quotations on this new item are now being accepted.

NDA PLUTONIUM ASSAY

Commercial production of the Brookhaven National Laboratory's Neutron Well Coincidence Counter has been undertaken by National Nuclear Corp., Redwood City, Calif. NNC recently built and delivered one of these plutonium assay instruments to Westinghouse-Hanford for use in the FFTF program. Before being turned over to commercial production, several of these instruments were built by BNL for use in the AEC Safeguards program.

The instrument assays plutonium by measuring the spontaneous fission neutrons emitted from Pu^{240}. Fission neutrons and (,n) neutrons are distinguished by counting only neutrons in coincidence. The unknown sample is inserted in the counter well and counted for a preset time, usually 500 seconds. Comparison with standards provides Pu^{240} content. With known isotopic ratios and appropriate corrections, the amount of fissionable plutonium can be determined. Varying size samples can be accommodated.

Calibration tests performed at General Electric-Vallecitos and at Westinghouse-Hanford showed linearity of instrument response to amount of Pu^{240} over a three decade range (10 mg-10 g). Instrument precision of 2 per cent at 1 g Pu was measured and the instrument could detect amounts of Pu^{240} as small as 1 mg.

Additional information can be obtained from National Nuclear Corporation, telephone: (415) 364-2880.

SHIPPING CASK

ROCKVILLE, Md.—Licensing by the United States Atomic Energy Commission of the nuclear power industry's first cask for shipment of spent fuel

(Continued on Next Page)

Robert L. Delnay

N15 STANDARD STATUS

by

R. L. Delnay
Chairman

The number of proposed ANSI standards being prepared by N15 increased by six. This increase is the result of an AEC request to prepare the following standards:

N15.16 - "Standard for the Application of Statistics to Licensee Data." This standard will be prepared in Subcommittee N15-3 which is chaired by John Jaech.

N15.23 - "Standard for the Use of Fuel Rod Scanners for Measurement of Nuclear Material Content of Fuel Rods." Lynn Hurst is forming a new Subcommittee to write this standard.

N15.24 - "Standard for the Recordkeeping and Reporting of Licensee Inventory Data." The responsibility for this standard was assigned to Russ Weber, Chairman of N15-4 Subcommittee.

N15.25 - "Standard for Measuring Material in Process Equipment." Subcommittee N15-6, with Doug George as chairman, will produce this standard.

N15.26 - "Standard for Material Protection Considerations in Plutonium Scrap Recovery;" and,

N15.27 - "Standard for Material Protection Considerations in High Enriched Uranium Scrap Recovery." Both of these standards have been assigned to John VanHoomissen. He is busy putting together his subcommittee. I am sure he can use any who wish to volunteer for his subcommittee.

Armand R. Soucy is the new chairman of Task Group N15-1.4. This group is updating the standard N15.8, "Nuclear Materials Control System for Nuclear Power Reactors." Mr. Soucy has expanded the original task group in order to include material not covered in the first version of the standard.

Gene Miles has just recently become the chairman of Task Group N15-1.3. This group will be resolving the comments received during letter ballot on proposed standard N15.9, "Nuclear Material Control System in Fuel Fabrication Plants."

The American National Standards Institute has recently published standards N15.7-1972 and N15.10-1972. N15.7-1972 is the standard that covers the analytical procedures for accountability of uranium hexafluoride. N15.10-1972 is the scrap classification guide for plutonium. Both standards may be purchased from the American National Standards Institute, Inc.

assemblies from second generation reactors was announced by Nuclear Fuel Services, a pioneering leader in nuclear fuel manufacturing, reprocessing and transportation.

Designated the NFS-4 by Nuclear Fuel Services, the new cask is capable of safely transporting a broad range of second-generation Light Water Reactor (LWR) fuel assemblies. Capacity of the new cask is two Boiling Water Reactor (BWR) assemblies or one Pressurized Water Reactor (PWR) fuel assembly.

The 50,000 pound NFS-4 cask meets all AEC and U. S. Department of Transportation regulations relating to Fissile Class III shipment of large quantities of radioactive material. Principal means of transportation will be by specially-designed truck-trailer under sole use assignment. Rail or other modes of transportation also may be utilized.

AEC APPROVES FUEL ASSEMBLY INSERTIONS

ROCKVILLE, Md. — Insertion of four Nuclear Fuel Services, Inc., mixed oxide demonstration fuel assemblies into the Big Rock Point Reactor of Consumers Power Company in Michigan during the Spring of 1973 has been approved by the United States Atomic Energy Commission, according to an announcement here by Nuclear Fuel Services, Inc.

Loading of the assemblies marks another industry achievement for NFS since this will be the first time, commercially, that recycled uranium and plutonium have been returned, as mixed oxide fuel, to the same domestic reactor from which the spent fuel was discharged.

NEW MEMBERS OF I.N.M.M.

The following individuals have been accepted into INMM membership as of April 13, 1973. To each, the staff of the Journal extends congratulations. New members not mentioned in this issue of the Journal will be published in the summer issue to be mailed in late July or early August.

Richard N. Chanda, Senior Research Chemist, Dow Chemical, Golden, Colo.; Leonard F. Dow, Boston Edison, Boston, Mass.; Joe Dykstra Jr., Superintendent, Chemicals Operation Department, Union Carbide, Oak Ridge, Tenn.; A. T. Freeman, Nuclear Division, Union Carbide, Paducah, Ky.; and Robert M. Keller, 242 North Purdue Avenue, Oak Ridge, Tenn.

Kosta S. Kotti, 811 Jackson Avenue West, North Augusta, S. C.; Leonard Lanni, 1443 Via Loma, Walnut Creek, Calif.; James E. Morcom, Staff Assistant, Babcock & Wilcox Research Center, Lynchburg, Va.; William E. Pappanastos, Babcock & Wilcox, Lynchburg, Va. and Albert D. Parent, 507 Shady Lane, Cayce, S. C.

Dr. T. Douglas Reilly, Staff Member, Los Alamos (N.M.) Scientific Laboratory; Thomas J. Schmierer, P. O. Box 11116, Albuquerque, N. M.; William J. Shelley, Director, Regulation and Control, Kerr-McGee Nuclear Division, Oklahoma City, Okla.; Norman H. Weissert, Analytical Supervisor, W. R. Grace & Company, Clarksville, Md.; and Francis J. Wieczorek, Specialist, Nuclear Materials Management, General Electric, San Jose, Calif.

EDITORIALS

(Continued from Page 1)

and integrity from many disciplines to the still embryonic nuclear fuel cycle industry.

Yes, people, and as I look out across the city tonight from our 27th floor at the myriad electric "candles" that light our darkness, I'm reminded once again what the modern fuel cycle is all about. Micah said it:

". . . they shall beat their swords into plowshares,
and their spears into pruninghooks."

We're all proudly a part of that. Let's not forget where we've been and where we are to kow where we're going.

NEW SAFEGUARDS COURSE—Three Kansas State University nuclear engineering faculty members recently completed a programmed instructional full-semester course using individually prescribed instruction (IPI) techniques. Dr. Walter Meyer (l.), formerly of K-State and now head of nuclear engineering at the University of Missouri, Columbia, headed the project. Dr. Manny Kanter (r.) of Argonne (Ill.) National Laboratory was the project monitor.

INMM UPGRADING OF CERTIFICATION COMMITTEE, meeting during a few spare moments at the recent Atomic Industrial Forum Protection of Special Nuclear Materials Workshop in Key Largo, Fla (l. to r.)—John E. VanHoomissen, Harley L. Toy, James W. Lee and Russell Wischow.

ADDRESS CHANGES OF INMM MEMBERS

The following are new addresses for members of the Institute of Nuclear Materials Management:

Thomas B. Bowie, Manager, Nuclear Materials and Security, Combustion Engineering, Inc., 1000 Prospect Hill Rd., Windsor CT 06095; Joe Dykstra Jr., 624 Pennsylvania Ave., Oak Ridge TN 37830; C. Gordon Hough, 4606 S. 342nd, Auburn WA 98002; James E. Lovett, c/o International Atomic Energy Agency, P. O. Box 645, Kaerntnerring 11, A-1011 Vienna, Austria; and M. N. Wolfe, 10327 Tumblewood Dr., Sun City AZ 85351.

OUT OF CONTEXT

Items selected at random (more or less) from past proceedings of the INMM.

"Where do we go from here?" J. Vinciguerra, Washington, 1967.

"All of us who are responsible for safeguarding special nuclear material, both in Government and private industry, must avoid using material balance accounting information as a dog uses a lamppost — for relief rather than enlightment." L.D.Y. Ong, Palm Beach Shores, 1971.

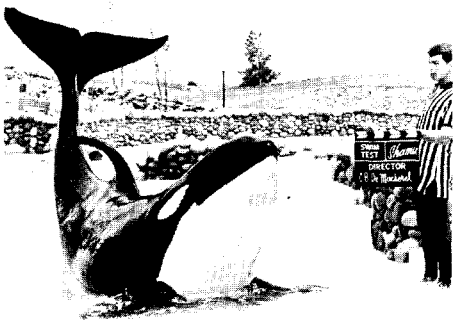
"Some of the major problems expectedly encountered in this study have included site selection, transportation of hot fuel, waste disposal, AEC licensing and insurance, and regulatory aspects of each." E. R. Johnson, Columbus, 1960.

". . . Las Vegas must be off limits to electric utility employees." A. R. Soucy, Gatlinburg, 1970.

"There are, however, certain problems with the licensee-station situation that would bear serious attention." Ed North, Columbus, 1960.

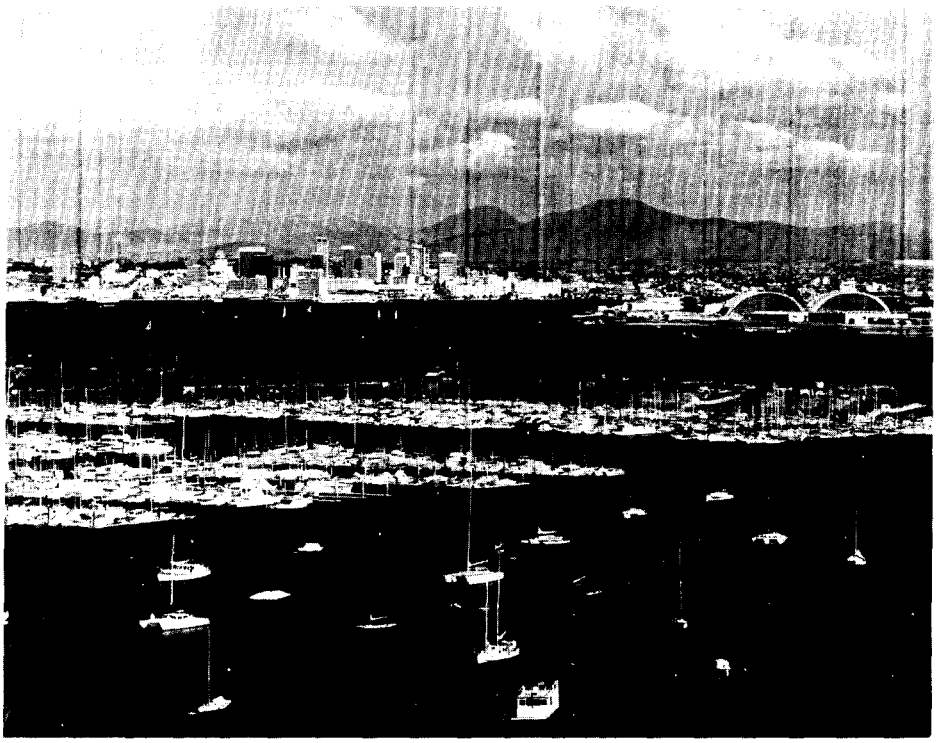
"I'm sure none of you will disagree with me." Charles Keller, Cincinnati, 1965.

"The U. S. safeguards staff has maintained close relations. . ." John Downing, Washington, 1967.



SCREEN TEST — Shamu, a 17-foot-long killer whale does a "slide out" to climax his screen test for a new contract in the "Shamu goes Hollywood" show at San Diego's famous Sea World marine show park. Among other things, the popular whale gives his trainer a thundering bareback ride and kisses a pretty girl in each of his shows.

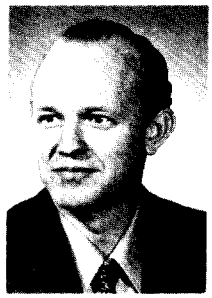
SAN DIEGO CONVENTION SITES



SAN DIEGO, California's historic birthplace and now the second largest city in the West, is nestled around San Diego Bay with the Laguna Mountains on the east and the Pacific Ocean on the west. In the foreground is the tip of Shelter Island, one of two man-made islands in San Diego Bay that house restaurants, hotels, motels, and boat marinas.

SAN DIEGO'S BALBOA PARK, infinite in variety and almost unsurpassable in beauty, covers 1400 acres within the heart of the city. The park is the site of many San Diego cultural and recreational activities —Shakespearean and contemporary theatre, concerts, Broadway musicals, art galleries, museums, golf, archery, tennis, and the world famous San Diego Zoo. Shown here is the Botanical Garden and the colorful lily pond.

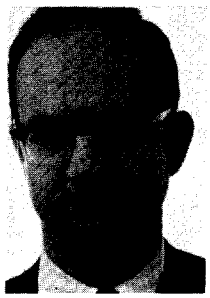
Forscher



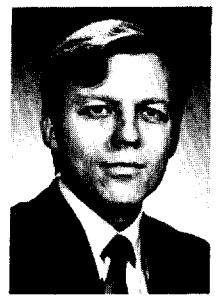
Christensen

Johnson

Minnick



Powers



Prezbindowski

AUTHORS FOR THIS ISSUE

Dean E. Christensen (M.A., Physics, Brigham Young University, 1962) is currently involved in the application of burnup relationships to chemical reprocessing plant data from spent nuclear fuels for possible safeguards use. His past experience includes conducting and directing both nondestructive and destructive burnup studies of nuclear fuels and the analysis of the resulting data.

Frederick Forscher (Ph.D., Applied Mechanics, Columbia Univ. 1953) He is currently Consultant in the fields of Nuclear Fuels, Standards, and Energy Management. He started his nuclear career at Bettis (1952-57) as Supervisor of Mechanical Metallurgy, was co-founder and V.P. of Operation for Numec from 1957 to 1967, and returned to Westinghouse as Manager of Advanced Fuel (1967-71). He is the author of some 30 papers in the fields of mechanics, materials engineering and social physics. He is also a Certified Nuclear Materials Manager.

E.R. Johnson is President of E.R. Johnson Associates, Inc. From 1957 to 1967, he held several positions with Nuclear Fuel Services, Inc., first as Technical Director (Erwin, Tennessee); Assistant General Manager (West Valley, New York) and finally as Vice President. From 1952 to 1957 he worked for the National Lead Company at the AEC Feed Material Production Center as Section Leader for the chemical development of recovery processes for uranium and other metals. Mr. Johnson holds a B.S. in chemistry from Bowling Green State University and has performed graduate work at Ohio State University and at Xavier University. He has authored numerous articles in the field of uranium recovery, spent fuel reprocessing, nuclear material control and safeguards and nuclear fuel cycle economics. He is a past Chairman of the Institute of Nuclear Materials Management and holds membership in the American Nuclear Society, the American Institute of Chemical Engineers and the American Chemical Society. He is Chairman of the ANSI Subcommittee N15 for the development of standard nuclear material control systems.

Lawrence E. Minnick is a vice president of Yankee Atomic Electric Company, Westboro, Mass. Minnick joined New England Electric System in 1948 and held various positions in the Steam Production Department until 1954. At that time he took a leave of absence and was assigned to Atomic Power Development Associates in Detroit, Mich. After three years in Detroit, Minnick returned to Boston and joined Yankee where he was made assistant vice president in 1963. He was named to his present position in 1966.

James A. Powers (Ph.D., Nuclear Chemistry, Purdue University), is Chief of the Material Protection Standards Branch, Directorate of Regulatory Standards, USAEC Regulation, a position he has held since May 1972. Dr. Powers has been employed by the AEC since early 1966. Prior to becoming associated with the nuclear material safeguards program, he worked for ten years in the isotope power program.

David L. Prezbindowski (Ph.D., Nuclear Engineering, Purdue University, 1967) has contributed to the understanding of the theoretical aspects of isotopic correlations at Battelle-Northwest. His responsibilities have included the performance of calculations of reactor isotopic inventory, criticality and shielding requirements.

ADVERTISING INDEX

MATERIALS AND PLANT PROTECTION STANDARDS

By James A. Powers, Chief
Materials Protection Standards Branch
Directorate of Regulatory Standards
U.S. Atomic Energy Commission
Washington, D.C.

Introduction

The Atomic Energy Commission has been encouraging the development of standards on an accelerated basis for some time. Both Commissioner James T. Ramey and AEC-Regulation Director L. Manning Muntzing emphasized the need for standards in remarks at the ANS sponsored Executive Conference on Nuclear Standards in May 1972.[1,2] Mr. Muntzing discussed the then-recent establishment of a Directorate of Regulatory Standards in order to, in his words, "put drive behind our standards work." He requested an intensified and better standards effort from industry. More recently, Commissioner Doub stated that standards development and implementation is "a joint responsibility of the industry and the AEC working within the framework of the national standards writing organizations."[3]

As part of the Directorate of Regulatory Standards, a staff was organized to develop materials and plant protection standards. This group has been working since May 1972 with the broad objective, in the interests of national security and public health and safety, of establishing balanced, graded regulations and regulatory guides for (1) physical security and accountability to protect against, deter, and detect theft or diversion of strategic quantities of special nuclear material and (2) physical security to protect against and deter industrial sabotage of plants containing, processing, or otherwise using such material.

The Need for Material and Plant Protection Standards

The need for material and plant protection standards is brought into focus by a brief look at the present and future status of the nuclear power industry. To date, U. S. utilities have ordered or announced plans for 177 commercial nuclear electric plants, 169 light water reactors and 8 high temperature graphite reactors. Of this number, 29 nuclear plants are now in operation, 51 are in various stages of construction, 74 are "on the drawing boards" and 23 have been announced as future additions to local capacities.[4]

Approximately 20 companies currently operate facilities in the nuclear fuel cycle other than for mining, milling, and power production, with about 80 separate nuclear operations at 30 different locations. Plans for the immediate future call for some 20 additional nuclear operations (see Table 1). Thus, within the next few years approximately 180 separate operations, including reactors, at more than 110 different locations will be handling or using large quantities of special nuclear material.

The quantities of material being processed and used in nuclear facilities and transported between them is measured in thousands of kilograms of plutonium and millions of kilograms of enriched uranium each year. Domestic annual production of fissile plutonium by recovery from power reactor fuels is estimated to at 3,400 kilograms per year by 1975 and 13,600 kilograms per year by 1980, with perhaps as much as half of this material entering the fuel cycle (the rest will be in storage). Current domestic plutonium recovery is about 400 kilograms per year, most of which is now being stored. Plutonium production in foreign free world countries is now about 3,500 kilograms per year, with an estimated increase to more than 16,000 kilograms per year by 1980.[5] In 1971, more than 1.5 million kilograms of enriched uranium was delivered to the domestic nuclear industry and about a half million kilograms was delivered to foreign users. Estimates of the growth of the nuclear industry indicate that the domestic requirements for special nuclear material each year will be of the order of 2.5 to 3 million kilograms of uranium plus plutonium by 1975 and more than 5 million kilograms by 1980.

With these quantities of material being processed and moving within the industry, an effective and credible program of material and plant protection is required to discharge the Commission's responsibility to protect the common defense and security as well as its responsibility for public health and safety. The extent to which nuclear materials should be protected against loss or diversion is continually under review within the Commission. At one extreme a system of intense physical security and accounting may be prescribed to guard against and promptly detect small losses of material. At the other extreme, a minimum level of accounting may be prescribed to provide a periodic detection mechanism for nuclear material losses or diversions. An effective and credible program of material protection sufficient to discharge the Commission's responsibilities lies between these extremes in a balanced system of physical protection and material accounting.

In addition to a balance with respect to physical security and material accounting, the system requirements also need to be graded commensurate with strategic significance and radiological hazards associated with the plants and with the quantities and physical and chemical forms of the materials used in the plants.

The Development of Standards

It is up to both industry and the AEC to accept the responsibility for assuring the proper use and handling of special nuclear material. Members of the AEC Regulatory Staff have actively sought out membership and participation in national standards groups. Of particular interest here are those sponsored by the Institute of Nuclear Materials Management, which are developing plant and material protection standards. A number of standards have been and are being developed in the areas of material accounting and measurements and in

statistical treatment of data. However, there are no similar groups actively developing standards for physical protection of either material or plants, except for reactors, or for the protection of material in transit.

The AEC recently issued effective and proposed rules encompassing the entire spectrum of material and plant protection, to amend 10 CFR Parts 50, 70 and 73. A considerable number of standards will need to be developed to assist in implementing these amendments.

The effective amendments to 10 CFR Part 73 limited the quantities of plutonium and certain types of uranium that could be shipped by passenger aircraft and were aimed at eliminating the possibility of significant quantities of special nuclear material being illegally obtained by highjacking a domestic passenger aircraft. Other proposed amendments relating to material in transit would apply to licensees who ship 5000 grams or more of uranium-235 (contained in uranium enriched to more than 20 percent), or 2000 grams of plutonium or uranium-233, or a combination of these materials which is less than 5000 grams if the plutonium or uranium-233 content is greater than 2000 grams.

To complement these proposed regulations, when they become effective, for truck shipments standards are needed in such areas as armed escorts traveling in a separate vehicle, trucks or trailers specially designed to protect against theft or diversion, continuous communication capability, and continuous monitoring methods. When cargo aircraft are used, the number of enroute transfers would be minimized and would be observed by monitoring personnel. Standards will be needed here.

Under the proposed amendments, operators of fuel fabrication and reprocessing plants would have to: (1) equip and train guards and watchmen to protect against industrial sabotage; (2) establish a "protected area," enclosed by a physical barrier; (3) provide for control of access by individuals, vehicles and packages to the protected area; (4) install lighting along the perimeter of the area; (5) develop a response capability to intrusion; (6) establish liaison with law enforcement authorities for assistance when necessary; and (7) establish an emergency two-way communication link with law enforcement authorities. Complementary standards will be needed in all these areas.

Assurance against undetected loss or diversion of material can be achieved only when the physical security system is backed up by physical inventories. Various systems of material control and accountability can be used to account for the material. However, a material balance based on a measured physical inventory is the only means for assuring that the physical security system is working and that no significant losses or diversions have gone undetected. In addition, the interval between verification of material balances should be shorter as the strategic value increases.

The proposed amendments to Part 70 would reduce the quantity of special nuclear material for which more comprehensive material control and accountability requirements would be applied from 5000 grams to one effective kilogram—a term used to define equivalent quantities of plutonium, uranium-233 and uranium-235 and determined by making specific calculations for each type of material; the term has been used by the International Atomic Energy Agency for a number of years in connection with material safeguards. Licensees authorized to possess more than one effective kilogram would have to perform more frequent inventories and the maximum time intervals between inventories would be: 30 days for plutonium, 60 days for uranium-233 and for uranium enriched 20 percent or more in uranium-235, and 180 days for low-enriched uranium. Annual physical inventories would be required by licensees authorized to possess more than 350 grams of special nuclear material but less than one effective kilogram.

Minimum standards for the quality of the inventories and for the overall material measurement program as well as additional reporting requirements are included in the proposed amendments to Part 70. All of these regulations are for the most part performance oriented aimed at instructing licensees on what shall be done rather than how it should be done. As these amendments to the rules become effective, a large number of industry standards and regulatory guides will be needed. The Regulatory Staff of AEC will continue to be working to develop guidance in material control and accounting, inventory methods and calculations. More guidance is needed for material and plant protection and for transportation security, and AEC staff will be working toward providing these more detailed guidelines.

In time, these guides will all be developed, but the time could be shortened with the help of outside groups. Presently, the outlook is for very limited outside participation in this effort and this is unfortunate. In the final analysis, as the actual custodian of the materials, it is the prime responsibility of industry to protect their plants and materials. Since this is the case, industry must become more involved in defining the procedures for carrying out the rules. The industry must recognize the immediacy of the need for these standards and guides and be willing to put forth the effort to develop them on a more timely basis than is presently the case. The N-15 Committee has attempted to obtain greater representation from industry and has developed a number of standards accepted by ANSI. For the most part, however, these standards are the easier ones, the general framework standards. Concerted effort is needed to develop some of the more specific standards mentioned earlier. Ten such standards in the materials and plant protection area were identified recently in correspondence with ANSI, but only seven have been assigned for committee action. Until groups outside the AEC do assume more responsibility in this area, standards development will continue, but at a pace much less than is needed.

Table 1

Type of Operation	Number of Operations Present	Planned
UO_2 Production	8	3
UO_2 Pelletizing	9	1
UO_2 Fuel Fabrication	10	1
Special Fuels	13	1
Plutonium Fuels	9	2
U and Pu Fuel R&D	14	2
Gold Scrap Recovery—U	8	4
Cold Scrap Recovery—Pu	4	5
Spent Fuel Reprocessing	1	2
U to UF_6 Conversion	3	3
Total	79	24

Thorium and depleted uranium processing facilities not included.

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PERSPECTIVES ON THE ENERGY CRISIS

By Frederick Forscher

If there is still any doubt that this nation suffers from an Energy Crisis, the following news items—all from Jan. 18, 1973,—should settle this issue. Even if all our fuel sources should be nationalized overnight, there still would be an energy crisis; that is to say, we are in a shortage situation, with or without the profit motive.

