

IN THIS ISSUE

- Nondestructive Assay Techniques for Recycled 233U Fuel for High-Temperature Gas-Cooled Reactors — J.E. Rushton, J.D. Jenkins, and S.R. McNeany, 18-35.
- Making Inferences about the Shipper's Variance in a Shipper-Receiver Difference Situation — John L. Jaech, 36-38.
- Development of a Complete System Determining Route Inspection Efforts and Timing for Fabrication Plants — Tohru Haginoya et al., 39-40.
- Some Thoughts on Constant and Variable Components of Systematic Error — S.C. Suda, 41-43.
- Some Thoughts on 'Some Thoughts on Random Errors, Systematic Errors, and Biases' by John L. Jaech — Roger H. Moore, 44-46.

NUCLEAR MATERIALS MANAGEMENT

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EDITORIAL

'Not to be **Undertaken Lightly'**

Dr. Higinbotham

By W.A. Higinbotham **Executive Editor**

For many years INMM has issued certificates to nuclear materials managers who met certain requirements for knowledge and understanding of the subject. This is an important responsibility, not to be undertaken lightly. If the criteria for certification are high and the procedures for certification are patently thorough and objective, the certificates may hope to gain credibility and acceptance from those segments of society who presently view INMM as an insider's group, automatically committed to defending nuclear power. Indeed, all of the members of INMM share in the responsibility which the Institute has should red. Its success or lack of success will be reflected in whether we as individuals are judged to be more concerned with the welfare of society or more narrowly with our personal welfare

In order to insure that the certification program is clearly defined, for all to see, and of the highest technical quality, the Institute has established a committee, headed by Fred Forscher, to redefine the certification procedure in the framework of The American National Standards Institute. This committee's report appears in a neighboring column and a draft standard will be distributed at the June meeting. This effort should receive critical attention from all of us.

The program addresses the responsibility of the individual materials manager to society. The materials manager or the safeguarder has a right to a guid-pro-guo. Consider the case of the BART engineers: While employed in design of The San Francisco Bay Area Rapid Transit System, they became aware of public hazards which were not being corrected. First they tried to work within the organization. Failing that they made a public statement and were fired. It took a long time to persuade the prestigious professional societies to come to their help. Eventually they did. The engineering societies are now working on standards for professional ethics for engineers and on standards to define the rights of engineers who work for industry or government.

Nucleonics Week of April 17 carries an item "Allegations of Nuclear Employee Intimidation Rise Again" which tells of (1) a nuclear engineer who was fired by Nuclear Services Corp. after criticizing some aspects of its QA program before a closed session of a Congressional committee, (2) a nuclear employee who was "dissuaded" from participating in a public forum by his employer and (3) an engineer who was fired for publicly calling attention to a sloppy job of containment coating on a reactor under construction.

When will an INMM member have to make that agonizing and potentially costly decision to embarrass his company or laboratory or government agency because his conscience says this is the right thing to do? The INMM budget is hardly prepared to take on big legal battles. I could, however, draft a bill of rights for safeguarders which would signal Institute support for the honest critic to the limits of our resources and of others that we might tap.-WAH

Only in exceptional circumstances, will the Journal publish such long and detailed articles as "Nondestructive Assay Techniques for Recycled U-233 Fuel for HTGR's," which appears below. At this time INMM cannot hope to compete with Science or Nuclear News. In this case, however, the article seems to be especially relevant to INMM. Recycle of U-233 may appear to be far down the road. But the technology is being developed now and this is the time to develop the safeguards techniques, not after the plans have been frozen for construction of the commercial facility. Safeguards measurement problems will also arise in connection with the reprocessing plant for HTGR fuels. Hopefully, a future issue will present this subject to Journal readers and to solicit their advice.-WAH



Mr. Soucy

Work of Standing Committees Important To Institute

By Armand R. Soucy

DYMAC, RETIMAC, ERDA, NRC--all acronyms which are an indication of the rapid developments in the area of safeguards. Sensational documentaries such as "The Plutonium Connection" and books such as "The Curve of Binding Energy" and "Nuclear Thefts and Safeguards" are examples of the sensational public furor over the issue of safeguards. How can the Institute of Nuclear Materials Management make a positive contribution in the "future shock" environment which exists in safeguards today?

This is an issue which your Officers and Executive Committee Members debated at length at our latest Executive Committee Meeting. There is no question that a professional organization such as INMM with a diverse membership of 400 individuals has difficulty in reacting quickly and promptly to documentaries such as "The Plutonium Connection." At this moment, the most effective rebuttals to public statements which distort the safeguards picture are those which are presented by you as individual professionals in our industry.

The Institute's overall contribution to the development of safeguards will probably never receive the public recognition which it deserves. However, in the long run it is our work with its slow but constant progress which is the foundation of a sound safeguards system.

Recently I have been tremendously impressed by the work of those of you who are involved in the various standing committees of the Institute.

STANDARDS—In the past six months, under the leadership of John Jaech, many INMM members have newly agreed to work as members of committees or as Committee Chairmen in the development of standards.

CERTIFICATION—The recent success of our Certification Committee whereby ANSI has accepted the Institute's certification program as an official ANSI Standard, is a major step forward in the progress of our certification program.

EDUCATION—In the area of education, the Institute's decision to fund for the first time the educational program of the Argonne Safeguards Center was admittedly made with a degree of trepidation. We were pleasantly surprised at the operational and financial success of our initial involvement in this program, and on the basis of that success we plan to again sponsor courses which will be offered in the Fall of 1975.

SAFEGUARDS