Transit fuel shortages are rapidly developing, and service curtailments may begin soon. Trucking, rail, barge, airline and mass transit representatives are meeting with U.S. officials.

Governor Shapp of Pennsylvania has ordered state offices to set thermostats at 68 degrees during working hours and 63 degrees during off hours, and urged homeowners to do the same. The governor also recommended that electric use be limited since much electric power is produced at plants burning oil.

Oil-import curbs were eased by President Nixon to meet surging U.S. energy needs. The unprecedented relaxation includes unlimited purchases of foreign home-heating oil and raises by over 50 percent the crude and refined products that may enter east of the Rockies.

It is hard to think of any other vital national issue which is so complex, so fraught with environmental controversy, so entangled with politics and special economic and regional interests, and so involved with U.S. import policies and even national security. It is also an issue that involves Technology at every level and in every phase of it. i.e.: supply, transportation, distribution and consumption. Therefore, it seems appropriate to present a few socio-technological considerations to help us evaluate the policies and proposals that will surely make headlines in the near future.

To begin with, we must learn to separate the idea of natural resources such as coal, oil, gas and uranium, from the concept of energy. Energy is different from anything we are exposed to, except perhaps time. Energy cannot be recycled! What we are using today, for whatever reason we think we are justified in doing so, will be gone forever. It will not be available to all the generations yet to be born; it will not grow back like the harvests in the fields or the trees in the forests. It will not renew itself like people, from generation to generation. It is literally gone forever like the time of day.

Matter can be recycled, energy cannot. Therefore, the only thing a consumer truly "consumes" is energy; everything else is, or could be, recycled.

The fundamental significance of energy in the physical and biological sciences is now well recognized. On the other hand, in the social sciences, the concept of energy per se, has not gained much importance, at least not up to now. While students of metabolism, particularly human metabolism, delve into the details of the life-giving energy-conversion mechanisms that metabolism represents, the corresponding study for the social organism has not even been given a name. I call it Social Physics—a scientific specialty of the future. Yet, the need for energy and natural resources are as great in our society to sustain its functions, and to survive, as energy and food is needed to sustain life in any organism.

Most of the energy comes to us today from the fossil fuels, or from uranium. A small portion is supplied by the potential energy of water and the kinetic energy of the wind. Geographically, like socially, we are not a closed system; we import much of our energy needs from abroad in the form of oil, LNG (liquified natural gas) and uranium. We have not yet perfected the means for large scale use of the sun's energy, except through the food chain. After multiple conversions, all of the various forms of energy, from all sources, end up as heat, and thus become unavailable for future use.

This is, of course, also true for the energy contained in the food we eat. An active person consumes about 1 million BTU per year in food energy. Since the days of the caveman, we have learned to make our life easier by the controlled use of various energy sources: from the first tool man ever made with the help of fire, to the sophisticated construction of nuclear power stations; from the days when man lived by the sweat of his brow (one million BTU per year), to today's standard of living, where each American uses over 300 million BTU per year. Today's use of energy is equivalent to having 300 slaves work for each one of us, every single day of the year. Incidentally, this compares to 160 million BTU for the average European, and 5 million BTU for the average Indian.; in other words, Americans live about twice as well as the average European, and 60 times as well as the average Indian.

We see that our society, and other societies, have come a long way from the days of Adam and Eve. For some students of history, the increased use of energy per capita represents a measure of "progress." Even if one does not agree with such a definition of progress, one must agree that life has become progressively easier — not necessarily fuller in the sense of quality of life — but easier with increasing usage of energy per capita. It is therefore not surprising to find almost general agreement, that the average energy per capita — in any society, and at any historical age—is a valid measure of the "Living Standard" of this society. The U.S. standard of living is the highest that was ever achieved anywhere. We use about 40 percent of the world's resources to achieve this standard for 6 percent of the world's population.

The energy crisis seems to say: this cannot go on for much longer. The humanitarian in us will agree, we must decrease our consumption. But where and what are we going to cut, and still maintain our growth, social momentum, and expectations for an even better life? The answer involves the idea of enough energy to do all the things, society needs to do. But the hard question, the non-technical question remains: How much is enough?

NUCLEAR POWER

By L. E. Minnick
Vice President
Yankee Atomic Electric Co.
Westboro, Mass.

About fifteen years ago, the primary interest was centered around how a nuclear plant worked. "What is fission and what are neutrons?" After we had completed the Yankee plant, we found that people wanted to hear about our experiences with the plant and about our plans to put that experience to work in future plants here in New England. Then, perhaps six or seven years ago, and up until about three years ago, it seemed that people lost interest in things nuclear. During that particular period, we received very few requests to make presentations. As a result, during that period we who are engaged in the nuclear industry made what turned out to be a serious mistake. We assumed that nuclear plants had been accepted and that our job was simply to design them and to operate them as economically and as safely as we knew how.

In recent years, of course, we have found how wrong we were. We have suddenly found that there were more questions being asked, more accusations leveled and more criticisms articulated than any of us could hope to cope with in a dozen sessions such as this one,—especially considering that these concerns range in subject matter over a spectrum beginning with the general and philosophical and extending all the way to the most technical and complex details.

In any event, we have found in the last year or two that most opportunities to discuss nuclear power are provided to us in the form of a debate. Such presentations have the minor and single virtue of establishing that our subject matter must consist primarily of the best defense that we can muster to the particular concern of the opposition on that particular occasion.

Today it seems to me you have presented an opportunity to do something different and I will try to take advantage of it.

What I hope I can do today, and intend to ask you to do also, is to step back a little from the fray and to try to view nuclear power and the nuclear power controversy from a somewhat larger point of view—and with more emphasis on perspective and on a sense of proportion relative to other aspects of the real world and of our way of life.

If I may, perhaps I should begin by talking about the point of view which is most familiar to me. In the eyes of a utility company nuclear power has been proven—partly through our own efforts here in New England on the early Yankee plants in Massachusetts and in Connecticut—to be a reliable and economical source of electrical energy and one with less effect on the environment than any other available to us. Additionally, we see the cost of nuclear fuel still gradually being reduced, whereas we see the cost of oil and coal moving upward at a continuing and alarming rate. From our particular point of view the primary disadvantage of nuclear power—and a serious one at the moment—is that it is the least susceptible to reasonable scheduling as to when or even whether any particular new plant once committed can be expected to be available to produce power. This uncertainty as to scheduling, together with significant uncertainties as to specific requirements for plant design, result in compounded uncertainties in any prediction of the ultimate cost of future nuclear plants. There uncertainties are largely a direct result of the controversy regarding nuclear power and the environment in general and of the reaction of government and the regulatory agencies to that controversy.

At this point then it may be appropriate to try to look at nuclear power from a national point of view. At the moment, there are 28 nuclear power plants operating in this country. There are 52 plants under construction and 70 more on order. The total capability represented is 130,000,000 kilowatts which is the equivalent of about one third of the total generating capacity in existence at the present time.

Viewed from the standpoint of overall sources of energy, we find that American sources of gas, oil and coal are reaching limits of capacity to supply our energy requirements. Natural gas supplies, in particular, can barely meet the demands already created. There is considerable quantity of coal still in reserve, but it generally contains unacceptable quantities of sulfur, is difficult to mine with acceptable costs, safety and environmental impact, and is difficult, expensive and dirty to transport and to store. Our domestic petroleum sources are limited in extent and must be supplemented massively from foreign sources. Already some twenty percent of our oil requirements are imported at an annual penalty to our balance of payments of $5 Billion. Corresponding predictions for the year 1985 are 50 percent and 25 Billion. Troublesome as trade deficits are, even those problems are outweighed by the risks involved in becoming so dependent on distant sources, for something as basic to our civilization as energy. We run the risk of blackmail in terms of price at the source, of unstable policies and governments in the Middle East and, finally in terms of vulnerable transportation over thousands of miles of open ocean.

At this point it seems worth emphasizing that the only basic energy raw material available in unlimited quantities within the continental United States and, therefore, not subject to any of the considerations affecting the conventional fuels is uranium.

Other potential sources of energy are being mentioned more and more frequently, as the proportions of the energy crisis become clearer. Under the circumstances there can be little disagreement that any reasonable approach should be thoroughly evaluated—and this procedure is continuing. Each of the suggestions, however, whether based on the fusion process, solar power, geothermal energy or even harnessing the wind suffers from the same major difficulty; over and above immediate technical difficulties. The simple fact is that each is so early in its development that it is nearly inconceivable that it can be moved ahead rapidly enough to contribute significantly in the critical twenty years ahead.

At this point it is probably appropriate that we attempt to understand the concerns about nuclear power from the point of view of those who are sincerely trying to improve our en-

vironment. In all honesty this is very difficult for me to do since I am convinced that some of the major advantages of nuclear power come under the general heading of minimal effect on the environment.

Certainly nuclear power is relatively clean. Beginning at the source there are no oil wells and no pipelines, no tankers and no oil spills, and no strip mining. Nuclear plants themselves are clean and surrounded not by oil tanks or coal piles, but rather by acres of land held in its natural state. Finally, there are no smoke, fumes or ash. In only one respect can one's concern for the environment lead to greater concern in regard to a nuclear plant. That is that it takes larger quantities of water to condense the steam which drives the turbine in a nuclear plant. The water is not necessarily one degree warmer; it simply takes more of it. This aspect is clearly a legitimate concern and one which is extremely difficult to completely allay. Exact predictions of future effects are difficult, if not impossible, about which to be absolutely and unarguably positive. On the other hand a team of biologists has been studying the Connecticut Yankee plant for years—both prior to and ever since it began to operate in 1967—without finding any significant effect on the ecology of the Connecticut River.

Perhaps the most understandable concern for nuclear power in general has to do with its safety. I say this despite my own feeling that nuclear power represents only a minute risk to the public and a risk which is certainly less than many we accept casually every day of our lives. The concern is understandable because of the unfortunate genesis of nuclear power; because of its complexity and unfamiliarity; and oddly enough, at least partially, I think, because such stringent efforts have been made to be certain that it is as safe as is humanly possible.

In the early 1950's when President Eisenhower announced the Atoms for Peace Program and the Atomic Energy Act was passed, it was perfectly clear that unlike any other new industry, the implicit risks were recognized and the basic philosophy was established that the safety and well-being of the public would be protected at all costs. This mandate has been carried out in exemplary fashion. Although there are and have been for years many dozens of large and small reactors in operation, there has never been a single instance of harm to a member of the public from this source. Is there any other major industry that can make that statement? Is there any other industry which as a prerequisite to being allowed to proceed must prepare several technical volumes which not only describe every significant design detail, but further postulate and analyze every credible combination of unfortunate events and must demonstrate that the end result of all postulations is acceptable? Is there any other industry which makes such documents available to anyone who wishes to examine them and which must ultimately answer in a public hearing any questions anyone wishes to ask? I submit that the answer to my questions is "No, there is no similar industry" and that that is not only one of the reasons for our remarkable safety record, but possibly one of the reasons why at least some people are more concerned about nuclear power than about some other risks that have never been so thoroughly and publicly scrutinized.

There are, of course, many other factors contributing to the concern for nuclear power and to the controversy surrounding it. As I have mentioned, it is a complex technology. It is difficult to answer effectively, convincingly and briefly even a majority of the many complex questions which can be asked. In many cases there is no absolute answer which can be given. No honest engineer can state that anything is absolutely safe. Under the circumstances it is certainly not too surprising that there are occasional differences of opinion—even among "experts."

I feel, however, over and above the circumstances already mentioned that contributing to this controversy are many of the more fundamental questions which make the times we live in more and more filled with controversy on many subjects. I have in mind the growing distrust in some quarters of technology in general, of industry, of government and, for that matter, of the utilities. Perhaps we are, at least to some degree, experiencing one manifestation of what has been characterized as the "Age of No"; wherein it seems that some people are far more interested in why things should not be done than in why they should be done.

My intention today has not been to change your mind if you are concerned about nuclear power. I don't feel it would be reasonable to expect that from either you or me in such a short time. What I do hope is that I have convinced you that it is an important question and that to the extent you may be concerned that you should make the effort to examine and to understand the whole situation.

Further, I think that we should all realize that although there may be some residual risks to proceeding with nuclear power, or any other technology for that matter, that there are also risks in not proceeding, which deserve equal consideration.

Editor's Note. Mr. Minnick's talk was sent to us with a recommendation that the Journal be used as a vehicle for its message. The agreement in favor was that "the utility viewpoint is one which is seldom heard." While the content of Mr. Minnick's talk is not what we had in mind for the Journal, after reading and re-reading and carefully noting many subtleties, we herewith submit a "utility viewpoint."—Curtis G. Chezem.

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Facts for Authors

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PLUTONIUM

By E. R. Johnson

What is Plutonium?

Plutonium, element number 94, was discovered in 1940 by Glenn T. Seaborg, Arthur C. Wahl and Joseph W. Kennedy at the University of California.

There are a total of 15 isotopes of plutonium whose half lives range from 20 minutes to 76 million years and whose atomic weights range from 232 to 246. All plutonium isotopes except the isotope of atomic weight 241, are αemitters; some of the longer-lived isotopes also emit some β-radiation and plutonium 241 is a nearly pure β-emitter. These isotopes are formed by a variety of mechanisms, most of which involve the bombardment of certain uranium isotopes with α-particles, neutrons or deuterons.

Plutonium does occur in nature, although in such small amounts that it does not constitute a practical source of the element. Some pitchblende ores (once a principal source of uranium and radium) contain as much as 1 part plutonium in 10^{11}.

Plutonium is formed in nuclear power reactors principally through the neutron bombardment of U-238. Since most of the commercial nuclear power reactors are fueled with uranium which has a low level of enrichment in uranium-235, the bulk of the uranium contained in the fuel is U-238. The principal plutonium isotope produced is Pu-239.

$$\text{U-238} + \text{n} \longrightarrow \text{U-239} \xrightarrow{\beta^-} \text{Np-239} \xrightarrow{\beta^-} \text{Pu-239}$$

However, depending upon the operating conditions of the reactor and the degree of exposure to neutrons, a number of the other plutonium isotopes are produced. For example, Table 1 shows the principal isotopes of plutonium in the spent fuel of a typical nuclear power reactor, at current design exposure levels.

TABLE 1

PRINCIPAL ISOTOPES OF PLUTONIUM
TYPICAL POWER REACTOR SPENT FUEL*

	Half Life	Grams/MTU	% of Total Pu
Pu-238	86 years	166	1.8
Pu-239 (fissile)	24,360 years	5380	59.3
Pu-240 (fertile)	6,580 years	2170	23.9
Pu-241 (fissile)	13 years	1010	11.1
Pu-242	379,000 years	349	3.9
		9075 g	100.0

* 1060 MWe PWR @ 33,000 MWD/MTU

What are the Properties of Plutonium?

Plutonium can exist in the form of metal, alloys and a number of chemical compounds. Plutonium metal can be melted into ingots, machined, rolled and extruded to produce rod, foil, wire and other fabricated forms. Plutonium metal forms alloys and intermetallic compounds with most other metals, with the exception of certain alkali, alkaline earth and rare earth metals which are immiscible with plutonium in both solid and liquid forms. Plutonium has four oxidation states (III), (IV), (V) and (VI) and forms stable compounds with all the nonmetallic elements except the rare gases. Plutonium can exist in the form of plutonium oxide, plutonium carbide, plutonium nitrate, plutonium nitride and many other compounds.

Plutonium in all chemical and physical forms has some properties that are specifically unique and which make it an extremely useful material.

1. Some plutonium isotopes are fissionable — Pu-239 and Pu-241 are the most abundant fissile isotopes which result from spent nuclear fuel from light water reactors. This fissionable nature of Pu-239 and Pu-241 makes by-product plutonium from nuclear power reactors valuable as a reactor fuel. Fissile plutonium can be used in place of U-235 in light water reactors and when this is done the fuel is called plutonium recycle fuel inasmuch as the plutonium resulting as a by-product of reactor operation is recycled after separation from spent fuel. Fissile plutonium is a particularly good fuel material for the breeder reactors being developed in the U.S. and abroad and expected to be a source of commercial electric power by 1990. Of course, the fissile nature of these plutonium isotopes also requires that considerable care be exercised during fuel fabrication and reprocessing of spent fuel to prevent the occurrence of an accidental criticality (sustained nuclear fission) in such processing facilities.
2. Plutonium is self-heating — The α-emission from plutonium when absored into the crystal structure of a plutonium compound, mixture or metal causes the mass of the structure to increase in temperature. This property is readily noticeable in plutonium metal which has relatively low thermal conductivity compared to most metal. Pu-239 metal is warm to the touch and Pu-238 metal is too hot to hold in the hand without thermal protection. This property of Pu-238 makes it a valuable source of heat for use in thermoelectric generators, heart pacers, and other prospective applications requiring a concentrated and reliable source of heat. Pu-238 has been and is being used as a heat source in thermoelectric generators employed in a number of space missions, both manned and unmanned. The self-heating property of plutonium and the radioactive decay products associated with the α-emission also present some problems.
3. Plutonium is extremely toxic—This toxicity is also due to the fact that plutonium emits α-particles which have little penetrating power (less than 4 centimeters in air, and less than 50 microns in body tissue). Because of this low penetrating power the particles dissipate their entire energy in the tissues which surround the point of disposition in the body. Plutonium can enter the body in three basic ways:

(Continued on Next Page)

(i) by inhalation and absorption through the lungs,

(ii) by ingestion and absorption through the gastrointestinal tract,

(iii) by absorption through the skin.

The bulk of the plutonium that is retained by the body is deposited in the skeleton. Plutonium retained in the body can result in cancer, leukemia, or otherwise damage the various organs of the body depending upon the mechanism of intake and the rate of intake. Protection of persons working in plutonium processing facilities from the toxic consequences of plutonium intake is accomplished through the inclusion of extensive health safeguarding equipment in the basic design of the facilities as well as by carefully considered operating procedures and practices. Protection of persons outside of plutonium processing facilities is accomplished through the special processing and monitoring of all wastes, and through the inclusion of features in the basic facility design for the protection of the facility against natural phenomena and hypothetical serious accident situations.

The impact that this value of plutonium has on nuclear fuel cycle costs is demonstrated in Table 3. From this table it can be seen that, at equilibrium operation, a typical 1000 megawatt electric pressurized water reactor has annual fuel costs $10-11 million. This can be offset significantly by reprocessing the spent fuel for recovery of contained uranium and plutonium. The recovered uranium is worth about $940,000 annually, and the plutonium generated is worth about $1.3 million annually. The plutonium thus can be used to offset approximately 12 percent of the yearly fuel cost, based on current fuel cycle economics.

What is the Significance of Plutonium on Nuclear Fuel Cycle Economy?

Plutonium is useful both as a fuel for light water reactors of the type being used commercially at the present time and as a fuel for breeder reactors which are expected to be used for the commercial production of electric power by 1990. Since significant quantities are produced as a by-product of the operation of light water reactors, the early use of such by-product plutonium by recycling in the fuel of light water reactors has a profound effect upon the reactor's fuel cost.

To illustrate this effect let us assume that, at equilibrium, a reactor is fueled with 3.2 per cent enriched uranium. At present day prices 3.2 percent enriched uranium in the form of uranium hexafluoride (the feed material to the fuel fabrication process) costs about $286/kilogram; therefore, the value of the contained uranium-235 is about $8938/kilogram. Let us also assume that fuel containing plutonium is 50 percent more expensive to fabricate because of the higher costs for the fabrication facility and higher attendant operating costs—both due to the toxic nature of plutonium and the need to adequately protect the workers in the fabrication facility as well as the general public which require more safety features to be designed into the facility as well as the use of more stringent operational procedures. If the fabrication of low enrichment uranium fuel costs $70/kilogram and the fabrication of mixed oxide fuel (uranium oxide/plutonium oxide) fuel costs $105/kilogram. Then the added cost of fabricating plutonium-bearing fuel for light water reactors is $35/kilogram. This results in an effective reduction of $1522/kilogram in the value of fissile plutonium contained in fuel which has a fissile plutonium content of 2.3 percent—which means that the net value of fissile plutonium is $7416/kilogram. These calculations are set forth in Table 2.

What is the Significance of the Use of Plutonium in Conserving Reserves of Uranium?

One reactor can be refueled exclusively with plutonium from the plutonium produced each year by about four reactors of the same size. However, new reactors being started up require more fuel initially—3 to 4 times the annual replacement requirement.

The recycling of plutonium in the fuel of commercial nuclear power reactors will have a significant impact upon the future needs for uranium raw material and separative work capacity. Table 4 shows a projection of both uranium raw material and separative work requirements in the absence of plutonium recycle and with plutonium being recycled. While the impact of plutonium recycle is very small at the present and in the next few years, the recycling of plutonium a decade from now will reduce the amount of uranium and the attendant separative work required by about 14-15 percent. This will enable us to satisfy the expected demand for nuclear fuel while conserving a valuable natural resource and minimizing the amount of expensive separative work capacity required—and will enable us to do so at reasonable cost.

TABLE 2

ESTIMATED VALUE OF FISSILE PLUTONIUM
(In Light Water Reactors)

(1) Assume Fissile Pu Equivalent to U-235 for Fuel Use

(2) Assume Value of 3.2% Enriched Uranium is $286/kg

$ 7.00/lb U_3O_8	for yellow cake
$ 1.20/lb U	for conversion
$36.00/swu	for enrichment

Value of U-235 in 3.2% enriched U is $8938/kg

(3) Assume Plutonium Fuel Costs 50% More to Fabricate Than Uranium

($105/kg vs. $70/kg or $35/kg fuel)

(4) Assume Plutonium Recycle LWR Fuel Contains 2.3% Fissile Plutonium

(.023 kg/kg fuel)

(5) Assume Penalty on Value of Plutonium for Increased Fabrication Cost

$$\frac{\$35/\text{kg fuel}}{.023 \text{ kg fissile Pu/kg fuel}} = \$1{,}522/\text{kg fissile Pu}$$

(6) Value of Fissile Plutonium

Value U-235	$8,938/kg
Fabrication Penalty	1,522/kg
Net Pu Value	$7,416/kg

TABLE 3

EQUILIBRIUM ANNUAL FUEL CYCLE COST
FOR A MODEL 1000 MWe PWR*

ITEM	ANNUAL COSTS ($000) TOTAL	MILLS/KWH	% OF TOTAL FUEL COST
Yellow Cake			
(454,895 lb U_3O_8 @ $7)	$3,184	.427	29.8%
Conversion			
(174,943 kg U @ $2.65)	464	.062	4.3%
Enrichment			
(27,835 kg 3.2% enr U @ $36/swu)	4,314	.579	40.4%
Fuel Fabrication			
(27,000 kg 3.2% enr U @ $70)	1,890	.254	17.7%
Transport & Reprocessing			
(26,050 kg @ $32)	834	.112	7.8%
TOTAL	$10,686	1.434	
Recovered Uranium			
(25,790 kg 0.9% enr U)	(941)	(.126)	(8.8%)
Recovered Plutonium			
(245 kg @ $5,225)	(1,280)	(.172)	(12.0%)
NET COST	$8,465	1.136	

* Excluding capital charges.

TABLE 4

EFFECT OF PLUTONIUM RECYCLE ON YELLOW CAKE AND ENRICHMENT REQUIREMENTS*

(U.S. Reactors)

	U_3O_8 REQUIREMENTS (SHORT TONS)			SEPARATIVE WORK (1000 METRIC TON UNITS)		
YEAR	WITHOUT Pu RECYCLE	WITH Pu RECYCLE	% CHANGE	WITHOUT Pu RECYCLE	WITH Pu RECYCLE	% CHANGE
1974	17.3	16.7	3.5%	9.1	8.9	2.2%
1976	22.6	21.1	6.6%	12.5	11.7	6.4%
1978	30.7	28.6	6.8%	17.5	16.0	8.6%
1980	38.6	34.2	11.4%	22.7	20.5	9.7%
1982	50.8	44.3	12.8%	30.8	26.5	14.0%
1984	62.6	53.9	13.9%	39.2	33.4	14.8%

*Source: WASH-1139

Where Does Plutonium Exist in the Nuclear Fuel Cycle and How is it Handled?

There are five general areas in the nuclear fuel cycle where plutonium is likely to exist and where special care must be taken to contain the material. These are (1) the reactor facility, (2) during transportation of spent fuel and radioactive wastes, (3) during spent fuel reprocessing and radioactive waste processing, (4) during fabrication of plutonium fuels and (5) during shipment of plutonium.

At the Reactor

The fuel assemblies used in a reactor are constructed in such a way that they totally contain the fuel material, encapsulated in Zircaloy or stainless steel. In the event of a loss of this containment during the residence of the fuel in the reactor, through the development of leaks in the fuel rod cladding, contamination of the reactor's primary coolant and water in the spent fuel storage pool will result—but such contamination will consist largely of volatile fission products which can migrate through perforations in the cladding. Only in the unlikely event of a major cladding rupture would there be even trace quantities of plutonium, which would, of course, be accompanied by highly radioactive non-volatile fission products. Provisions are made in the design of a nuclear power reactor for the cleanup of the primary coolant for removal of essentially all radioactive contamination. Most cleanup systems involve the use of filtration, evaporation, and ion exchange and are designed to handle high levels of contamination of the primary coolant as well as contaminated water from the spent fuel storage pool. The radioactive material removed from the primary coolant and spent fuel pool water is concentrated and then either fixed in concrete or bitumen and buried in controlled burial areas. It should be pointed out that in connection with the operation of nuclear power reactors and the wastes resulting therefrom, the contamination problems posed by plutonium are minimal since its existence in the fuel is as plutonium oxide which is generally in solid solution with uranium oxide in a dense pellet from which is very insoluble in the coolant water.

During Transportation of Spent Fuel and Radioactive Waste

The shipment of spent fuel from the nuclear power reactor to the spent fuel reprocessing plant is accomplished in large shielded shipping casks which are transported either by rail or by truck. Principal design criteria for the casks include requirements intended to minimize the probability of escape of radioactive material or loss of shielding during serious accident conditions. The cost of these shipping casks ranges from $100,000 for a cask to handle 2 PWR assemblies to $600,000 for one capable of carrying 10 PWR assemblies. Shipping casks are really large shipping boxes; but as the cost of them might suggest, they are of sophisticated design and construction. These same types of shipping casks will also be used in the future for the transport of high level radioactive waste which has been solidified and is destined for a central waste storage repository. It should be emphasized, however, that plutonium in the spent fuel elements is still largely contained, as it is in the reactor, and given the design criteria applicable to the casks themselves, is likely to remain so, and is thus not, per se, a hazard in shipping.

During Reprocessing

The reprocessing of spent fuel to recover unburned uranium and plutonium values is accomplished in elaborately designed facilities—so designed to protect workers as well as the general public from the high level of radioactivity from fission products, during both normal operating conditions and accident conditions.

The reprocessing operation involves the shearing of the fuel into small pieces, dissolution of the sheared fuel in nitric acid, solvent extraction of the dissolved fuel to separate the uranium and plutonium from fission products and from each other, final purification and concentration of uranium and plutonium and conversion into forms suitable for their reuse. Uranium is generally recovered as uranyl nitrate solution or as uranium hexafluoride. Plutonium is generally recovered as plutonium nitrate solution or plutonium oxide. All such forms are suitable for recycle to fuel fabrication operations.

The reprocessing plant is a massive concrete structure which is subdivided into processing cubicles or cells which contain remotely controlled and operated equipment. All operations except the final conversion operations are conducted in such cells. Equipment in the cells is operated remotely by operators using servomechanisms and other devices with visual access through shielded windows, periscopes and television cameras. Maintenance and replacement of equipment is performed remotely.

Ventilation systems in reprocessing plants are elaborately instrumented to balance air flows and to monitor pressures, radioactivity and the like. Air flows are directed from the areas of least contamination potential to those of the highest potential. The air pressure in the process cells is maintained at a lower operating level than in the non-process and auxiliary areas where there is little or no contamination. Acid fumes generated in the process cells are removed from the ventilation air by scrubbing. Radioactive gases are removed by passing through beds of absorbing materials. Radioactive particles are removed from exhaust air by filtration through high efficiency filters in banks, and in series with one aother. Liquid wastes from a reprocessing plant are processed by a number of different methods for removal of radioactivity down to an acceptably safe level prior to discharge to the environment, depending upon the level and nature of the radioactivity contained in the waste. All liquid effluents that are discharged from the plant are carefully monitored prior to such discharge. Generally speaking, the radioactivity in reprocessing plant wastes (including the plutonium component) is concentrated in solution form for storage. At the present time this concentrated waste is stored in underground storage tanks as a neutralized solution at the reprocessing plant site but a recently enacted AEC regulation requires for the future that a reprocessing plant solidify these wastes after no more than five years of storage and transfer the solidified wastes to a central Federal waste repository after no more than five years from the time they were solidified.

As may be evidenced by the foregoing description of the extensive nature of the reprocessing facilities, the facility design is strongly influenced by the radioactivity of the materials being handled and the attendant need to protect the plant workers, and to prevent the release of more than the minimal quantities of radioactive materials to the environment which is necessary in order to adequately protect the general public. The cost of a reprocessing plant having a capability for processing 5000 kilograms of uranium and plutonium per day is in the range of $70-90 million, whereas absent the radioacitivity (and toxicity) problem, the same operations could be conducted in a facility costing on the order of $2-3 million.

During Fuel Fabrication

The fabrication of plutonium bearing fuel is also accomplished in elaborately designed facilities—so designed to protect both the workers and the general public from contact with plutonium. However, such fabrication facilities are not as elaborately designed as the spent fuel reprocessing facilities since the same high level of radioactivity does not prevail in such fabrication facilities.

The fuel fabrication operation involves the conversion of uranium hexafluoride to uranium dioxide, blending uranium and plutonium oxides together, pressing the blended oxides into pellets, high temperature sintering of the pellets, grinding the pellets to final dimensional requirements, encapsulating the pellets in Zircaloy or stainless steel tubing and assembling the resulting fuel rods into fuel bundles or assemblies. Plutonium-bearing scrap material produced during fabrication operations is processed for recovery of both uranium and plutonium by a process similar to that used for the reprocessing of spent fuel.

Plutonium fabrication facilities are contained in structures which are capable of withstanding assault by natural phenomena such as tornadoes, hurricanes, earthquakes and floods. There are three basic levels of containment built into a plutonium fabrication facility. The primary containment is effected through the use of glove boxes, equipment enclosures and totally enclosed transfer devices. The secondary containment is effected by dividing the processes into individual process areas which are totally separated from one another. The tertiary containment is provided by the building structure which is designed to retain its integrity under credible conditions of physical stress.

While the ventilation systems of plutonium fuel fabrication facilities vary in design, it is usually the practice to either have ventilating air pass from the area of the lowest prospective contamination into the areas of progressively higher contamination or to separately ventilate primary containment and structures. A negative pressure is maintained in all containment areas, with the negative pressure in the primary containment areas being greater than that maintained in the individual process areas and the structure as a whole. Emergency power is provided to operate the facility ventilation systems in the event of a power failure. The air from each of the containment areas is exhausted through high efficiency air filters to remove particulate plutonium from the air streams. These air filters are tested for efficiency periodically. All exhaust air, as well as air in working areas, is continuously monitored and operations are stopped if an unsafe level of activity is detected.

Contaminated waste (mostly liquid) from plutonium fuel fabrication operations is processed to concentrate the contamination and the resulting concentrate solidified in concrete or bitumen prior to burial in controlled areas. All liquid effluents discharged from the facility are carefully monitored prior to such discharge.

Plutonium-bearing materials are removed from primary containment areas only after they have been suitably packaged, or encapsulated. Such removal is effected through air locks and utilizing bagging procedures on the item removed. Shielding is provided for glove box enclosures where radioactivity levels are sufficiently high to require it for safety of the facility workers.

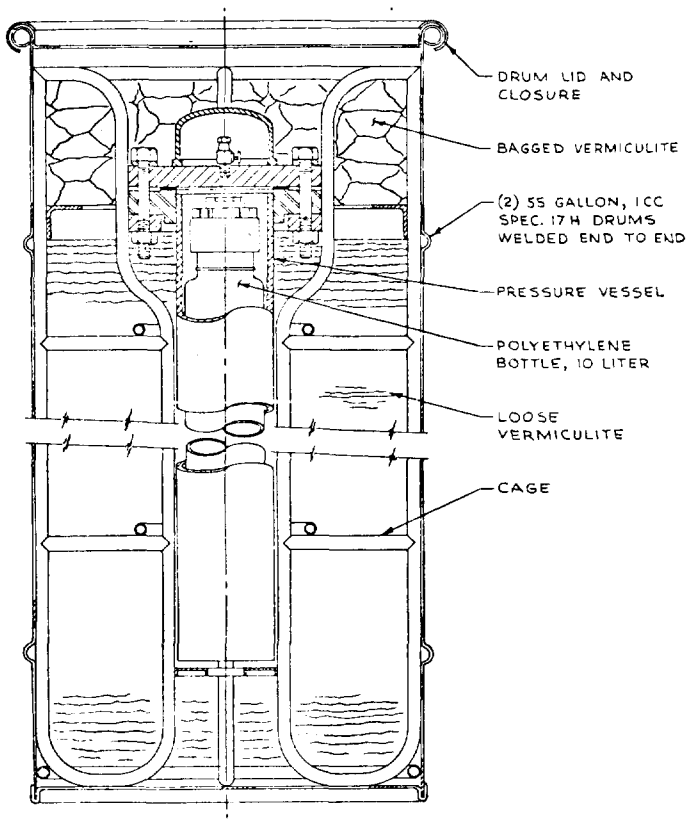
As is evidenced by the foregoing description of the nature of the plutonium fuel fabrication facilities, the facility design is strongly influenced by the toxicity of the plutonium being handled and the attendant need to protect the plant workers and to prevent the release of plutonium to the environment in quantities which could present a hazard to the general public. The cost of a plutonium fuel fabrication plant is about 1.5-2 times as high as that for a correspondingly-sized uranium fuel fabrication plant which processes only low enrichment uranium. This is a result of the additional safety features required and the provisions for special operational procedures that are necessary for the processing of a toxic material.

During Plutonium Transport

Special care must also be taken during the shipment of plutonium in order to protect against the possibility of release of contamination to the environment.

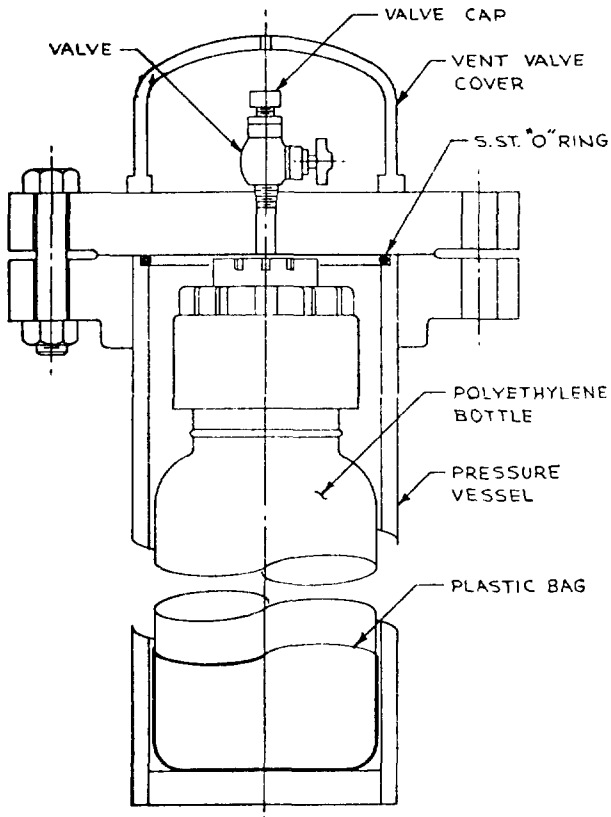
As an example, shipment of plutonium nitrate solution is generally effected in a polyethylene bottle with a vented screw cap. This polyethylene bottle is placed inside a steel container which is equipped with a bolted flange on the top. The steel containment vessel is positioned in a spider-like device inside a 55-gallon drum or two 55-gallon drums, welded end to end. The steel container is so held in the spider arrangement that it is maintained suspended in the center of the drum(s). Vermiculite is used to fill the voids. In the event of an accident condition resulting in rupture of the polyethylene bottle, the solution would be contained in the steel containment shell. In the event the steel containment shell were ruptured, the plutonium nitrate solution would be absorbed in the vermiculite. Figures 1, 2 and 3 show a typical plutonium nitrate

FIGURE 1



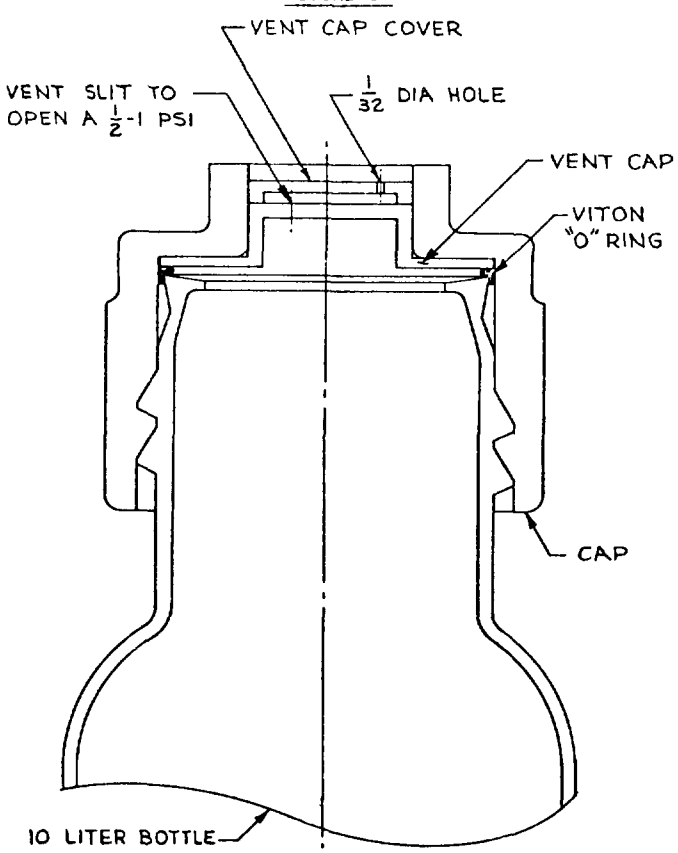
PLUTONIUM NITRATE SHIPPING CONTAINER (3-LITER, 10-LITER)

FIGURE 2



PLUTONIUM SHIPPING CONTAINER
DETAIL OF PRESSURE VESSEL CLOSURE

FIGURE 3



PLUTONIUM SHIPPING CONTAINER
DETAIL OF VENTED CAP, INNER BOTTLE

TABLE 5

COMPARISON OF MPC FOR VARIOUS RADIOACTIVE MATERIALS

	Concentrations, Microcuries/Milliliter*	
	In Air	In Water
Plutonium (239, 240)		
Soluble	6×10^{-14}	5×10^{-6}
Insoluble	1×10^{-12}	3×10^{-5}
Uranium (Natural)		
Soluble	3×10^{-12}	2×10^{-5}
Insoluble	2×10^{-12}	2×10^{-5}
Uranium (235)		
Soluble	2×10^{-11}	3×10^{-5}
Insoluble	4×10^{-12}	3×10^{-5}
Thorium (Natural)		
Soluble	1×10^{-12}	1×10^{-6}
Insoluble	1×10^{-12}	1×10^{-5}
Strontium (90)		
Soluble	3×10^{-11}	3×10^{-7}
Insoluble	2×10^{-10}	4×10^{-5}

* In unrestricted area: 10CFR20, Appendix B, Table II

(Note: The "low as practicable" limits reduce some of these values by factors ranging from 15 to 100; air borne particulates by a factor of 100,000)

TABLE 6

COMPARISON OF LICENSING REQUIREMENTS
FOR PROCESSING FISSILE MATERIAL

	PLUTONIUM	URANIUM
PRELIMINARY APPROVALS REQUIRED		
Site	X	
Design Bases for		
. Plant Structures	X	
. Process Systems and Components	X	
Quality Assurance Program for		
Structures and Components (Must Meet 10CFR50, Appendix B)	X	
SPECIFIC LICENSING REQUIREMENTS		
Resistance of Structures and Components to Effects of Natural Phenomena	X	
Qualified Staff	X	X
Appropriate Equipment	X	X
Nuclear Materials Controls	X	X
Safety Planning and Procedures	X	X
Criticality Analyses	X	X
Nuclear Criticality Alarms	X	X
Emergency Planning	X	X
Double Containment	X	
Shielding	X	

shipping container and the detail of the vent closure on the container bottle.

In all nuclear fuel cycle operations the maximum permissible concentration (MPC) of plutonium and other radioactive materials for release to the environment are set by 10CFR20. A comparison of the MPC values for plutonium, uranium, thorium and strontium-90 are shown in Table 5. From this table it can be seen that the environmental release limits for plutonium are low with respect to other nuclear fuel materials (uranium and thorium), particularly when it is recognized that the specific activity of plutonium is higher than such materials. A comparison of the general AEC licensing requirements for uranium and plutonium fuel processing facilities is shown in Table 6. From this table the more extensive nature of the design requirements and approvals involved in plutonium facilities over those which are necessary for purely uranium processing facilities is demonstrated.

The stringent release limits which must be strictly observed in the operation of nuclear fuel cycle activities; the complex requirements which must be met in the design, construction and licensing of nuclear fuel processing facilities, with respect to the containment and safe handling of plutonium; and the technical and operational experience which has been gained in over 30 years of handling and processing of plutonium have resulted in the nuclear industry having a capability for the safe handling of plutonium now that the need for large scale processing of plutonium has materialized.

What are the Safeguards Problems Associated with Plutonium?

Plutonium can be used to make what most weapons experts refer to as an "unsophisticated" nuclear weapon. This has caused some concern over the possibility of diversion of plutonium from peaceful uses to the production of weapons—by nations who do not have a nuclear weapons capability, by criminal interests, or by extremist groups. Table 7 shows the projected quantities of plutonium which are expected to be produced by nuclear power reactors in the United States and the rest of the free world between now and 1984. As can be seen from this table, significant quantities of plutonium will be produced in future years, representing a large nuclear weapons potential. The United Nations General Assembly recognized this problem and as a result of extensive discussions and deliberations, succeeded in negotiating a non-proliferation treaty which has been ratified by most member nations. This agreement against the further proliferation of nuclear weapons in the world will be enforced by inspections on all nuclear power plants, reprocessing plants, fuel fabrication plants and other nuclear facilities throughout the world by the International Atomic Energy Agency (IAEA) in Vienna, Austria. It is the objective of the IAEA inspections to be able to promptly detect any diversion of nuclear material to unauthorized uses. The United States has long conditioned the supply of nuclear materials to non-nuclear weapons states on bilateral agreements which limit the uses of such materials to peaceful applications, and provide rights of inspection in the receiving country which the United States has exercised. The United States, through the Atomic Energy Commission (AEC), has also established very stringent regulatory controls upon domestic licensees in connection with the handling and processing of nuclear materials in order to minimize the possibility of their diversion to unauthorized activities. These regulatory requirements have been directed toward the prompt

TABLE 7

PROJECTED QUANTITIES OF PLUTONIUM EXPECTED
TO BE PRODUCED BY NUCLEAR POWER REACTORS

YEAR	FISSILE PLUTONIUM RECOVERED PER YEAR (kg) U. S.	REST OF FREE WORLD
1974	2,100	4,500
1976	6,400	6,700
1978	10,800	10,600
1980	15,600	16,200
1982	22,500	23,800
1984	32,300	32,200

Source: WASH-1139

TABLE 8

VALUE OF PLUTONIUM COMPARED WITH PRECIOUS METALS

Uranium (93% Enriched)	$12,000/kg	(373.25/troy oz.)
Plutonium (70% Fissile)	$ 5,200/kg	(161.74/troy oz.)
Platinum	$ 4,180/kg	(130.00/troy oz.)
Gold	$ 1,447/kg	(45.00/troy oz.)
Palladium	$ 1,157/kg	(36.00/troy oz.)
Silver	$ 58/kg	(1.80/troy oz.)

32.15 troy oz. = 1 kg

detection of a diversion of nuclear material in the nuclear fuel cycle through comprehensive nuclear material control procedures within the individual facilities involved. Of course, it should also be recognized that nuclear materials have very high values and that individual commercial organizations involved in nuclear fuel cycle activities have strong economic motivation to protect these materials from loss, pilferage and the like. Table 8 shows the relative value of plutonium and uranium with that of platinum, gold, palladium, and silver—the latter metals being well recognized as precious materials compared to most other items involved in commerce today. From this table it can be seen that plutonium is more valuable than platinum, nearly four times as valuable as gold, more than four times as valuable as palladium, and nearly 90 times as valuable as silver.

In addition to the accounting-type requirements the U.S. Government has more recently imposed strict requirements for the physical protection of nuclear materials, setting minimum standards for physical protection while nuclear materials are in process and being transported.

The problem of diversion of plutonium (or uranium) from a nuclear power plant is extremely remote. Plutonium is present at the reactor in one of three forms:

(i) Plutonium contained in new fuel

(ii) Plutonium contained in fuel which is in the reactor undergoing fission, and

(iii) Plutonium contained in spent fuel.

Plutonium in these forms and under these circumstances is very inaccessible for diversion. Plutonium in new fuel is present to the extent of only about 2 percent, with the remainder of the fuel being uranium. A significant investment in facilities and knowhow would be required to recover this plutonium from diverted reactor fuel in a pure and useful form for weapons production. Plutonium is even more inaccessible in irradiated fuel (either in the reactor or as spent fuel). Not only is the plutonium content of irradiated fuel very low but the presence of highly radioactive fission products requires that, during handling and processing, such fuel be heavily shielded and handled remotely — thus requiring an even higher (factor of 10 or more) investment in facilities and knowhow to effect the recovery of plutonium in a useful form.

The possibilities of diversion of plutonium are greatest in fuel fabrication and spent fuel reprocessing plants, where at some point in the processing plutonium occurs in relatively pure form, and during the transportation of plutonium in pure form. An increasing quantities of plutonium enter the nuclear fuel cycle, more stringent safeguards against prospective diversion are being applied. Extensive material control and accountability techniques, coupled with frequent physical inventories, provide early warning of diversions, and prudent physical security measures make actual diversion to unauthorized uses increasingly difficult to accomplish.

In Summary

Plutonium is a valuable national energy resource, both for the commercial production of electricity and for use as a heat source to power thermoelectric and thermionic generators such as those used in satellites, space missions and heart pacers. With the fossil fuel sources of the world diminishing, plutonium can be expected to play a major role in meeting the increasing demand for energy in the future. The very attributes of plutonium that make it a vital source of energy for the future also make it a potentially dangerous material. But the extensive experience and knowhow in the handling and utilization of this valuable material which has been gained since its discovery in 1940 enables us to exploit its use safely now and in the future.

★ ★ ★

ATLANTA EXECUTIVE MEETING—The INMM Executive Committee met in Atlanta in February for a working meeting. This photo was taken by James W. Lee. Seated (l. to r.)—Walt Martin, Lynn Hurst, Harley Toy, Ralph Jones, Armand Soucy and Fred Forscher. Standing—Tom Gerdis, Joe Shaver, Vince DeVito, Shelley Kops, Manny Kanter, Bob Delaney, Dick Alto and Roy Cardwell.

ISOTOPIC CORRELATION SAFEGUARDS TECHNIQUES: REACTOR CHARACTERISTICS AS OBSERVED FROM MEASURED SPENT FUEL DATA

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ABSTRACT

Families of burnup curves have been determined, based on measured isotopic composition data resulting from the chemical dissolution of spent fuel assemblies, which represent:

a) Various initial ^{235}U content fuels - from natural uranium to 5.0 weight percent ^{235}U, and
b) Various reactor types - heavy water reactors, boiling water reactors and pressurized water reactors - as a function of increasing exposure.

As a result, figures and tables are available which compare burnup properties of various fuels and reactor types. The figures and tables may be used to

a) Verify stated information about future spent fuels from the above reactors processed at a chemical reprocessing plant, such as initial enrichment and exposure;
b) Verify burnup curves which will be evident from future measured data of the reactors considered; and
c) Predict the burnup curves for spent fuels from the above reactors which may be of different initial enrichment.

INTRODUCTION

To date, the development of isotopic correlation safeguards techniques as applied to spent fuel data measured at a chemical reprocessing plant has dealt principally with the verification of the measured plutonium. [1-7] The verification has been accomplished by the use of isotopic ratios involving both the plutonium-to-uranium ratio and ratios of several uranium and plutonium isotopes. The ratios have been observed to be generally independent of increasing exposure for single enrichment fuels. Some attention has also been given to the differences in relative isotopic composition observed from the measured data which were seen to depend on initial enrichment, reactor type, exposure level and other parameters. Brief summaries of these observations have appeared in previous reports. [2,4,6] The purpose of this report is to discuss the differences in relative isotopic composition in more detail and to indicate differences that tend to characterize the initial fuel enrichment and reactor type.

The term characterize is used in the sense that the relative composition of the measured isotopic data tends to vary uniquely between initial enrichments and the type or class or reactor in which the nuclear fuels are irradiated.

The available data base of chemical reprocessing measurements assembled at Battelle-Northwest now includes data representing heavy water reactors (HWRs), boiling water reactors (BWRs) and pressurized water reactors (PWRs), with fuels ranging from natural uranium to 4.94 wt% ^{235}U enriched. Because relative isotopic composition of nuclear fuels is a useful index, this report also includes graphite moderated-gas cooled reactor (GCR) and plutonium recycle data. These latter data resulted from burnup experiments rather than from the dissolution of spent assemblies and serve to indicate what can be expected as chemical reprocessing data are made available for these fuels.

The measured isotopic data have shown remarkable consistency and have been used to obtain the position and shape of the reactor fuel burnup paths for all isotopes and to identify the variables which affect changes in the burnup paths. The consistency of the data has been a basic building block permitting fuels and reactor types to be characterized. In addition, it has been demonstrated that families of burnup curves based on the measured isotopic data can be determined empirically as a function of initial enrichment, reactor type and increasing exposure by extrapolation and interpolation. Calculated burnup results were used in some cases to aid in defining the shape of the burnup curves but not the positions of the curves. The empirical burnup curves apply within the limitations of enrichment and reactor type represented by the available data.

As a result figures and tables are presented which compare isotopic data from GCRs, HWRs, BWRs and PWRs. From the comparisons, identifying characteristics are indicated particularly for BWR and PWR data and their applications to isotopic correlation safeguards techniques are discussed.

DISCUSSION

There is more information in measured data from chemical reprocessing plants than just the total amounts of uranium and plutonium and their fissile contents. A key to extracting as much information as possible is the underlying transmutation equations which are germane to the isotopic ratios developed thus far. Just as equations can be manipulated to provide information, so can the measured data be manipulated and information extracted by trends evident from data manipulation. The transmutation equations have not been used explicitly, but they are central to the development of identifying characteristics using measured isotopic data.

Data Base

A key feature of any identification process is the central data or information file. Use of characteristics for identification is only as good as the file from which they are derived. A substantial measured data base is now available, and efforts are continuing to collect both newly measured data and data measured in the past but not collected.

The amount of data collected to date is shown in Parts A and B of Table I. Chemical reprocessing plant data include measured isotopic data, that is ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu, for input dissolution batches of fuel from Candu, [4] Dresden I, [6,8,9]

TABLE I
MEASURED DATA COLLECTED
PART A

REACTOR TYPE	NO. BATCHES	DATA SET	ENRICHMENT
HWR	9	CANDU	0.7114
GMR	–	CALDER HALL*	0.712
BWR	17	DRESDEN I, Fuel Lot 1	1.474
BWR	17	DRESDEN I, Fuel Lot 2	1.474
BWR	12	DRESDEN I, Fuel Lot 4	1.474
BWR	9	DRESDEN I, Fuel Lot 3	2.50
BWR	11	HUMBOLDT BAY	2.578
PWR	9	TRINO	2.72
PWR	16	YANKEE ROWE CORE I	3.404
PWR	14	YANKEE ROWE CORE II & III	3.404
PWR	7	YANKEE ROWE CORE IV	4.101
PWR	11	YANKEE ROWE CORE V	4.101
PWR	11	YANKEE ROWE CORE VI	4.935
PWR	12	YANKEE ROWE CORE VII	4.935
PWR	11	YANKEE ROWE CORE VIII	4.941
PWR	–	SAXTON (Pu recycle)*	0.712 (U) 6.6 (Pu)

***Measured data have resulted from a burnup experiment**

TABLE I (continued)
MEASURED DATA COLLECTED
PART B

REACTOR TYPE	NO. BATCHES	DATA SET	ENRICHMENT
BWR	1	DRESDEN I, Fuel Lot 4	1.59
BWR	1	DRESDEN I, Fulel Lot 4	1.60
BWR	1	DRESDEN I, Fuel Lot 4	1.62
BWR	1	DRESDEN I, Fuel Lot 4	1.77
BWR	1	DRESDEN I, Fuel Lot 4	1.83
BWR	1	DRESDEN I, Fuel Lot 4	2.18
BWR	1	HUMBOLDT BAY	2.258
BWR	1	HUMBOLDT BAY	2.276
BWR	3	HUMBOLDT BAY	2.310
BWR	2	VAK	2.33
BWR	1	VAK	2.40
BWR	1	HUMBOLDT BAY	2.402
BWR	1	HUMBOLDT BAY	2.433
BWR	1	VAK	2.51
BWR	1	HUMBOLDT BAY	2.557
BWR	1	HUMBOLDT BAY	2.567
PWR	1	YANKEE ROWE CORE V	2.90
PWR	1	TRINO	2.92
PWR	3	TRINO	3.13
PWR	1	TRINO	3.31
PWR	1	YANKEE ROWE CORE VII	3.50
PWR	1	YANKEE ROWE CORE VII	3.59
PWR	4	YANKEE ROWE CORE IV	3.90

Humboldt Bay, [7] VAK, [4] Trino [4,10] and Yankee Rowe[1,2,3,5,6,8,11,12] reactors. The enrichments of the initial fuels varied from natural uranium to 4.94 wt% ^{235}U enriched. Part A of Table I lists those data sets where enough fuel of a given initial enrichment was processed to result in at least seven measured batches. Several of the batches listed in Part B resulted from mixing fuel assemblies of different initial enrichments in a given batch. This was the case for the single batches from Dresden 1, fuel lot 4, the single batches from the Humboldt Bay and the four batches from Yankee Rowe Core IV. Part B of Table I also lists those data sets which had less than seven batches of single enrichment fuels. This was the case for the remaining three batches of Humboldt Bay as well as the batches of VAK, Trino and Yankee Rowe Cores V and VII fuels which are listed.

The measured data collected for Calder Hall[13] and Saxton[14] reactors resulted from burnup experiments rather than dissolution of spent fuel assemblies. Least-squares fitting results, in the case of Saxton, and calculated results which matched the measured data, in the case of Calder Hall, are used for ease in presentation.

A large amount of independently measured data has also been collected from the dissolution of Candu, Dresden I, Humboldt Bay, VAK and Yankee Rowe spent fuels. These data have served to reinforce the conclusion derived from the data measured at the chemical reprocessing plants.

Reactors for which partial sets of measured data have been collected include Douglas Point I,[15] Sena[15] and Big Rock Point[16] reactors. Complete data sets are expected to be collected in the future.

In addition to the measured data, calculated results have been collected from calculations of Candu, Calder Hall, Dresden I, Humboldt Bay, Big Rock Point, KRB,[15] Browns Ferry,[17] Trino,[15] Fort Calhoun,[17] Yankee Rowe and Saxton reactors. Some of the calculated data are incomplete as far as the isotopic data are concerned and application of these results were limited, but nevertheless have proved useful.

All measured data listed in Table I and mentioned in this section have been provided through cooperation and agreements with the fuel owners and the reprocessors. The calculated data have been provided by the reactor operators except for Browns Ferry and Fort Calhoun. The receipt of all data is gratefully acknowledged.

Comparison of Plotted Data

The data described above have been used to compare the isotopic burnup properties of operating HWR, BWR, and PWR reactor types as well as to look at GCR and plutonium recycle data and note how isotopic correlations can apply to these types. Presenting the collective data in various plotted formats makes it possible to visually compare the data. Information is derived pertaining to data consistency from batch to batch and from reactor to reactor, burnup path shape and position, and identification of trends that may be present in the isotopic data. Each figure also serves as an information data bank to which new set of measurements can be compared.

The ^{236}U, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu data are shown as a function of ^{235}U in Figures 1 through 5. In all figures the initial ^{235}U values (and the initial plutonium values for the Saxton plutonium recycle fuel) are shown. In Figure 1, an initial ^{236}U value is also shown, except for natural uranium, and the ^{236}U values are based on isotopic measurements of several of the initial fuels. Since ^{236}U does not occur naturally, it was presumed that its presence was from uranium irradiated in a reactor and recycled through an enrichment plant. A small amount of ^{236}U was therefore assumed to be present in all enriched ^{235}U fuels. The values were obtained from a plot of initial ^{236}U versus initial ^{235}U. All the isotopic value units are expressed as weight percent per total uranium for ^{235}U and ^{236}U and per total plutonium for ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu.

Each data point shown in Figures 1 through 5, other than those representing initial values, represent measured isotopic values of individual batches of the dissolved fuels listed in Table I. Those that appear as groups and which lie near the curves shown represent the major data sets listed in Part A of Table I. Those data points that appear scattered about the graphs represent the mixed initial enrichment fuel batches and small sets of single enrichment fuel batches listed in Part B of Table I. Further information pertaining to the data points is given in Appendix A.

As can be seen by examination of Figures 1 through 5, all the measured data from spent fuels irradiated in the various reactors listed in Table I show that

1). Definition relationships exist between the uranium and plutonium isotopes for HWR, GCR, BWR, PWR and plutonium recycle data.

2). The relationships are well defined, making it possible to indicate the burnup paths by inspection. These curves have been drawn by connecting the measured batch data points with the initial (or zero burnup) data point for the major sets of data listed in Part A of Table

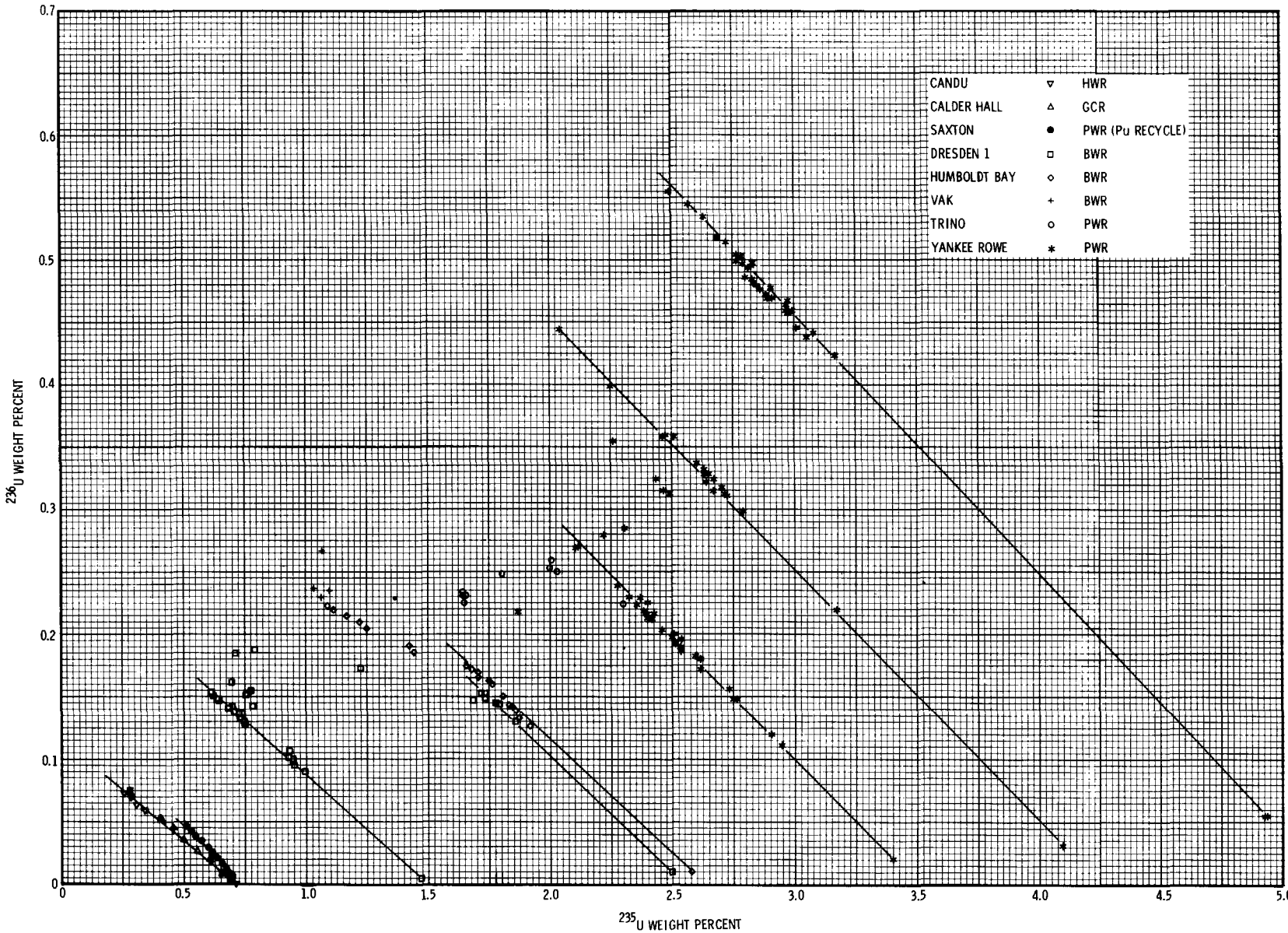

FIGURE 1. URANIUM-236 WEIGHT PERCENT AS A FUNCTION OF ^{235}U WEIGHT PERCENT

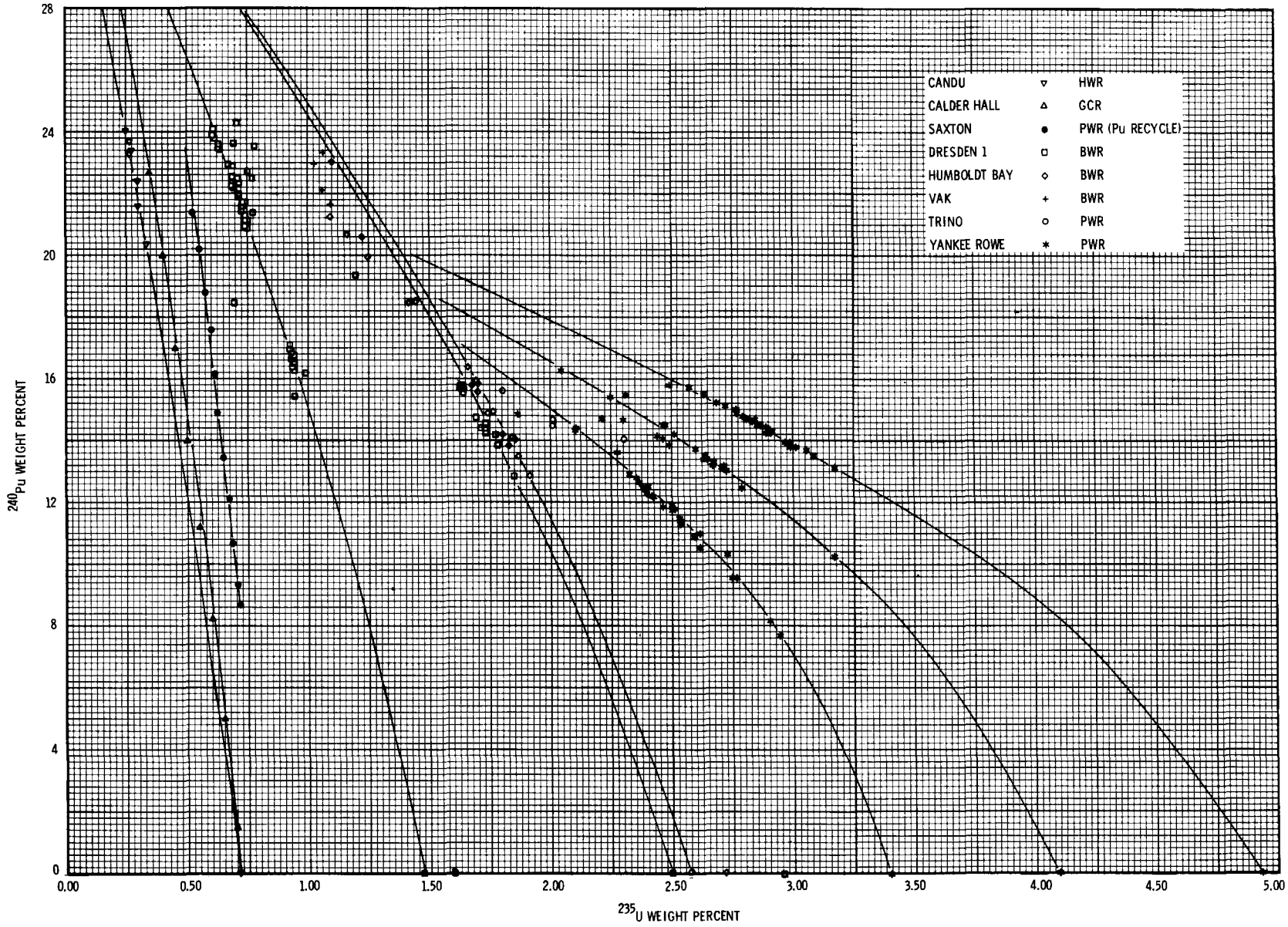

FIGURE 3. PLUTONIUM-240 WEIGHT PERCENT AS A FUNCTION OF ^{235}U WEIGHT PERCENT

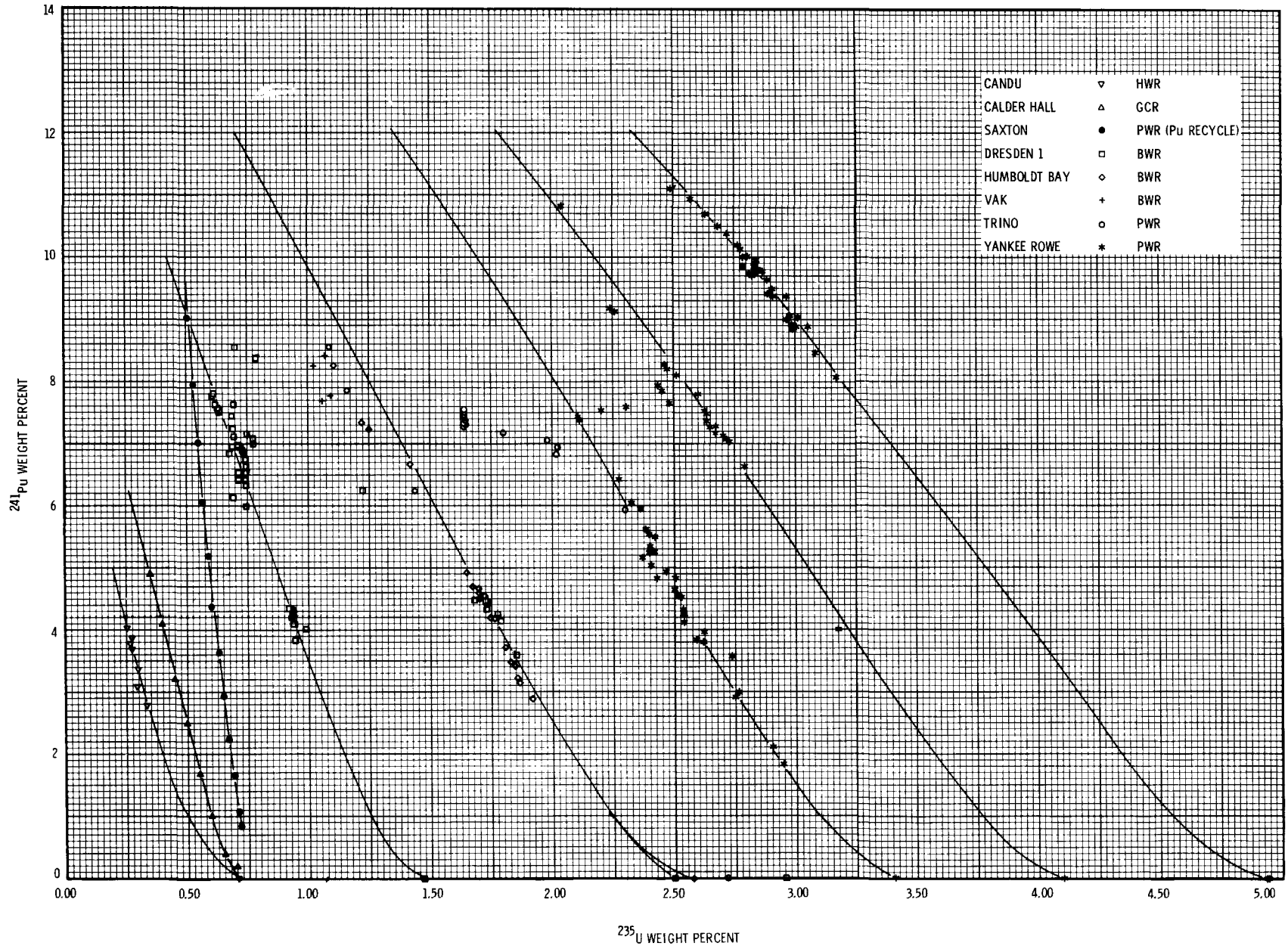

FIGURE 4. PLUTONIUM-241 WEIGHT PERCENT AS A FUNCTION OF ^{235}U WEIGHT PERCENT

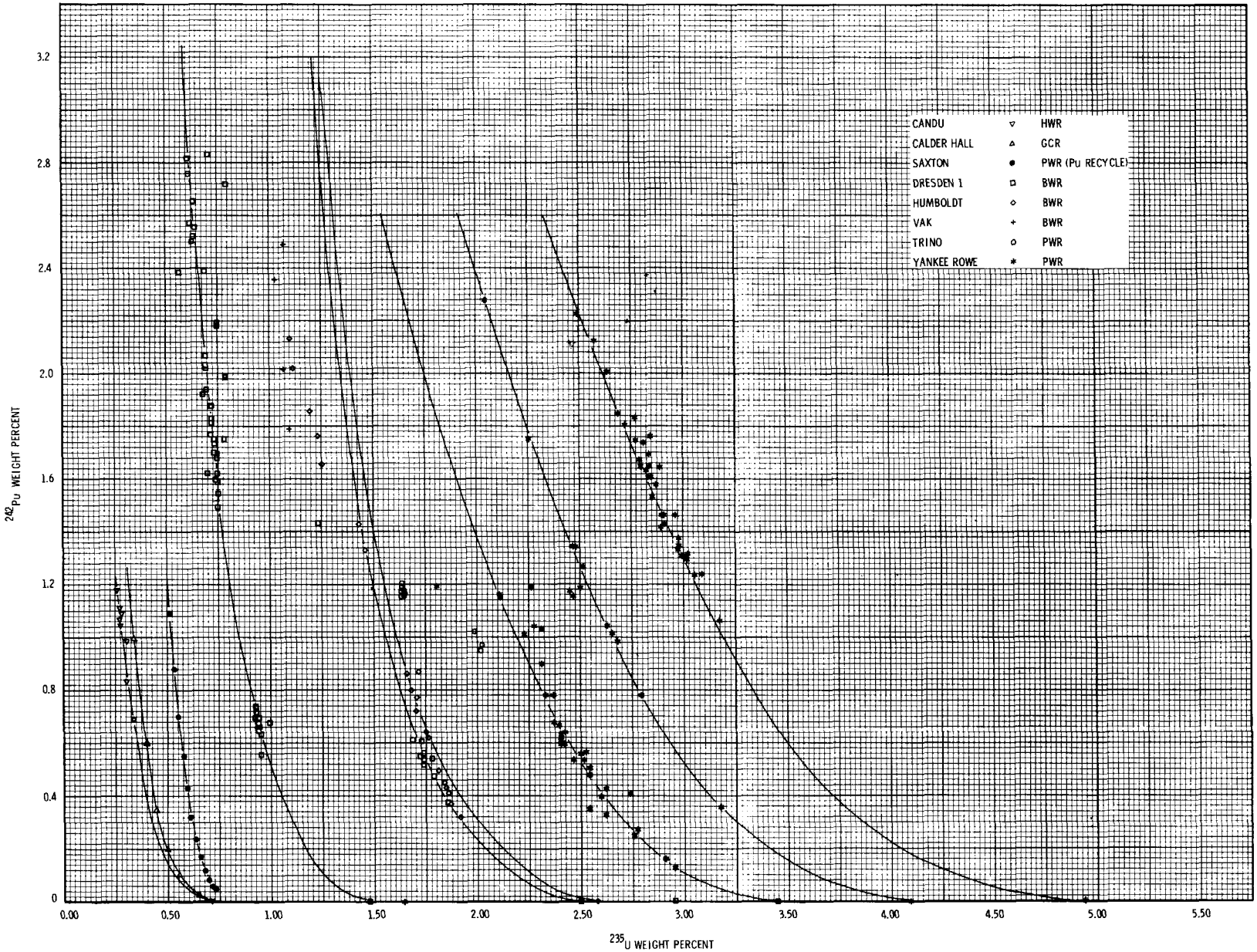

FIGURE 5. PLUTONIUM-242 WEIGHT PERCENT AS A FUNCTION OF ^{235}U WEIGHT PERCENT

I, except trino. The shapes of the burnup curves in regions where no measured data were available were verified by comparing calculated burnup curves with the curves drawn by inspection. Good agreement between the shapes of the burnup curves shown in Figures 1 through 5 and calculated burnup curves exists. The position of the burnup curves shown, however, was dictated by the measured data rather than the calculated data.

By comparing the shapes and positions of the burnup curves, indentification trends were noted. However, these characteristics are discussed after the isotopic tables are presented.

Isotopic Data Tables

To compare isotopic values for several enrichments as a function of equal increments of exposure, the measured data shown in Figures 1 through 5 were used to empirically determine families of burnup curves. The curves were then used to compile tables of isotopic values, Table II-A through F representing the reactors listed in Table I.

The empirical burnup curves (dashed lines) at initial ^{235}U enrichments of 1.5, 2.0, 2.5, 3.0 wt% for the BWR data and 3.0, 3.5, 4.0 4.5 and 5.0 wt% for the PWR data are shown in Figure 6 for the case of ^{236}U versus ^{235}U as an example. These enrichments apply to BWRs and PWRs particularly rather than HWRs. Additional data are needed from HWRs or GCRs representing initial enrichments from natural uranium to 1.5 wt% ^{235}U in order to define intermediate burnup curves in this range.

Empirical burnup curves for the plutonium isotopes considered in Figures 2 through 5 were also determined. For the ^{235}U values listed in Table II-A, the corresponding ^{236}U, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu values were read from the empirical burnup curves and tabulated in Table II-B, C, D, E and F, respectively.

The determination of values shown in Tables II-B through F was based on ^{235}U values rather than the exposure values given in Table II. Generally speaking, there exist no measured values of exposure from the dissolution of spent fuels at a chemical reprocessing plant. The exposure values used were calculated values which appeared on material transfer sheets or were received directly from utilities. The exposure values were plotted as a function of ^{235}U, and empirical burnup curves (see Figure 7) were determined by the same procedures used to determine the isotopic curves. The ^{235}U values tabulated in Table II-A were then determined at the exposure levels listed in the table. Although the exposure values are not considered as accurate as the ^{235}U and the corresponding ^{236}U, etc, isotopic values, the exposure values are sufficiently accurate in order to be able to compare isotopic values as a function of increasing exposure for the initial enrichment fuels listed.

The general procedures used to empirically determine the dashed burnup curves by interpolation and extrapolation are discussed in Appendix A. In addition to the general procedure outlined in Appendix A which constrained the empirical burnup curves to be very similar to the burnup curves representing the measured data, several other constraints were used both from the measured data and from calculated data to ensure that the values given in Table II were representative of the reactors listed in Table I. As mentioned, calculated burnup curve shapes were used and empirical burnup curve shapes were required to show agreement with calculated shapes. Several cross checks were used as well. In addition to the internal agreement between ^{239}Pu and ^{235}U, for example, internal agreement between ^{239}Pu versus Pu/U and ^{235}U versus Pu/U was required. For the higher plutonium isotopes, consistent agreement of ^{240}Pu versus ^{239}Pu, ^{241}Pu versus ^{239}Pu and ^{240}Pu, ^{242}Pu versus ^{239}Pu, ^{240}Pu and ^{241}Pu was also expected. A last constraint used was the requirement that the plutonium isotopic weight percents would add to one hundred minus the ^{238}Pu wt% (100 − ^{238}Pu wt%).

IDENTIFYING CHARACTERISTICS

The isotopic values given in Table II are based on reactor spent fuel data from Candu, an HWR, from Dresden I and Humboldt Bay, both BWRs and from Yankee Rowe and Trino, both PWRs. The isotopic values from the GCR and plutonium recycle data are also listed in Table II and these values were obtained directly from the respective burnup curves shown in Figures 1 through 5. The values of exposure that apply to reprocessed spent fuels are from 5,000 to 30,000 MWD/MTM. However, isotopic values at 1,000, 2,000 and 3,000 MWD/MTM are also listed to define the regions of low exposure which are needed to define the burnup curves. Since ^{235}U is not the only isotope with initial values, the initial isotopic values, other than ^{235}U, are listed in the row of zero exposure in Table II.

That neutron spectrum differences in these reactor types result in different relative isotopic compositions can be seen from Figures 1 through 6 and from Table II-A through F. Differences that characterize reactor type are summarized by isotope in the subsections that follow.

Uranium 235

The ^{235}U indicates initial enrichment and to designates possible reactor types which utilize fuel in a given enrichment range. For example, a measured ^{235}U value of 2.4 wt% representing a batch of dissolved fuel indicates that HWR or BWR fuels initially enriched to less than 2.4 wt% ^{235}U are not involved, while BWR or PWR fuels initially enriched to values above 2.4 wt% ^{235}U are involved. The measured ^{235}U was the basic parameter that was used throughout to determine both the empirical burnup curves and the tabulated isotopic values.

From the values given in Table II-A, initially equal ^{235}U content fuels in different reactor types, that is, natural uranium in both a HWR or GCR and 3.0 wt% ^{235}U in both a BWR or a PWR, utilize different amounts of ^{235}U to reach the same exposure levels. The HWR and BWRs used more ^{235}U than the GCR or PWRs, respectively, and therefore varying amounts of plutonium are produced and fissioned in the different reactor types when enriched to the same level.

The ^{235}U values are also an index to indicate exposure levels. The values tabulated in Table II-A indicate that to reach an exposure level of 30,000 MWD/MTM, a fuel which is to be charged in reactors of similar designs must be enriched to more than 2.5 wt% ^{235}U. The initial enrichments decrease that much in the case of the 4.0, 4.5 and 5.0 wt% ^{235}U fuels. All ^{235}U enriched fuels decreased an approximate equivalent enrichment of 1 wt% ^{235}U to reach the 10,000 MWD/MTM exposure level. Thus, natural uranium fuels operate within the 10,000 MWD/MTM exposure range and at the same time are required to utilize a larger amount of plutonium fissions to even acquire exposures over 5,000 MWD/MTM. The other alternative is to enrich with plutonium as was the Saxton fuel (6.6 wt% Pu) in order to reach high burnups such as 30,000 MWD/MTM in which the contribution from ^{235}U to total exposure can be estimated to be less than 3,000 MWD/MTM from Table II-A.

The measured ^{235}U content (in wt%) of spent fuels is thus seen to be a general indicator of the initial enrichment of the fuel, of possible reactor types which burn fuels of that enrichment range and of the exposure level that might be expected from the fuel since the higher the enrichment the higher the expected exposure whether enriched with ^{235}U or plutonium.

Uranium 236

The ^{235}U values, as seen in Table II-B, offer little more in the way of characterizing reactor type than already realized from ^{235}U. The ^{236}U is produced from ^{235}U and thus depends on initial ^{235}U enrichment or amount of ^{235}U, the neutron capture cross section of ^{235}U and the exposure level. The ^{235}U capture is more commonly included in a^{235}, the capture-to-fission ratio of ^{235}U, and as the initial enrichment increases, the increasing ^{236}U values indicate that a^{235} increases. Thus, more ^{236}U is produced in higher enriched fuels for the same amount of ^{235}U burned.

However, using the ^{235}U, ^{236}U and ^{238}Pu values and the procedures to empirically determine burnup curves, a graphical method has resulted, see Figure 8, whereby the initial enrichment of spent fuels can be identified to within ± 3%. This result was obtained by observing the measured data in many plotting formats until one format was noted which was apparently independent of the various reactor design parameters except the ^{235}U initial enrichment. The burnup paths at intermediate enrichments to those represented by the measured data were also determined, resulting in a family of burnup curves, each curve representing a given initial ^{235}U enrichment as seen in Figure 8.

TABLE II

A

SPENT FUEL ISOTOPIC CONTENT BASED ON MEASURED DATA

ISOTOPE	^{235}U Weight Percent											
REACTOR TYPE	HWR	BWR				PWR					Pu RECYCLE (SAXTON-PWR)	GCR
INITIAL ENRICHMENT	NAT.	1.500	2.000	2.500	3.000	3.000	3.500	4.000	4.500	5.000	NAT.-6.6	NAT.
EXPOSURE (MWD/MTM)												
0	0.712	1.500	2.000	2.500	3.000	3.000	3.500	4.000	4.500	5.000	0.720	0.720
1000	0.603	1.380	1.870	2.372	2.873	2.890	3.387	3.890	4.390	4.890	0.715	0.616
2000	0.522	1.270	1.752	2.250	2.750	2.777	3.275	3.776	4.275	4.776	0.708	0.534
3000	0.440	1.162	1.647	2.137	2.642	2.677	3.173	3.675	4.177	4.672	0.695	0.464
5000	0.315	0.987	1.454	1.937	2.432	2.490	2.975	3.462	3.955	4.450	0.680	0.356
10000	0.126	0.648	1.065	1.512	1.975	2.080	2.540	3.002	3.475	3.953	0.644	
15000		0.400	0.745	1.143	1.575	1.727	2.167	2.602	3.050	3.500	0.605	
20000			0.503	0.850	1.250	1.427	1.840	2.254	2.672	3.100	0.567	
25000				0.612	0.955	1.172	1.554	1.942	2.327	2.725	0.530	
30000								1.665	2.026	2.398	0.495	

TABLE II (Cont'd)

B

ISOTOPE	^{236}U Weight Percent											
REACTOR TYPE	HWR	BWR				PWR					Pu RECYCLE (SAXTON-PWR)	GCR
INITIAL ENRICHMENT	NAT.	1.500	2.000	2.500	3.000	3.000	3.500	4.000	4.500	5.000	NAT.-6.6	NAT.
EXPOSURE (MWD/MTM)												
0	0.000	0.005	0.008	0.012	0.015	0.015	0.021	0.030	0.042	0.059	0.000	0.000
1000	0.017	0.026	0.033	0.035	0.038	0.036	0.043	0.053	0.064	0.082	0.002	0.017
2000	0.030	0.046	0.053	0.057	0.063	0.057	0.066	0.076	0.088	0.106	0.004	0.031
3000	0.043	0.064	0.072	0.077	0.082	0.076	0.086	0.096	0.109	0.128	0.006	0.042
5000	0.063	0.095	0.106	0.113	0.122	0.112	0.124	0.138	0.155	0.174	0.009	0.060
10000	0.096	0.155	0.176	0.192	0.211	0.190	0.218	0.231	0.253	0.277	0.017	
15000		0.198	0.233	0.258	0.287	0.258	0.284	0.313	0.340	0.372	0.027	
20000			0.275	0.312	0.348	0.315	0.348	0.382	0.418	0.455	0.035	
25000					0.406	0.364	0.406	0.444	0.489	0.532	0.043	
30000								0.501	0.551	0.601	0.050	

TABLE II (Cont'd)

C

ISOTOPE	^{239}Pu Weight Percent											
REACTOR TYPE	HWR	BWR				PWR					Pu RECYCLE (SAXTON-PWR)	GCR
INITIAL ENRICHMENT	NAT.	1.500	2.000	2.500	3.000	3.000	3.500	4.000	4.500	5.000	NAT.-6.6	NAT.
EXPOSURE (MWD/MTM)												
0	100.00*	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	90.41	100.00
1000	93.17	95.07	95.80	96.60	97.15	96.88	97.37	97.89	98.24	98.49	89.85	92.10
2000	88.05	90.81	92.02	93.37	94.44	94.15	95.00	95.75	96.34	96.80	89.09	85.90
3000	82.78	86.34	88.76	90.36	91.92	91.85	92.98	93.97	94.79	95.26	88.16	80.40
5000	74.39	79.48	82.76	85.31	87.14	88.07	89.35	90.37	91.30	92.16	86.40	71.80
10000	60.84	65.58	70.76	74.60	77.65	80.59	82.26	83.45	84.60	85.83	83.00	
15000		54.30	59.63	64.55	68.60	74.69	76.45	77.97	79.18	80.30	78.61	
20000				56.60	61.20	69.74	71.78	73.20	74.48	75.60	74.00	
25000						65.43	67.60	69.23	70.24	71.34	69.70	
30000								65.60	66.72	67.68	65.60	

*In terms of weight percents, initially ^{239}Pu is 100.00 wt % of the first atom of plutonium formed.

TABLE II (Cont'd)

D

ISOTOPE	^{240}Pu Weight Percent											
REACTOR TYPE	HWR	BWR				PWR					Pu RECYCLE (SAXTON-PWR)	GCR
INITIAL ENRICHMENT	NAT.	1.500	2.000	2.500	3.000	3.000	3.500	4.000	4.500	5.000	NAT.-6.6	NAT.
EXPOSURE (MWD/MTM)												
0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	8.66	0.00
1000	6.50	4.50	3.78	3.00	2.48	2.83	2.40	1.90	1.56	1.32	9.10	7.15
2000	11.00	8.07	6.88	5.60	4.65	5.08	4.37	3.60	3.07	2.60	9.65	12.30
3000	15.40	11.30	9.20	7.85	6.55	6.80	5.85	4.93	4.20	3.78	10.26	16.35
5000	21.60	15.92	13.30	11.40	9.90	9.35	8.23	7.32	6.50	5.80	11.50	22.50
10000	29.58	23.68	20.45	17.92	15.95	13.20	11.90	10.98	10.15	9.38	13.60	
15000		28.84	25.38	22.90	20.60	15.58	14.35	13.37	12.55	11.82	16.53	
20000			29.03	26.60	24.10	17.32	16.10	15.16	14.36	13.60	19.10	
25000				29.30	27.18	18.68	17.50	16.50	15.85	15.18	21.54	
30000								17.80	17.16	16.50	23.58	

TABLE II (Cont'd)

E

ISOTOPE												
REACTOR TYPE	HWR	BWR				PWR					Pu RECYCLE (SAXTON-PWR)	GCR
INITIAL ENRICHMENT	NAT.	1.500	2.000	2.500	3.000	3.000	3.500	4.000	4.500	5.000	NAT.-6.6	NAT.
EXPOSURE (MWD/MTM)												
0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.88	0.00
1000	0.29	0.40	0.39	0.37	0.35	0.27	0.21	0.20	0.19	0.18	1.00	0.71
2000	0.81	1.00	1.00	0.92	0.86	0.67	0.58	0.56	0.52	0.48	1.20	1.78
3000	1.52	2.08	1.83	1.61	1.40	1.16	1.01	0.95	0.88	0.82	1.50	2.95
5000	3.10	3.91	3.44	2.90	2.60	2.23	2.12	2.02	1.94	1.77	2.00	4.77
10000	6.47	7.72	6.87	6.05	5.35	5.14	4.92	4.79	4.60	4.32	3.21	
15000		10.47	9.67	8.78	7.95	7.70	7.38	7.19	6.97	6.73	4.52	
20000			11.85	11.00	10.00	9.68	9.32	9.20	9.00	8.75	6.34	
25000				12.75	11.87	11.22	11.02	10.82	10.70	10.52	7.90	
30000								12.20	12.06	11.92	9.60	

TABLE II (Cont'd)

F

ISOTOPE	^{242}Pu Weight Percent											
REACTOR TYPE	HWR	BWR				PWR					Pu RECYCLE (SAXTON-PWR)	GCR
INITIAL ENRICHMENT	Nat.	1.500	2.000	2.500	3.000	3.000	3.500	4.000	4.500	5.000	NAT.-6.6	NAT.
EXPOSURE (MWD/MTM)												
0	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.046	0.000
1000	0.035	0.022	0.020	0.018	0.015	0.013	0.010	0.005	0.003	0.002	0.048	0.040
2000	0.113	0.081	0.065	0.058	0.045	0.040	0.030	0.024	0.021	0.020	0.060	0.120
3000	0.265	0.222	0.152	0.118	0.085	0.080	0.060	0.048	0.037	0.036	0.080	0.300
5000	0.790	0.582	0.390	0.270	0.208	0.202	0.151	0.119	0.105	0.090	0.100	0.930
10000	2.915	2.680	1.650	1.100	0.784	0.701	0.548	0.439	0.351	0.280	0.189	
15000		5.30	4.32	3.040	2.08	1.416	1.157	0.995	0.798	0.664	0.340	
20000				4.900	3.810	2.300	1.905	1.632	1.415	1.234	0.560	
25000						3.395	2.740	2.377	2.109	1.895	0.860	
30000								3.161	2.827	2.550	1.220	

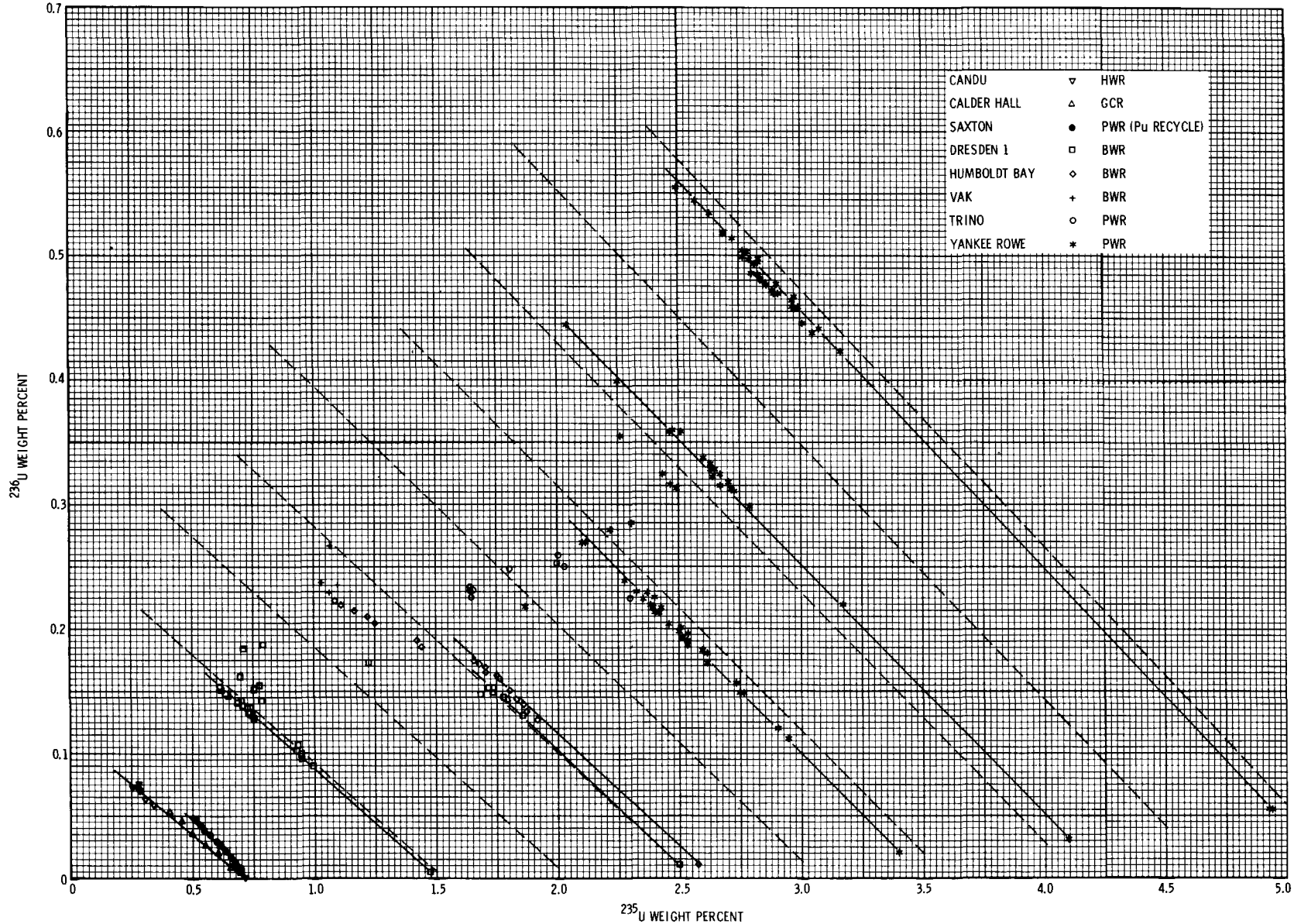

FIGURE 6. URANIUM-236 WEIGHT PERCENT AS A FUNCTION OF ^{235}U WEIGHT PERCENT

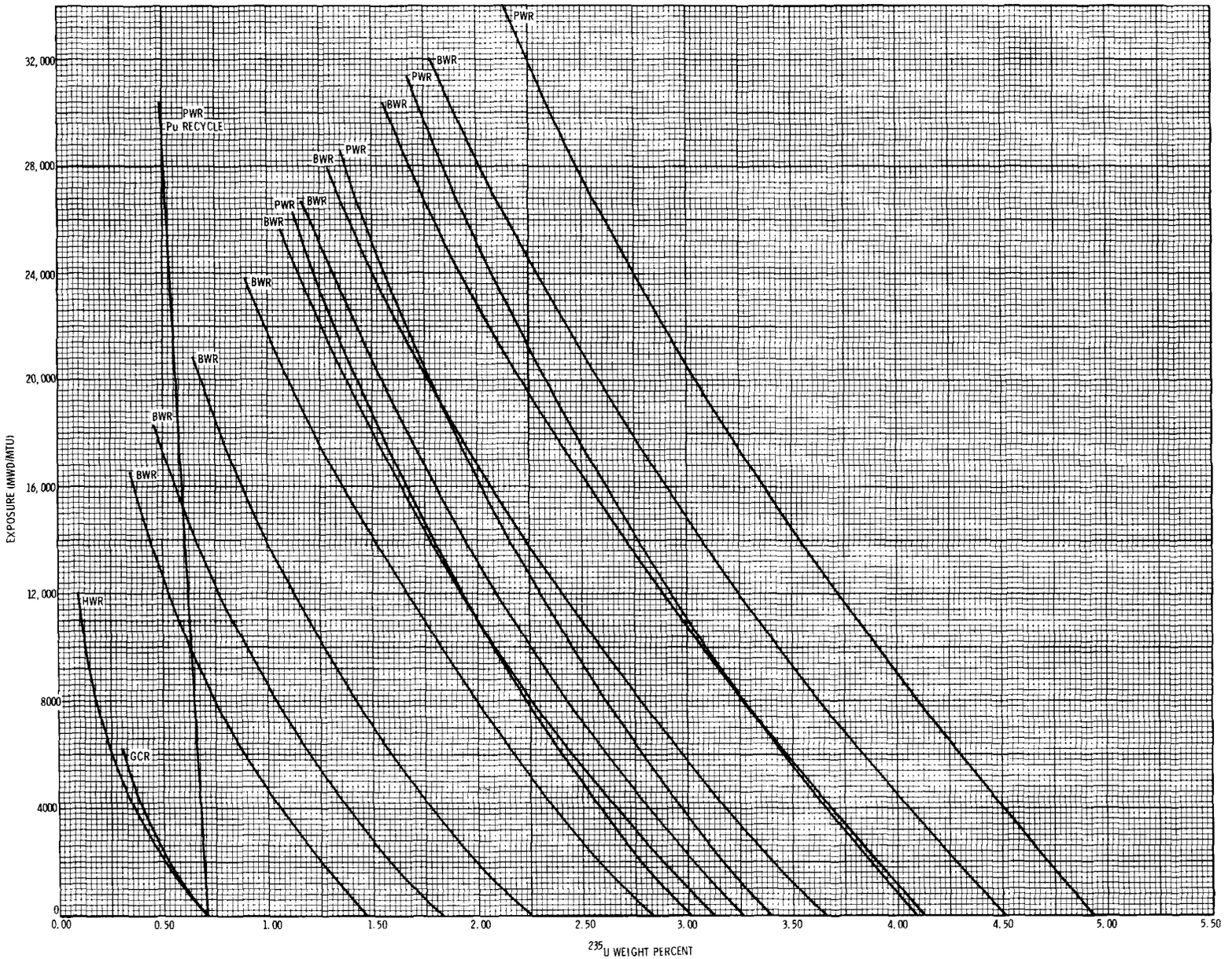

FIGURE 7. EXPOSURE AS A FUNCTION OF ^{235}U WEIGHT PERCENT

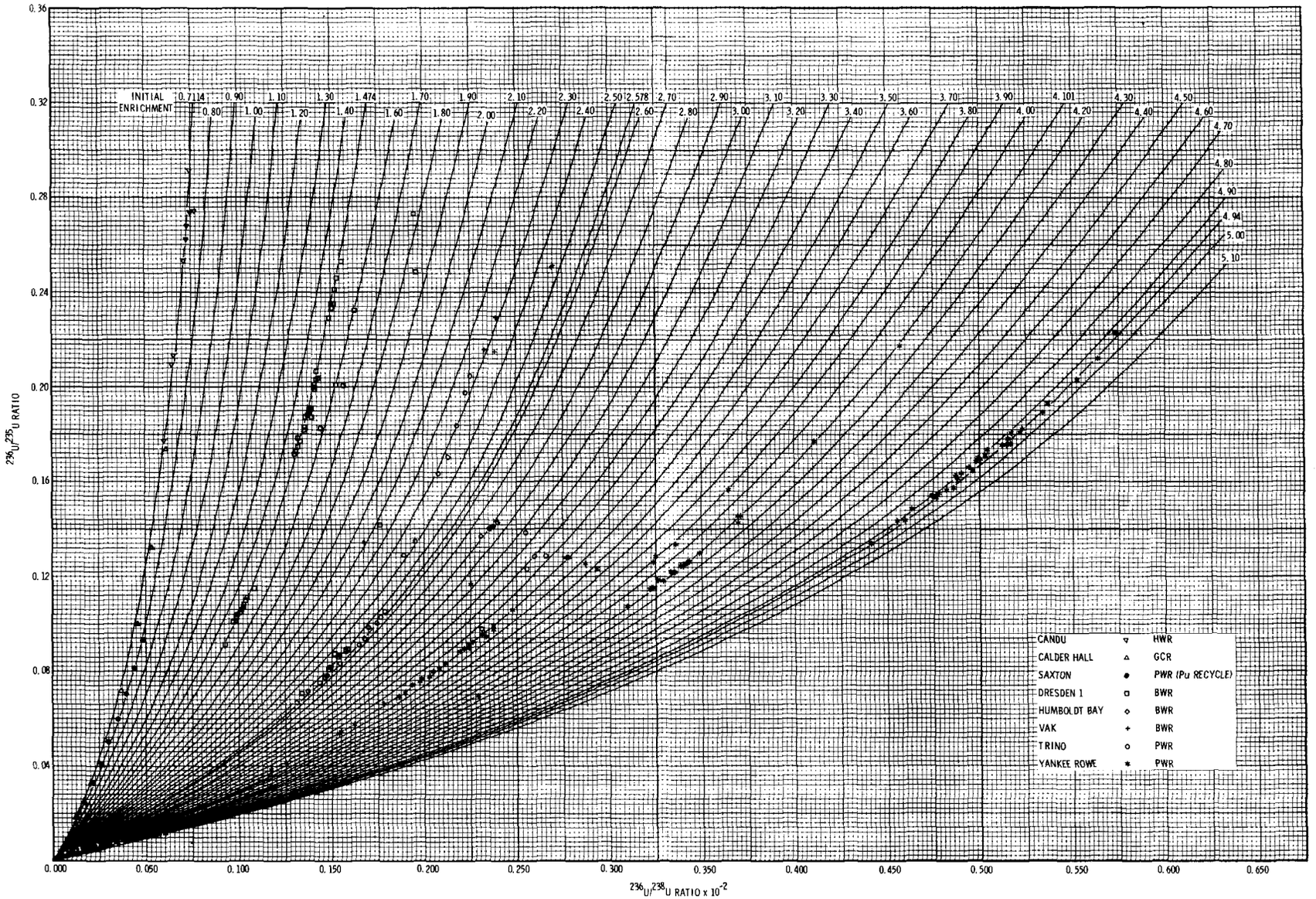

FIGURE 8. THE $^{236}U/^{235}U$ RATIO AS A FUNCTION OF THE $^{236}U/^{238}U$ RATIO

TABLE III
COMPARISON OF INITIAL ^{235}U ENRICHMENT FROM FUEL RECORDS AND AS DETERMINED FROM FIGURE 8

NO. OF BATCHES	DATA SET	INITIAL ENRICHMENT FROM RECORDS Wt %	INITIAL ENRICHMENT FROM FIGURE 8 Wt %	Diff. (%)
1	DRESDEN I, Fuel Lot 4	1.59	1.60	+0.6
1	DRESDEN I, Fuel Lot 4	1.60	1.60	0.0
1	DRESDEN I, Fuel Lot 4	1.62	1.64	+1.2
1	DRESDEN I, Fuel Lot 4	1.77	1.78	−0.6
1	DRESDEN I, Fuel Lot 4	1.83	1.85	+1.1
1	DRESDEN I, Fuel Lot 4	2.18	2.13	−2.3
1	HUMBOLDT BAY	2.26	2.27	+0.4
1	HUMBOLDT BAY	2.28	2.27	−0.4
3	HUMBOLDT BAY	2.310	2.30	−0.4
2	VAK	2.33	2.28	−2.1
1	VAK	2.40	2.32	−3.3
1	HUMBOLDT BAY	2.40	2.39	−0.4
1	HUMBOLDT BAY	2.43	2.42	−0.4
9	DRESDEN I, Fuel Lot 3*	2.50	2.50	−0.0
1	VAK	2.51	2.46	−2.0
1	HUMBOLDT BAY	2.56	2.56	0.0
1	HUMBOLDT BAY	2.57	2.57	0.0
9	TRINO*	2.72	2.82	+3.7
1	YANKEE ROWE CORE V	2.90	2.97	+2.4
1	TRINO	2.92	3.03	+3.8
3	TRINO	3.13	3.22	+2.9
1	TRINO	3.31	3.34	+0.9
1	YANKEE ROWE CORE VII	3.47	3.53	+1.7
1	YANKEE ROWE CORE VII	3.53	3.64	+3.1
4	YANKEE ROWE CORE IV	3.90	3.91	+0.3

***Major data set.**

To use Figure 8 with new measured data from the reactors listed in Table I, $^{236}U/^{235}U$ and $^{236}U/^{238}U$ ratios are formed and the point representing the values is plotted on Figure 8. The point will plot on or near a burnup curve which represents a given initial enrichment. The measured ^{235}U, ^{236}U and ^{238}U values are then identified as having resulted from irradiated fuel initially enriched to a value consistent with the burnup path upon which the plotted point falls.

The method was developed using the major sets of data listed in Part A of Table I (except Trino and Dresden I, fuel lot 3) and has been tested using the data listed in Part B, Table I plus Dresden I, fuel lot 3 and Trino data of Part A, Table I. That is, the initial ^{235}U enrichments which result from plotting the measured ^{235}U, ^{236}U and ^{238}U data for batches listed in Part B were determined and are listed in Table II.

The agreement between the ^{235}U initial enrichment as obtained from fuel records and as determined from Figure 8 is generally to within 3%. This agreement is considered reasonable in light of several complicating factors which can affect the initial enrichments obtained from fuel records. These complications include, for example, weighting factors, residual heels in dissolver tanks, recycle acid and measurement biases for ^{236}U, and are discussed in Appendix A.

Plutonium 239

The uranium isotopes were thus seen applicable to the determination of initial enrichments from isotopic measurements of spent fuel. The plutonium isotopes appear to be a general guide to further definition of reactor type.

The ^{239}Pu wt% decreases most rapidly for HWR and BWR fuels as seen from Table II-C even though the shapes of the burnup curves representing ^{239}Pu versus ^{235}U for HWR, BWR and PWR data are similar, as seen from Figure 2. At a given level of exposure, 10,000 MWD/MTM for example, the ^{239}Pu wt% of the total measured plutonium increases as the initial enrichment increases for a given type of reactor. Fuels of equal initial enrichment but irradiated in different reactor types, for example natural uranium in a GCR or HWR and 3.0 wt% ^{235}U in a BWR or a PWR, show ^{239}Pu wt% values that are different for equal exposure levels. This results partly from the different amounts of ^{235}U consumed and partly from the different rates of utilizing plutonium in the various reactor types.

The ^{239}Pu wt% values are thus higher for PWR data than HWR and BWR data, and the differences between respective values of equal enrichment fuel increases as exposure increases.

Plutonium 240

The shapes of the ^{240}Pu versus ^{235}U burnup curves, see Figure 3, are distinctly different for HWR and BWR data as compared to PWR data. The ^{240}Pu burnup curves increase more linearly in a HWR and BWR, and thus a distinguishing feature of HWR and BWR data is the larger ^{240}Pu wt% relative to the ^{240}Pu wt% in a PWR as seen in Table II-D.

For a given level of exposure, e.g., 10,000 MWD/MTM, the ^{240}Pu wt% decreases as the initial enrichment increases. This trend is coupled with the ^{239}Pu trend in the $^{240}Pu/^{239}Pu$ ratio which therefore exhibits even greater differences between BWR and PWR fuels of equal enrichment than the individual ^{239}Pu and ^{240}Pu values. This is seen in Table IV. The $^{240}Pu/^{239}Pu$ ratio differentiates between the BWR 3.0 wt% and the PWR 3.0 wt% fuels above an exposure level of 10,000 MWD/MTM. Differentiation however is not possible between BWR and PWR fuels of equal enrichment below 10,000 MWD/MTM.

The uranium isotopes can thus be used to indicate initial enrichment. The plutonium isotopics can be used to differentiate between a BWR or a PWR if the initial enrichment indicates the possibility that the fuel could be used in both types and if the exposure is over 10,000 MWD/MTM.

Plutonium 241

The burnup curves representing the ^{241}Pu wt% data versus ^{235}U, Figure 4, for the HWR, BWR and PWR reactor are similar in shape. At a given exposure level, 10,000 MWD/MTM, the ^{241}Pu wt%, see Table II-E, decreases as the initial enrichment increases, except for HWR data, where the ^{241}Pu values are lower than the 1.5 wt% BWR values. The ^{241}Pu wt% values for 3.0 wt% ^{235}U fuels in a BWR and a PWR are seen to be approximately equal even though the other isotopic values differ. The ^{241}Pu could not be used to differentiate between BWR and PWR fuels of the same initial enrichment. However, ^{241}Pu may provide differentiation between HWR and BWR fuels of the same initial enrichment. Sufficient measured data are not available to make a conclusion. Also, the ^{241}Pu data have not been corrected for decay. The data at present do not have a common time base, which complicates making a decision. Data from higher enriched fuels in a HWR should indicate what differentiation is possible from ^{241}Pu wt% values.

Plutonium 242

As seen from Figure 5 and Table II-F, the ^{242}Pu evidences trends similar to ^{240}Pu. That is, at a given exposure, 10,000 MWD/MTM, the ^{242}Pu wt% decreases as the ^{235}U enrichment increases. The decrease of ^{242}Pu wt% from natural uranium to 5.0 wt% ^{235}U is approximately a factor of ten for an equal exposure level of 10,000 MWD/MTM. There is a distinct difference between BWR and PWR ^{242}Pu values for 3.0 wt% ^{235}U enriched fuels once the fuels reached an exposure level above 10,000 MWD/MTM. Thus, the $^{242}Pu/^{239}Pu$ ratio whose values are given in Table V is a sensitive index to differentiate reactor type for BWR and PWR fuels of equal initial ^{235}U enrichment which have accumulated more than 10,000 MWD/MTM.

APPLICATION TO VERIFICATION FOR SAFEGUARDS

It has been shown by the data plotted in Figures 1 through 5 and the isotopic values tabulated in Tables II, IV and V that the measured data from spent fuels listed in Table I provide very useful information in addition to total amounts of uranium and plutonium and their fissile compositions. An estimate of the initial enrichment to within ± 3% was

TABLE IV

THE $^{240}Pu/^{239}Pu$ RATIO OF SPENT FUEL BASED ON MEASURED DATA

ISOTOPE	$^{240}Pu/^{239}Pu$*											
REACTOR TYPE	HWR	BWR				PWR					Pu RECYCLE (SAXTON-PWR)	GCR
INITIAL ENRICHMENT	NAT.	1.500	2.000	2.500	3.000	3.000	3.500	4.000	4.500	5.000	NAT.-6.6	NAT.
EXPOSURE (MWD/MTM)												
0	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.096	0.000
1000	0.070	0.047	0.039	0.031	0.026	0.029	0.025	0.019	0.016	0.013	0.101	0.078
2000	0.125	0.089	0.075	0.060	0.049	0.054	0.046	0.038	0.032	0.027	0.108	0.142
3000	0.186	0.131	0.104	0.087	0.071	0.074	0.063	0.052	0.044	0.040	0.116	0.203
5000	0.290	0.200	0.161	0.134	0.114	0.106	0.092	0.081	0.071	0.063	0.133	0.313
10000	0.486	0.361	0.289	0.240	0.205	0.164	0.145	0.132	0.120	0.109	0.164	
15000		0.531	0.426	0.355	0.300	0.209	0.188	0.171	0.158	0.147	0.210	
20000				0.470	0.394	0.248	0.224	0.207	0.193	0.180	0.258	
25000						0.285	0.259	0.238	0.226	0.213	0.309	
30000								0.271	0.257	0.244	0.359	

*Using weight percents.

TABLE V

THE $^{242}Pu/^{239}Pu$ RATIO OF SPENT FUEL BASED ON MEASURED DATA

ISOTOPE	$^{242}Pu/^{239}Pu$ Ratio*											
REACTOR TYPE	HWR	BWR				PWR					Pu RECYCLE (SAXTON-PWR)	GCR
INITIAL ENRICHMENT	NAT.	1.500	2.000	2.500	3.000	3.000	3.500	4.000	4.500	5.000	NAT.-6.6	NAT.
EXPOSURE (MWD/MTM)												
0	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.051	0.000
1000	0.037**	0.023	0.021	0.019	0.015	0.013	0.010	0.005	0.003	0.002	0.053	0.043
2000	0.128	0.089	0.071	0.062	0.048	0.042	0.032	0.025	0.022	0.021	0.067	0.140
3000	0.320	0.257	0.171	0.131	0.092	0.087	0.065	0.051	0.039	0.038	0.091	0.373
5000	1.06	0.732	0.471	0.316	0.239	0.229	0.169	0.132	0.115	0.098	0.116	1.30
10000	4.78	4.09	2.33	1.47	1.01	0.870	0.666	0.526	0.415	0.326	0.228	
15000		9.76	7.24	4.71	3.03	1.90	1.51	1.28	1.01	0.827	0.433	
20000				8.66	6.23	3.30	2.65	2.23	1.90	1.63	0.757	
25000						5.19	4.05	3.43	3.00	2.66	1.23	
30000								4.82	4.24	3.77	1.86	

*Using weight percents.

**All numbers are multiplied by 10^{-2}, such as 0.037×10^{-2}.

seen to result using the measured uranium isotopes and Figure 8. A useful safeguards procedure is then to reverify the initial enrichment as shown in records and reports of the reactor, fuel fabrication and enrichment facilities by means of the uranium isotopic data measured where spent fuel has been dissolved.

Information as to reactor type in which the fuels were irradiated in, a BWR or a PWR, was seen to result from the uranium and plutonium isotopic values. The differentiation between a BWR and a PWR which burn equally enriched ^{235}U fuels was seen to be possible at exposures of 10,000 MWD/MTM and above using plutonium isotopic values from spent fuels. In the area of safeguards verification, the reactor type derived from the measured data should agree with the reactor type as listed in fuel records which are obtained from sources external to the chemical reprocessing plant.

Having determined the initial ^{235}U enrichment and reactor type from the measured data, it is a straightforward process to estimate the exposure of a batch of spent fuel using Figure 7 and the measured ^{235}U. All that is necessary is to select the appropriate burnup curve as designated by initial enrichment and reactor type and pick off the exposure value at the point which corresponds to the final measured ^{235}U. It is expected that the exposure values are valid to $\pm$ 10%.

In the realm for safeguards verification, these estimated exposure values can be expected to be consistent with calculated exposures obtained from fuel records. This last procedure requires that the safeguards authority also knows the fuel assemblies which make up individual dissolution batches in order to know correct weighting factors to be applied to calculated exposures listed by fuel assembly.

A consideration relevant to the above three determinations is that it is not possible to arrive at the measured isotopic values by an unlimited number of burnup paths when the initial starting point, the reactor type and the approximate exposure level are known. That is, it is impossible to arrive at the isotopic composition of Yankee Rowe fuel burned to 30,000 MWD/MTM starting with 1.5 wt% ^{235}U fuel in a BWR. Thus, the relative isotopic composition of the measured data immediately indicates limitations as to the number of possible burnup paths that are feasible in arriving at the specific isotopic values.

Upon receiving newly measured data from processed fuels of the BWR and PWR reactors listed in Table I, a logical order of procedure to determine initial ^{235}U enrichment, reactor type and exposure from the data is as follows:

1). From the measured $^{236}U/^{235}U$ and $^{236}U/^{238}U$ ratios, determine the initial ^{235}U enrichment by plotting the values on Figure 8.

2). From the measured uranium and plutonium isotopic values, determine reactor type by comparing measured values to tabulated values of Tables II, IV and V and by plotting values on Figures 1 through 5.

3). From the measured ^{235}U values, determine the exposures of the input dissolution batches by selecting an appropriate burnup curve from Figure 7 as dictated by 1) and 2) above and read off the exposures at points which correspond to the final ^{235}U values.

4.). Compare the information obtained in steps 1), 2) and 3) above with data from the reactor, fuel fabrication and enrichment facility. This includes comparisons of burnup curve shape and position.

Future Application

The above procedures, the empirical burnup curves shown in Figures 6, 7 and 8 and the values tabulated in Tables II, IV and V can best be shown to provide meaningful verification as new measured data are obtained from the reactors listed in Table I. More data are expected as each reactor listed continues to operate. It is but a matter of time until additional data are received and the procedures applied to the data.

Whether or not the burnup curves shown in Figure 8, which were seen to be the most independent of reactor design parameters, can be applied to other reactor types or newer generation reactors of similar types utilizing natural uranium to 5.0 wt% ^{235}U enriched fuels can be determined as measured data representing other reactors are collected. For example, complete measured data from Douglas Point I, Sena and Big Rock Point are being collected and will be compared to the data presented in this report. Should these data, as well as data for other possible operating reactors, apply in the case of Figure 8, it can be concluded that the burnup curves shown in Figure 8 are truly independent of reactor design parameters, except the initial enrichment.

It is expected that future isotopic data from operating BWR and PWR reactors other than those listed in Table I will evidence different isotopic compositions. To date, comparisons of data from one BWR versus another BWR and one PWR versus another PWR have shown data differences which lead one to conclude that reactors of the same type have different neutron spectra. Therefore, for fuel of equal ^{235}U enrichments, the burnup curves, such as those shown in Figures 1 through 5, are expected to change for other reactors due to different neutron spectra present in these reactors. Thus, isotopic composition will be different from those shown in Tables II, IV and V and the magnitude of difference to be seen will depend on the isotope.

Based on what has already been observed from measured data, the ^{236}U versus ^{235}U burnup curves are the least likely to change and most likely to apply to data from equal initial enrichment fuels of other reactors in addition to the burnup curves shown in Figure 8. The ^{240}Pu versus ^{235}U burnup curves are the least likely to apply. That is, the magnitude of change varies per isotope and the ^{236}U versus ^{235}U burnup curves, Figure 1, changed the least and the ^{240}Pu versus ^{235}U burnup curves, Figure 3, changed the most due to spectrum differences in a BWR versus a PWR.

Although isotopic compositions are expected to be different for other BWR and PWR fuels, particularly for ^{240}Pu, the changes are not expected to be as large as already seen from the BWR and PWR data presented. Calculations of Fort Calhoun[17] show that ^{240}Pu burnup curves in a newer generation PWR will be similar in shape to those PWR curves shown in Figure 3, but they will be positioned one or two units of ^{240}Pu wt% higher. Thus, it is expected that separate families of burnup curves representing different generation PWRs will possibly be necessary when applying future measured data to burnup curves that vary due to neutron spectrum differences.

Another parameter that is expected to change, but for another reason, is the exposure values used in Table II. As mentioned, the exposure values resulted from burnup calculations, and it is planned that exposure values in the future are to be determined more directly from the measured spent fuel isotopic data. A method referred to as the heavy element or mass spectrometric method[18] is to be applied. The capture-to-fission ratios for the fissile isotopes are needed in this method of determining exposure, and these ratios are to be obtained by least-squares methods [19] applied to the isotopic data. These experimental numbers are to replace the calculated values, and thus the exposure values are subject to change.

Application to GCR and Plutonium Recycle Data

From the GCR and plutonium recycle data shown in Figures 1 through 8 and Tables II, IV and V it was evident that these data are also amenable to isotopic correlation techniques. The burnup curves representing the GCR data were similar in shape to the HWR data. However, their positions were different indicating that the different neutron spectrum in a GCR results in slightly displaced burnup curves for natural uranium fuel as compared to the HWR burnup curves. The result was that isotopic composition for GCR and HWR data were different, which may lead to information concerning reactor type. Also, from the GCR data shown in Figure 8, it was indicated that the empirical curves of this figure will likely apply to GCR data even though neutron spectrum differences are evident from other figures. The probable application is indicated since the GCR data follow quite closely along the natural uranium burnup curve in Figure 8.

In the case of the plutonium recycle data, the main purpose of presenting the data was to show that isotopic relationships do indeed exist for these data, and thus isotopic correlations can be applied to plutonium recycle data. The specific isotopic ratios presently used for natural uranium and ^{235}U enriched fuels may not be as useful for plutonium recycle data. However, because isotopic relationships do exist, other useful ratios can be derived to use when reprocessing data from mixed oxide fuels are finally available.

From the plutonium recycle data shown in Figure 8, the empirical burnup curves may not apply since the data indicate an initial ^{235}U

enrichment of 0.80 wt% ^{235}U rather than natural uranium. However, to derive empirical burnup curves from plutonium recycle as has been done for natural uranium and slightly enriched ^{235}U fuels should be possible once sufficient measured data are available.

CONCLUSIONS

The purpose of presenting measured chemical plant data in this report was to observe isotopic composition differences in several reactor fuels. Analyzing the collective data has achieved the desired goal and at the same time has illustrated the derivation of useful information from a central data file such as has been collected to date. An important conclusion reached from the analyses was that the data file can play an important role in future safeguards verification procedures.

As a result of the analyses done, it is particularly important that isotopic relationships be developed from which information can be derived that has maximum verification from external information sources. It has been shown that information such as the initial enrichment, reactor type and exposure can be derived from measured spent fuel data, and the intent is to pursue other possible relationships to determine what additional information can be derived.

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APPENDIX A

METHOD TO DETERMINE EMPIRICAL BURNUP CURVES

The data sets listed in Table 1 of this report represent several reactors of different designs. For example, design differences in initial enrichments, cladding materials, lattice pitch, fuel temperatures, coolants, voids, moderators and achieved exposures are represented. These design differences as well as others resulted in neutron spectra differences in the various reactors and the isotopic composition data were seen to reflect the neutron spectra differences. Various qualifications were therefore necessary to collectively analyze the data sets in order to empirically interpolate and extrapolate the data to families of burnup curves. The purpose of this appendix is to describe the empirical methods used and to define the qualifications applied.

The procedure used to interpolate and extrapolate empirically from the data and burnup curves shown in Figures 1 through 5 of the report to families of burnup curves is illustrated using as an example the ^{236}U versus ^{235}U case. To determine the dashed burnup curves shown previously in Figure 6, the following was done:

1) Seven values of ^{236}U were selected which covered the ^{236}U range of the collective data (0.05, 0.10, 0.20, 0.30, 0.40, 0.50 and 0.60 wt%).

2) At the intersection of the ^{236}U selected values with the burnup curves of Candu, Dresden I, Humboldt Bay and Yankee Rowe, corresponding ^{235}U values were read from Figure 1. These burnup curves represented fuels whose initial ^{235}U wt% were 0.7114 (Candu), 1.474 (Dresden I), 2.578 (Humboldt Bay), 3.406, 4.101 and 4.94 (Yankee Rowe).

3) The ^{235}U values thus determined were plotted versus the initial enrichment values given in 2) above and points representative of a single ^{236}U value were connected by a continuous curve through them. The result was seven curves representing each selected ^{236}U value. The curves indicated the ^{235}U values corresponding to the selected ^{236}U values at any initial enrichment from natural uranium through 5.0 wt% ^{235}U.

4) For the initial ^{235}U enrichments of 1.5, 2.0, 2.5 and 3.0 wt% for a BWR and 3.0, 3.5, 4.0, 4.5 and 5.0 wt% for a PWR, ^{235}U values and corresponding ^{236}U values were read from the curves of step 3 and plotted on Figure 1. A dashed line was then drawn connecting these points with the initial enrichment point as shown in Figure 6.

It was seen that the curves determined in step 3) above were approximately linear and were thus approximated by a straight line.

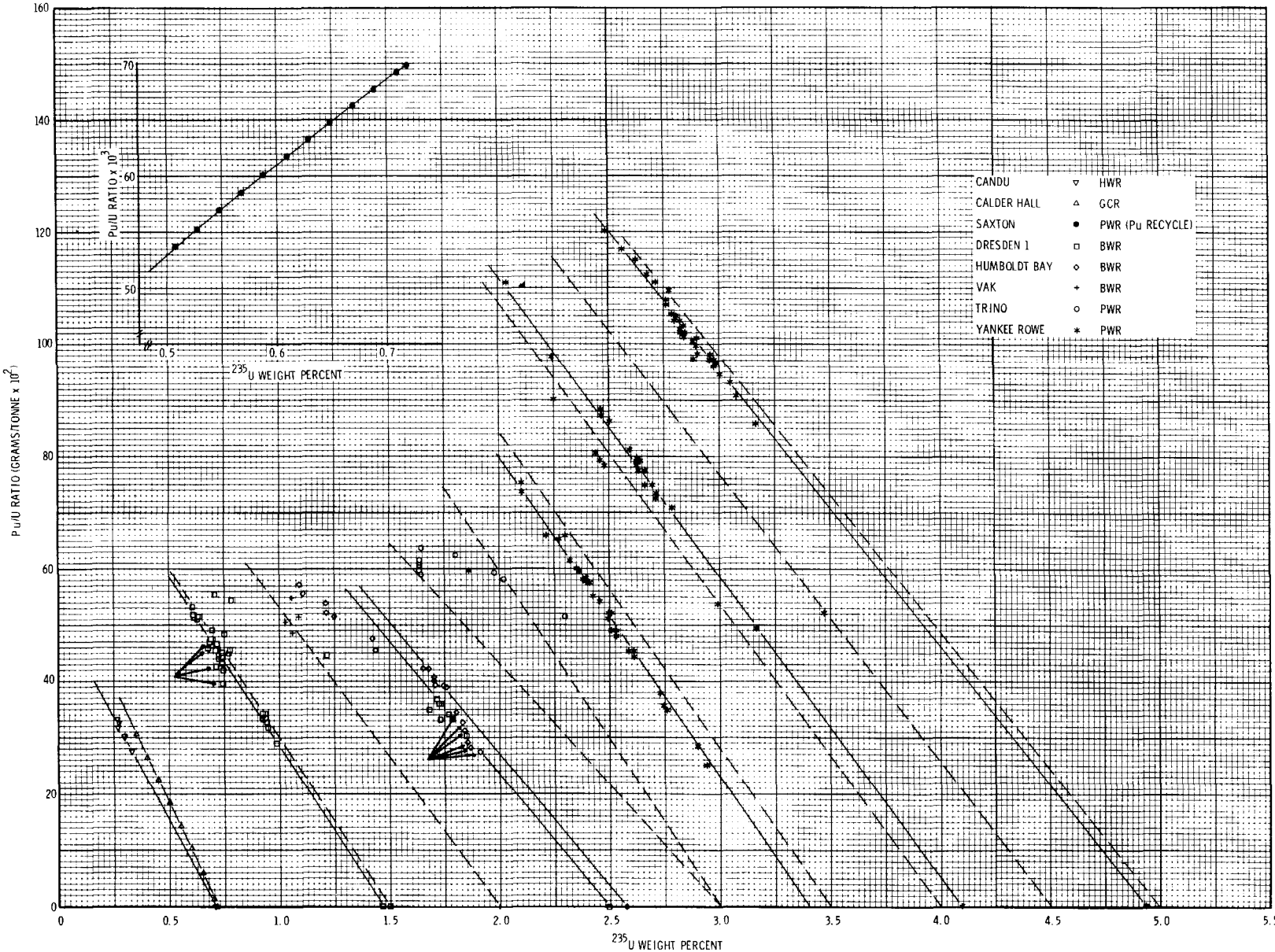

FIGURE A-1 PLUTONIUM-TO-URANIUM RATIO AS A FUNCTION OF ^{235}U WEIGHT PERCENT.

The curves were also continuous at 3.0 wt% ^{235}U which represented equally enriched ^{235}U fuels in a BWR or a PWR. This result indicated that at least for the ^{236}U versus ^{235}U, the BWR and PWR burnup curves of 3.0 wt% ^{235}U fuel are similar for the reactors considered.

Each plutonium isotope, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu, can be substituted for ^{236}U in the four steps outlined above along with a selection of values that covers the data range of each plutonium isotope. By performing the steps for the plutonium isotopes, it was seen that step 3) above did not result in continuous curves for a given selected value. Rather, the curves connecting the points representative of a selected value were discontinuous for BWR and PWR equally enriched fuels. Two approximately linear curves were seen to result for a given selected value of the plutonium isotopes, one for BWR data and one for PWR data. Thus, the differences in neutron spectra were more evident from the plutonium isotopic data. The fact that the step 3) curves were discontinuous may indicate a third curve for HWR data from natural uranium to 1.5 wt% ^{235}U and therefore no interpolation was done in this enrichment range for a HWR.

Due to neutron spectra differences from reactor type to reactor type, from reactor to reactor of the same type as well as differences within a given reactor core, particularly a BWR, certain procedures were followed in order to understand the effect of neutronic difference on the burnup curves. The isotopic data were plotted as a function of ^{235}U because one parameter that neutron spectra, and thus the burnup curves, are dependent on is the initial ^{235}U enrichment. Choosing ^{235}U as the basic plotting parameter helped to identify the effect that ^{235}U enrichment had on the burnup curves and separate this effect from other effects. For BWR data from Humboldt Bay and Dresden I, it was seen that, in addition to changes in burnup path due to enrichment, data from fuels which resided in the central region of the cores evidenced slightly different burnup paths than data from equally enriched fuels which came from outer regions of the core.

This second effect was not referred to when presenting Figures 1 through 5 and is therefore illustrated in Figure A-1 where the Pu/U ratio is shown as a function of ^{235}U. The data points representing fuels from the outer region of the Dresden I and Humboldt Bay cores are identified by the arrows and the data points representing the fuels from the central regions of both cores lie on or near the burnup curves shown for those sets of data. It is seen that the fuels from the outer regions of the core evidence lower burnup paths [6,7] than fuel from the center of the core. These lower lying data points were neglected in performing the procedure to determine empirical burnup curves. Thus, the empirical BWR burnup curves represent fuels which were irradiated in the central regions of Dresden I and Humboldt Bay reactors. This qualification pertains not only to the curves determined for the Pu/U ratio versus ^{235}U (dashed curves in Figure A-1) but to all empirical BWR curves determined except those curves seen in Figure 8.

Another qualification pertained to the initially enriched fuel of 4.94 wt% ^{235}U from Yankee Rowe Cores VI, VII and VIII. The uranium isotopic data from Core VIII evidenced slightly different burnup curves than the burnup curves from Cores VI and VII. The data points from Core VIII in Figure 1 (or 6) were consistently lower and in Figure 8 were consistently higher with respect to the burnup curves shown which were drawn through the Core VI and VII data. This effect was not evident from the plutonium isotopic data as seen from any of the figures involving the plutonium isotopes which would suggest that the effect was due to the initial ^{235}U, ^{236}U or to measurement bias involving the uranium isotopic values. That is, the initial enrichment may have been 4.90 wt% ^{235}U instead of 4.94% (1% difference), the ^{236}U initially may not have been as high as previous Core VI and VII fuels or a bias of 1% or less was present when the uranium isotopic data were measured. The analysis of Core VIII data has not been finalized as yet, however, at this point in time the effect is felt to be due to the initial ^{236}U or ^{236}U values being slightly lower than previous fuels. The interpolation and extrapolation was therefore done using the data from Core VI and VII.

Neutronic effects other than those due to initial ^{235}U enrichment were not observed when the burnup curves shown in Figure 8 were determined. The burnup curves for Dresden I and Humboldt Bay were seen to pass through the data points representing fuels from the center and outer regions of the cores equally well. Several qualifications that pertain to Figure 8 were with respect to the dissolution process and initial ^{236}U values rather than the irradiation process. Various considerations affect how well the initial ^{235}U enrichment values can be known from fabrication and fuel records for the batches listed in Part B of Table I presented early in the report. The complications which pertain to the processing of spent fuels are as follows:

1) In the case of batches of mixed initial enriched fuels, the number of assemblies of a given enrichment was not exactly known since a part of an assembly was included in a mixed batch and an accurate estimate of how much was not available. The weighted initial enrichment from fuel records for such a batch was thus an estimate only. For these cases the values from Figure 8 are more accurate to indicate initial enrichment.

2) Residual heels in the dissolver tank representing previously processed fuels of a higher or lower initial enrichment than the enrichment of newly dissolved fuel assemblies were mixed with the newly processed fuel during dissolution. A weighted initial enrichment would thus result due to the mixing which would be higher or lower depending on how large the heel was and how much higher or lower the initial enrichment of the previously processed fuel was. The initial enrichment value from Figure 8 would therefore be an indication of how large the residual heel was for batches of single enrichment fuels processed after processing fuel of higher or lower enrichments.

3) Use of recycle acid containing uranium of a different initial enrichment can complicate the accuracy by which the isotopic values are known. Although corrections are applied for recycle acid, the corrections add to the variations of the ^{236}U and ^{235}U.

4) Biases in mass spectrometer measurements such as have been noted for ^{236}U [4] also are complicating effects. Three significant figures for ^{236}U are needed for application of measured uranium isotopic data to Figure 8 and biases, recycle uncertainties and low mass spectrometer resolution tend to limit the application.

These complications along with the possibility that initial ^{236}U values may be different than the values assumed need to be taken into consideration when comparing the initial ^{235}U enrichment available from fuel records against the enrichments determined from Figure 8 which were shown earlier in Table III. A higher initial ^{236}U (or lower) value can result in a higher initial ^{235}U (or lower) value being determined from Figure 8.

ISOTOPIC CORRELATION SAFEGUARDS TECHNIQUES: PROPERTIES OF CHEMICAL REPROCESSING PLANT MEASUREMENTS RELATING TO BURNUP

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ABSTRACT

Data resulting from analytical measurements on representative samples of dissolution batches of spent reactor fuels at a chemical reprocessing plant have been evaluated employing techniques used in the past with burnup data. It was determined that burnup relationships between the depletion and growth of uranium and plutonium isotopes were readily evident from chemical plant data. This result is of importance since the measured batch data thus evidence a property termed as data consistency. Various evaluation techniques based on this result were applied to the measured batch data and it was seen that information pertaining to evaluation of data consistency was obtained.

INTRODUCTION

The purpose of reactor burnup experiments such as those conducted in the Yankee Core Evaluation Program[1] has been to provide measurements of the concentration and isotopic changes occurring in nuclear fuels while undergoing neutron irradiation. Extensive sampling of irradiated fuels has been done by cutting $^1/_2$ to 3 in. long specimens from selected fuel rods, resulting in small samples that were dissolved and analyzed. Various analytical methods have been used to measure the uranium (U) and plutonium (Pu) concentrations and isotopic compositions, and in recent years these methods have included sophisticated isotopic dilution techniques. The resulting data (hereafter refered to simply as burnup data) have indicated the relationships that exist between the depletion and growth of the U and Pu isotopes and these data have been used as a "benchmark" in the development and normalization of reactor design codes.

In 1967, chemical reprocessing measurements on spent reactor cores, namely Dresden I[2] and Yankee Rowe[3,4,5], became available for the first time. Isotopic ratios for U and Pu along with total U and Pu of dissolved spent fuel batches were measured at Nuclear Fuel Services, Inc., West Valley, New York, using isotopic dilution techniques. These data (referred to as chemical plant data) are similar to burnup data but the measurements are made on larger portions of the reactor core, as large as two metric tons U (tonne), than those made to obtain burnup data.

This report presents a comparison of burnup and chemical plant data to determine the extent that the relationships between the depletion and growth of U and Pu isotopes are evident from the data. Based on burnup relationships, the report illustrates data consistency procedures used in the past to analyze burnup data where the procedures have now been applied to chemical plant data. The term data consistency is defined and a value index is assigned in order to indicate the information available from applying data consistency evaluation procedures to chemical plant data. The report is intended as an introduction into data consistency procedures and does not involve detailed statistical methods.

Several safeguard techniques have been developed based on the use of spent fuel isotopic data[6-9] and include a) the verification of plutonium content of irradiated fuels measured at input to a chemical reprocessing plant, b) confirmation of available historical information, c) data consistency evaluation and d) diagnostic evaluation to define burnup characteristics from the measured data. All the techniques are interrelated and those techniques pertaining to a) and b) have been discussed in detail in other reports.[6-9] This report and a companion report[10] are written to describe in detail the development of the techniques being used in the areas of c) and d) above. This report considers isotopic relationship properties as they apply to the evaluation of data consistency.

DATA COLLECTION PROCEDURES

Burnup Data

The burnup data considered in this report resulted from the analysis of small fuel samples collected from Yankee Rowe core I fuels during the Yankee Core Evaluation Program.[1] Details of measurement procedures have been outlined in Reference 1 and are therefore not discussed here other than to point out that the measurements were performed utilizing the technique of isotopic dilution. Of the data measured, the U and Pu concentrations and ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu data are used in this report.

Burnup data include measurements of the concentration of a selected fission product to provide exposure values. However, chemical plant measurements do not normally include fission product determinations and the presentation of Yankee Rowe burnup data proceeds without specific use of exposure values.

B. Chemical Plant Data

The chemical plant data were also measured by isotopic dilution techniques.[3] Not all of the chemical plant data considered are normally required for material accountability. Total U and Pu plus ^{235}U, ^{239}Pu and ^{241}Pu weight percents (wt%) are normally required. Nevertheless, by making these measurements, ^{234}U, ^{236}U, ^{238}Pu, ^{238}U, ^{240}Pu and ^{242}Pu compositions are also measured. Details on the analytical procedures used in obtaining these data are given in Reference 3. The measurements for Dresden I and Yankee Rowe cores represented a first time in history for the direct and accurate measurement of both U and Pu isotopic compositions of input dissolver solutions on a routine basis. We will be concerned here with measurements of Yankee Rowe cores

I through IV as reported in Table A5.1 of Reference 4.

Common procedure used in reprocessing a spent reactor core, or a portion thereof, is to divide the total number of spent fuel assemblies to be reprocessed into groups as dictated by dissolver batch size. In the case of the reprocessing of Yankee Rowe core I[3], 75 fuel assemblies were divided into 16 dissolution batches which were sampled and measured. The amount of U in a batch varied between 2166 to 944 kgs. The assemblies were processed in random order and the 16 batches did not necessarily consist of whole assemblies. A total of 76 assemblies from cores II and III were processed in 14 batches. The material from core IV, 36 fuel assemblies, was processed in 11 measured batches. During the time each batch was in the input accountability tank, volume measurements were made and samples of each batch were obtained for analysis. Resulting U and Pu isotopic and concentration data from measurements made on each input batch sample are the data considered in this report.

COMPARISON OF BURNUP AND CHEMICAL PLANT DATA

It is useful when considering burnup relationships of spent fuel data to also include the zero burnup values of the fuel. The initial ^{235}U enrichment of cores I through III was 3.406 wt%[4] (an average) and for core IV fuel, 4.101 wt%. The initial ^{236}U wt% was 0.020 and the ^{238}U wt% was 96.553 for cores I through III. A slight drawback was encountered in not knowing the initial ^{236}U for core IV. However, an assumed value of 0.032 wt% was used along with 95.847 wt% for ^{238}U. In the case of four dissolution batches from core IV, fuels of differing initial enrichments were dissolved together (referred to as mixed batches). An average weighted initial ^{235}U enrichment of 3.90 wt%, 0.028 wt% for ^{236}U, and 96.047 wt% for ^{238}U was used for these batches. The weighted averages were estimated from information given in Table 16, Reference 4.

The burnup data collected from Yankee Rowe core I fuels during the Yankee Core Evaluation Program[1] are of particular interest since the set can be compared to chemical plant data that resulted from the dissolution of Yankee Rowe core I. Comparisons of these burnup and chemical plant data are presented in Figure 1, where the Pu/U ratio versus ^{235}U depletion and $^{241}Pu/^{239}Pu$ data versus $^{240}Pu/^{239}Pu$ data are shown. Decay corrections to date of discharge were made to compare the Pu isotopes. Only representative samples of small sample burnup data from regions which have been referred to[1] as the "perturbed" region, the "intermediate" region and the "asymptotic" region are shown. These regions correspond to different positions within a Yankee Rowe fuel assembly.

The purpose in comparing the data sets is to determine the relative consistency of chemical plant data versus that of burnup data. Qualitatively, if the data when plotted define a smooth curve which can be drawn by inspection and the data points have "small" scatter about the curve, then the data are said to be consistent. Burnup data previously analyzed[11-14] have usually been sufficiently consistent to indicate the burnup relationships (or the burnup curves) that exist between the U and Pu isotopes and concentrations. These relationships were apparent when the collective data of a burnup data set were plotted since the collective data defined a smooth burnup path or curve. The more consistent the burnup data were, the more well defined were the burnup curves and henceforth the burnup relationships were better defined. In this context, Yankee burnup data are seen to be consistent to the point that data from each region define different burnup paths, the Pu isotopes particularly.

The chemical plant data are also seen to be consistent. Collectively, they define an average burnup path which results from dissolving entire fuel assemblies instead of certain positions within the assembly. As expected because of the large amount of fuel dissolved, it can be seen that chemical plant data have greater consistency as evidenced by the decreased scatter of chemical plant data. Thus, the relationships between the depletion and growth of U and Pu isotopes and concentrations should be more evident and well defined when chemical plant data are plotted than has been evident from burnup data. A value index of data consistency is defined in the next section.

ANALYSIS OF CHEMICAL PLANT DATA

In the past, data plotting has been one of the many useful evaluation methods to evaluate data consistency and detect poorly measured data when applied to burnup data. The fact that greater consistency may be obtained from chemical plant data is the basis for applying these evaluation methods to these data also. Potentially, the methods have greater value to derive useful information when applied to chemical plant data due to the greater consistency, and the potential value will be looked for in the process of examining the quality of chemical plant measurements. Input batch data of Yankee cores I through IV are used to illustrate the techniques.

The use of plotting requires that we define contributions to the scatter (or consistency) of plotted data where the scatter is with reference to a curve representing the burnup relationship drawn through the data. For burnup data, contributions include:

1) Scatter due to random errors[15] of measurement, sampling and analytical sources. Systematic errors (or biases) are not detectable by the methods used.

2) Scatter due to variations in reactor conditions such as position of fuel sample, exposure level, coolant properties, etc., which effect change in burnup relationships.

Based on past experience with burnup data, two sigma (2σ) values representing the algebraic sum of 1) and 2) above were assigned to the Pu/U ratio and to U and Pu isotopic values as follows:

	2σ VALUE		2σ VALUE
Pu/U Ratio	**3–5%**	**^{239}Pu**	**1–2%**
^{235}U	**1–2%**	**^{240}Pu**	**2–3%**
^{236}	**4–7%**	**^{241}Pu**	**3–4%**
^{238}U	**0–2%**	**^{242}Pu**	**4–5%**

For the purposes of this report these values are defined as data consistency indexes.

It has been observed through analysis of burnup data that random errors due to measurement or variations due to reactor conditions which resulted in scatter greater than the values listed were detectable. The result was that a remeasurement was done or the data were considered representative of sufficiently different burnup conditions to be deleted. The above values can be used as a base for comparison as we proceed to examine chemical plant data by similar methods used with burnup data.

A. Qualitative Evaluation of Uranium Isotopic Data

The U isotopic data from chemical plant measurements are shown in Figure 2. To test the chemical plant data qualitatively, we observe whether or not the plotted data points lie on or near a curve that the collective data describe. To indicate the burnup curve or relationship between growth of ^{236}U and depletion of ^{235}U, a straight line from the zero burnup point through the collective batch data has been drawn by inspection. Two curves result due to the different initial enrichments of the fuel dissolved. The result of mixing differing initial enrichment fuels is also evident by the line drawn through the four data points representing the batches of mixed fuel.

Good data consistency, at least to ± 2 or 3% of the curve drawn by inspection, is evident from Figure 2, since all the data points lie on or near the burnup curves (plus or minus 2% error bars are shown for each curve for ease of examination). The consistency index of ^{235}U is roughly estimated to be ± 1 to 1.5% (2σ) and for ^{236}U to be approximately ± 2 to 3% (2σ) based on the fact that all data points lie on or near the curves. By the above procedure, plotting is thus seen to provide a qualitative estimate of data consistency. A more quantitative approach is presented in the next subsection.

A note of interest was observed from the data representing the mixed initial enrichment fuel batches. The mixed fuel batches are identified in Reference 4 as batches 35, 38, 41 and 43. However, the isotopic data shown in Figure 2 indicate the identification to be batches 35, 38, 42 and 43. Measured isotopic data were thus seen to provide information concerning which batches actually contained fuels of different initial enrichments.

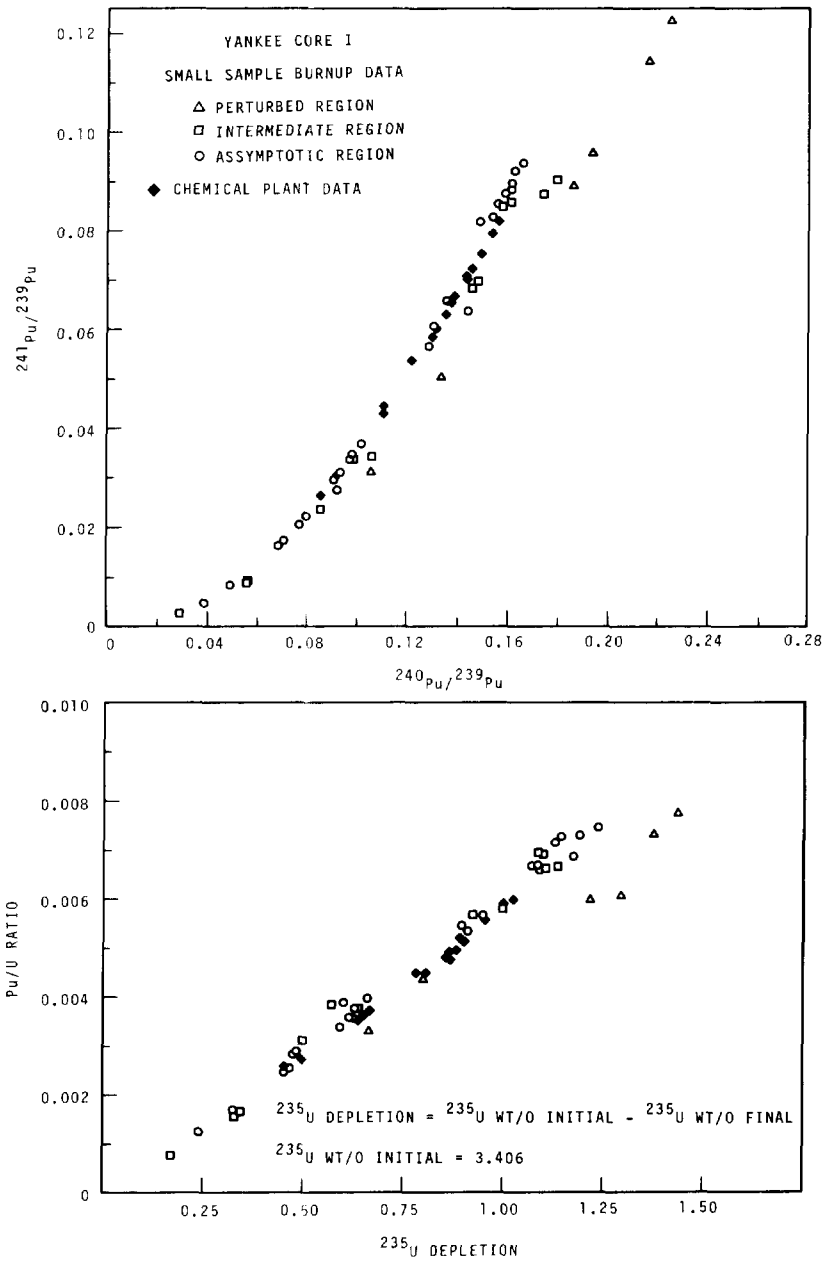

FIGURE 1. The $^{241}Pu/^{239}Pu$ Isotopic Ratio Versus the $^{240}Pu/^{239}Pu$ Isotopic Ratio and the Pu U Ratio Versus ^{235}U Depletion for Small Sample Burnup Data and Chemical Plant Data from Yankee Rowe Core I.

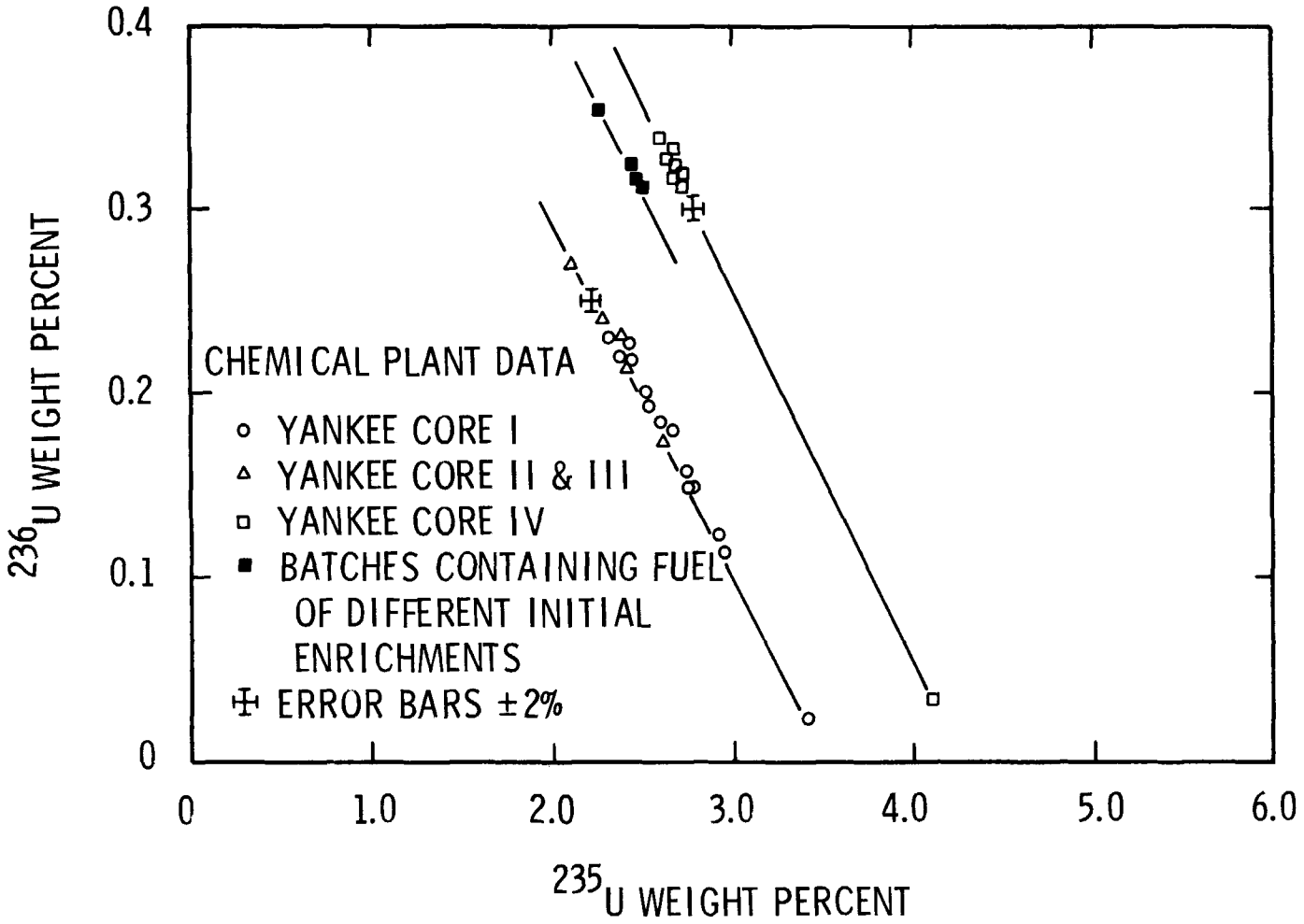

FIGURE 2. URANIUM ISOTOPIC DATA FROM CHEMICAL PLANT MEASUREMENTS

TABLE I

Ratios $1/(1 + 1/\alpha^{235})$ and $\sigma_a^{236}/\sigma_a^{235}$ at 0 and 25,000 MWD/MTM Exposure for the Yankee Rowe and Fort Calhoun Reactors

	$1/(1 + 1/\hat{\alpha}^{235})$			$\hat{\sigma}_a^{236}/\hat{\sigma}_a^{235}$		
	BOL*	EOL**	% Change	BOL*	EOL**	% Change
Yankee Rowe	0.2012	0.2182	7.8	0.21603	0.27322	+21%
Fort Calhoun	0.18959	0.19930	5	0.16711	0.20134	+17%

* Beginning of Life
**End of Life

B. Quantitative Estimate of Data Consistency

1. Theoretical Basis

The above plotting procedure which has been used to test the chemical plant's U isotopic data for consistency has its basis in the transmutation or burnup equations:

$$\frac{dN^i}{dt} = -\hat{\sigma}_a^i N^i \phi + \hat{\sigma}_c^j N^j \sigma - \lambda^i N^i ,$$

where j, i = ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu (1)

N^i is the isotopic concentration of the i^{th} isotope, $\hat{\sigma}_a^i$ is the spectrum averaged absorption cross section for the i^{th} isotope, $\hat{\sigma}_c^j$ is the spectrum averaged capture cross for the j isotope, ϕ is the neutron flux, λ^i is the decay constant of the i^{th} isotope and t is the time. The equations, when solved, provide the functional relationships that we are working with. From Equation (1), the slope of the lines shown in Figure 2 is

$$\frac{dN^{236}}{dN^{235}} = \text{SLOPE} = \frac{-1}{1+1/\hat{\alpha}^{235}} + \frac{\hat{\sigma}_a^{236} N^{236}}{\hat{\sigma}_a^{235} N^{235}}$$

where $\hat{\alpha}^{235}$ is the ratio of capture to fission for ^{235}U. (2)

Equation (2) can give some insight into the invariance of the slope seen in Figure 2. It is first clear from Equation (2) that dN^{236}/dN^{235} or the slope value is the difference of two terms. The term $1/(1 + 1/\hat{\alpha}^{235})$ increases about 8% in the burnup of the Yankee fuel up to 25,000 MWD/MTM (Megawatt Days Per Metric Tonne Metal) as determined from burnup calculations.[16] This increase is completely counter balanced by the term $N^{236}\hat{\sigma}_a^{236}/N^{235}\hat{\sigma}_a^{235}$ so that the net trend shown from calculations is for dN^{236}/dN^{235} to decrease slightly in absolute value at higher exposures. Initially the term $N^{236}\hat{\sigma}_a^{236}/N^{235}\hat{\sigma}_a^{235}$ exerts little influence because N^{236} is two orders of magnitude smaller than N^{235}. At this point, $dN^{236}/dN^{235} \approx -1/(1 + 1/\hat{\alpha}^{235})$,

Equation (2) shows the underlying physical reasons for the invariance of the slope in the plot of ^{236}U versus ^{235}U shown in Figure 2. Thus, we see that it is appropriate to represent the functional relationship between ^{236}U and ^{235}U as a straight line in Figure 2. Equation (2) further establishes that the slope will decrease (become less negative) at higher exposures and the decrease is caused by a correction term which is proportional to N^{236}/N^{235} and whole influence increases with exposure.

Table I gives some idea of the variation of $-1/(1 + 1/\hat{\alpha}^{235})$ and $\hat{\sigma}_a^{236}/\hat{\sigma}_a^{235}$ in both the Yankee and Fort Calhoun reactors as determined from burnup calculations.[16]

The percent change from beginning of life to end of life for the ratios in both reactors is similar. The absolute magnitude of the ratios varies from reactor to reactor because of the difference in the neutron energy spectra in these reactors. The Yankee Rowe neutron flux spectrum is different from the flux spectrum in more modern PWRs exemplified by Fort Calhoun. However, although differences are evident, the slope value of Equation (2) can be expected to be reasonably invariant.

2. Numerical Values of Linear Relationships

Since the function representing the relationship of U isotopic data for Yankee Rowe was approximately linear, it is worthwhile to determine an approximation to the numerical value of Equation (2) using individual batch data and the zero burnup data. That is, the value of Equation (2) can be approximated by

$$\text{SLOPE} \approx \frac{^{236}U_f - ^{236}U_o}{^{234}U_f - ^{235}U_o}$$

where f indicates the final values measured by the chemical plant for each batch and o indicates the initial values as discussed above. This approximation is good in cases where the functional relationship is highly linear. The procedure has the potential of providing more useful information than that obtained by plotting the U isotopic data. The absolute values of Equation (3) are tabulated in Table II. The listing of each batch is according to increasing exposure or decreasing ^{235}U content rather than numerical order. This has been done to identify any trend of the slope values to vary as the exposure increases, if that trend is present in the data.

First, we note by examining the values in Table II that there does not appear to be a trend for the values to decrease as a function of increasing exposure. Evidently, the fuel had not reached exposures where the correction term of Equation (2) would be significant. Secondly, the values are very constant from batch to batch.

TABLE II

TABULATION OF THE ABSOLUTE VALUES OF EQUATION 3 FOR INDIVIDUAL BATCH DATA FROM YANKEE ROWE CORES I THROUGH IV

Core I		Cores II and III		Core IV	
Batch Number	Slope Value	Batch Number	Slope Value	Batch Number	Slope Value
11	0.204	32	0.194	37	0.204
10	0.203	19	0.201	34	0.204
14	0.202	21	0.195	40	0.206
9	0.199	23	0.196	33	0.199
12	0.205	24	0.195	41	0.200
7	0.205	25	0.194	36	0.206
8	0.203	28	0.193	39	0.205
2	0.194	27	0.196	38*	0.202
15	0.205	26	0.196	35*	0.200
13	0.198	17	0.196	42*	0.203
5	0.195	22	0.196	43*	0.199
1	0.202	29	0.196		
6	0.198	30	0.194		
4	0.203	31	0.192		
3	0.207				
16	0.204				
MEAN	0.202		0.195		0.203
S^2	0.000014		0.0000044		0.0000073
STD.DEV.	0.004		0.002		0.003
%	1.9		1.1		1.3

*Batches containing fuels of differing initial enrichments. Weighted initial values were used.

The mean values of the batches for cores I, II & III and IV are listed in Table II and the standard deviations of individual batch values are also given. The mean values for core I and for cores II & III fuels whose initial ^{235}U enrichments were essentially the same are seen to be different, 0.202 and 0.195, respectively. The difference may indicate the presence of systematic error or shifts in systematic error or the difference may be due to real irradiation condition differences.[6] Nevertheless, the values are sufficiently close to provide quantitative information pertaining to data consistency.

The components of error for the values of Equation (3) include not only the random error due to the final ^{235}U and ^{236}U but also the random error of the initial ^{235}U and ^{236}U values. On the basis that the random errors of both the final and initial ^{235}U and ^{236}U values were the same, it is reasonable to conclude that the data consistency index of the ^{235}U is in a 0.5 to 1.0% (2σ) range and the ^{236}U is in a 1.0 to 3.0% (2σ) range. These ranges are smaller than derived from simple plotting of the data and were determined using standard propagation of error equations. The individual batch measurements are thus consistent enough to be able to use the procedure outlined above that permits the analyses of the data at a consistency level of approximately 1 to 3% (2σ). The procedure is useful to a safeguard agency to indicate the quality of measured chemical plant data since the scatter due to reactor conditions is reduced for chemical plant data and the consistency indexes more nearly reflect measurement errors.

C. Qualitative Evaluation of Plutonium Isotopes

These techniques of plotting and deriving numerical values for the slope can also be applied to the Pu isotopes to advantage as shown in Figure 3, and to a combination of U and Pu isotopes, as shown in Figure 4. The ^{241}Pu data shown in Figure 3 have been decay corrected to date of discharge and this required a weighted decay time for cores II & III fuels since eight of the batches contained fuels of different cooling times.

Again we note from inspection of the figures that the burnup paths (drawn by inspection) are well defined. The functional relationships are governed by the burnup equations also but are somewhat more complicated than those involving ^{235}U and ^{236}U and numerical parameters are not presented. However, from a qualitative point of view, the consistency of the Pu isotopic determinations are seen to be within a ± 2 to 3% envelope, Figure 3, as were the U isotopic determinations, Figure 2.

When the Pu and U isotopics are plotted versus each other as in Figure 4, which involves two separate mass spectrometer determinations, the consistency approaches the limits of $\pm$ 2 to 3% of the burnup path envelope as would be expected. Based on the qualitative results from Figures 2 and 3 as well as other plots not shown it was determined that a reasonable estimate of the 2σ range was 0.5 to 1.0% for ^{239}Pu, 1.0 to 2.0% for ^{240}Pu, 1.5 to 3.0% for ^{241}Pu and 2.0 to 4.0% for ^{242}Pu. Also, it appears from Figures 3 and 4 that the Pu isotopics for one batch (number 43) lies outside the envelope limit and the consistency of the Pu isotopic data of batch 43 are questionable. This is not to imply that all the Pu isotopic data for batch 43 are questionable. By examination of other graphs, it appeared that ^{239}Pu and ^{242}Pu were questionable which does not have important consequences except to indicate that inconsistent values may appear at times.

D. Qualitative and Quantitative Evaluation of Pu/U Ratio

Another example of the usefulness of plotting and numerical techniques for consistency evaluation involves the total U, total Pu and ^{235}U wt% as measured for each input batch at the chemical plant. The Pu/U ratio was formed and is shown plotted as a function of the change in ^{235}U wt%, i.e., ^{235}U depletion, in Figure 5. The collective data for each initial enrichment describe a straight line and it is evident that the individual batch data points generally lie within a ± 2 to 3% envelope. The lines as drawn by inspection may not be linear at higher exposures (x30,000 MWD/MTM) but are satisfactorily represented by straight lines over the range of the data[17] from core I through IV. The linearity of the relationship permits the use of the slope[6] as a consistency evaluation parameter where the slope in this case is expressed as

$$\text{SLOPE} = \frac{\text{Pu}}{\text{U}} \Big/ {}^{235}\text{U Depletion} \qquad (4)$$

$$\text{where } {}^{235}\text{U Depletion} = {}^{235}U_o - {}^{235}U_f . \qquad (5)$$

The units of Equation (4) are grams Pu per tonne U over ^{235}U loss in wt%.

The values derived are tabulated in Table III. The order of the batches is again arranged according to decreasing ^{235}U content. The mean values for each set of data were determined along with the standard deviations of the individual values and are listed in Table III. The four batches of mixed fuels in the core IV column were not included in the mean value shown since the values of Equation (4) vary as a function of initial enrichment as seen from Table III.

The means of fuels from core I and II & III which were initially enriched to the same value are different as was the case for values given in Table II. Again the differences are not large and may result from the same reasons mentioned previously. The relative standard deviations were approximately the same in this case as was seen for the values listed in Table II. Therefore, since the standard deviations are at this level and the 2σ range for ^{235}U was 0.5 to 1.0%, the consistency index for Pu/U values must be approximately 1.0 to 3%. (The ^{235}U consistency level matches that already seen in section IV.B). The estimated precision of the Pu/U ratio was quoted as 1.3% (two standard deviations).[4] Taking into account variations not necessarily due to measurement such as the fact that the relationship does vary slightly as exposure increases,[17] a 2σ range for data consistency of 1.0 to 3.0% is reasonable for the Pu/U ratio.

CONCLUSIONS AND SUMMARY

Data evaluation procedures based on the transmutation equations of nuclear fuel have been applied to data obtained from spent fuel measurements made at a chemical reprocessing plant. The relationships that exist between the depletion and growth of U and Pu isotopes were readily evident from the data since the burnup paths observed

TABLE III

TABULATION OF THE VALUES OF EQUATION 4 FOR INDIVIDUAL BATCH DATA FROM YANKEE ROWE CORES I THROUGH IV

Core I		Cores II and III		Core IV	
Batch Number	Slope B Value	Batch Number	Slope B Value	Batch Number	Slope B Value
11	5570	32	5781	37	5357
10	5760	19	5800	34	5266
14	5500	21	5773	40	5388
9	5500	23	5832	33	5266
12	5686	24	5831	41	5323
7	5683	25	5768	36	5362
8	5621	28	5739	39	5439
2	5579	27	5825	38*	5612
15	5688	26	5737	35*	5591
13	5581	17	5790	42*	5580
5	5551	22	5736	43*	5553
1	5846	29	5811		
6	5688	30	5826		
4	5711	31	5698		
3	5858				
16	5808				
MEAN	5664		5782		5343
S^2	13085		1788		4009
STD. DEV.	114		42		63
%	2.1		0.7		1.2

***Batches containing fuels of differing initial enrichments. Weighted initial values were used and the values derived were not used to obtain the mean value shown.**

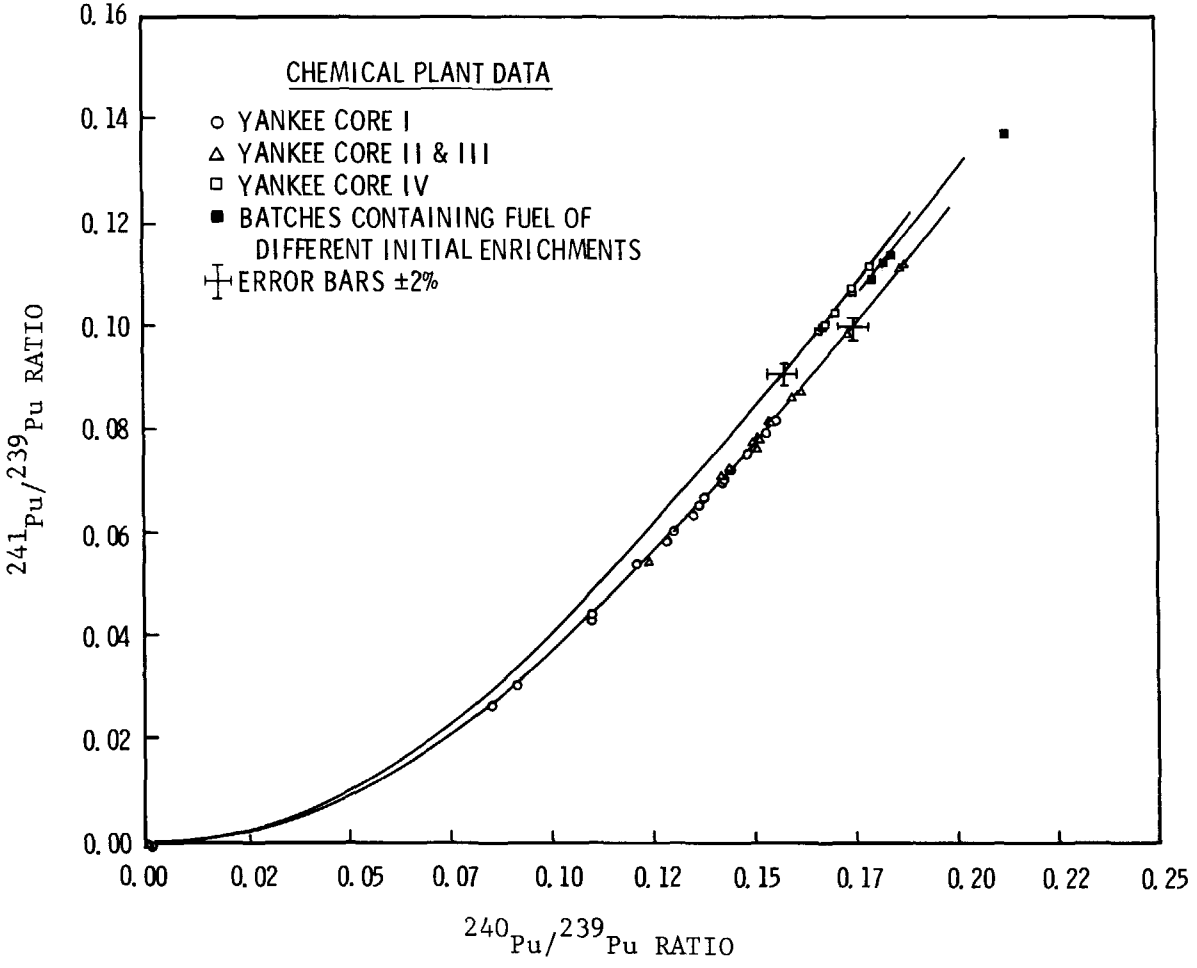

FIGURE 3. Plutonium Isotopic Data from Chemical Plant Measurements

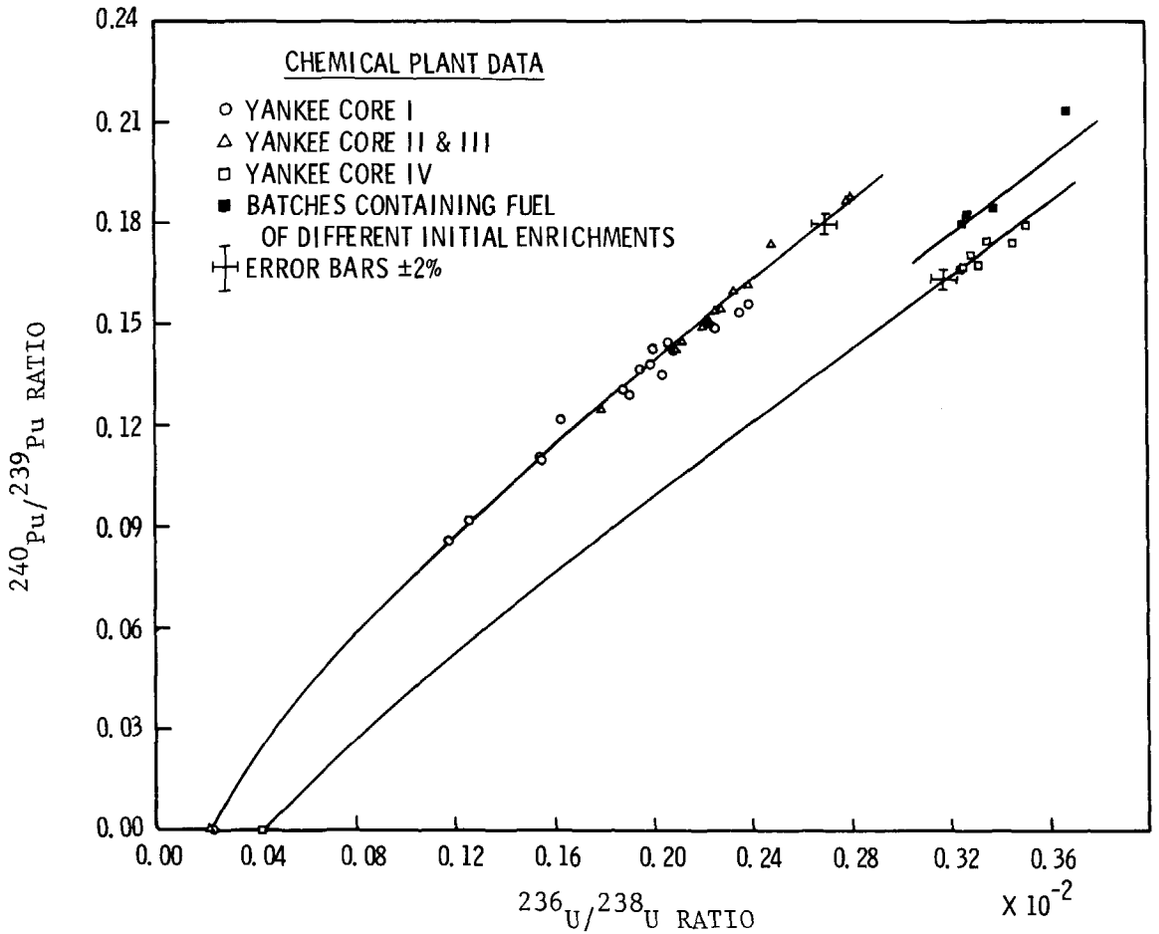

FIGURE 4. Uranium and Plutonium Isotopic Data from Chemical Plant Measurements

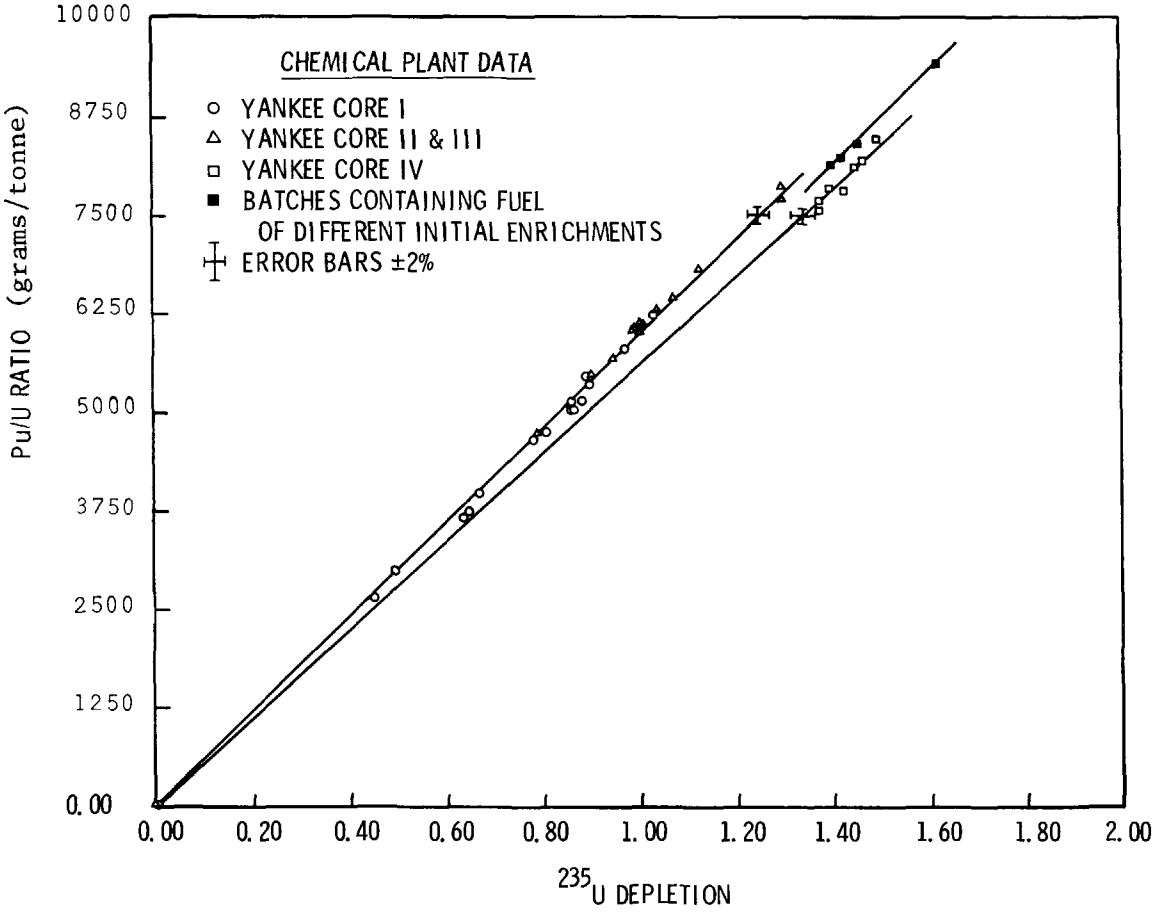

FIGURE 5. Pu/U Ratio as a Function of ^{235}U Depletion from Chemical Plant Measurements

from the plotted data data were well defined. This result was seen to provide a qualitative approach to evaluation of data consistency based on the following:

1) Burnup curves from zero burnup through the measured batch data points could be drawn by inspection of the plotted data.

2) The plotted data were seen to lie on the burnup curves or within a burnup curve envelope of ± 2 to 3%.

A qualitative indication of inconsistent data is obtained when a data point lies outside the burnup curve envelope. An example of data outside the envelope was the ^{239}Pu and ^{242}Pu values of batch number 43 of core IV.

Certain of the burnup relationships observed were essentially linear. This result was seen to provide a more quantitative approach to evaluate data consistency based on the following:

3) A numerical value representing the slope of a linear burnup function could be determined by determining the mean value from individual batch values of the slope.

4) A standard deviation for the individual batch values was also determined which gave an indication of the magnitude of the components contributing to the data consistency index.

An indication of questionable data is obtained when an individual batch value lies outside the two standard deviation values.

The above techniques thus serve to identify the individual measured batch data that are consistent with the burnup curves and consistent with the mean values determined from the collective data as well as those data that are inconsistent and thus questionable. The term data consistency analysis is therefore applied to the procedures used. In comparison to the 2σ value range of data consistency given for use with burnup data in section IV.A., it is evident that chemical plant data consistency is generally better such that one would use the following values:

	2σ VALUE		2σ VALUE
Pu/U Ratio	**1–3%**	**^{239}Pu**	**0.5–1.0%**
^{235}U	**0.5–1.0%**	**^{240}Pu**	**1.0–2.0%**
^{236}U	**1.0–3.0%**	**^{241}Pu**	**1.5–3.0%**
^{238}U	**0.2%**	**^{242}Pu**	**2.0–4.0%**

One reason that the above values are lower than those used for burnup data is that scatter due to reactor conditions is minimized when entire assemblies are dissolved in a batch.

Since the techniques provide information pertaining to measurement quality, independent remeasurement of every batch sample is not necessary to verify measurement accuracy. However, the procedures do not provide information concerning biases and a safeguards authority must plan to remeasure a random selection of samples to control possible biased results and to provide correct values when the data consistency procedures identify a questionable value. Thus, the techniques are economically advantageous by eliminating the need to remeasure every batch sample for accuracy verification.

It is important to note that the methods of plotting and deriving numerical slope values from chemical plant data are techniques that can be applied concurrent with the measurement of the data. Reliable information concerning data consistency is thus quickly available to a safeguards authority. However, use of the techniques are not restricted to safeguards and a chemical plant operator can conceivably use the techniques as a means of quality control. Chemical plant data should also be useful in the process of development and normalization of burnup codes[18] due to the fact that burnup relationships are evident from the data. This is not intended to mean normalization to core or campaign absolute totals only[4,5] but normalization of the calculated isotopic burnup paths to the isotopic burnup paths from chemical plant data as well.

We have restricted the discussion of one facet of isotopic correlations using isotopic data to measured data from one reactor. However, the methods discussed have been applied to measured data from several reactors and the methods have been seen to have broad application. Work has also been done by Stewart to quantify numerical parameters using least squares fitting methods.[19,20] Work is now underway to obtain quantitative information using a least square program specifically designed to analyze burnup data. The technique being applied utilizes the transmutation equations as incorporated in an approach developed by Matsen.[21] It is expected that the approach will realistically quantify the burnup curves and the numerical slope parameters as well.

To summarize, application of data consistency methods should be of significant value to a chemical plant operator as a means of improving quality control, to a fuel-reactor designer to aid in design code normalization and development and collectively to all concerned with nuclear materials safeguards in order to derive safeguards information and to minimize active surveillance.

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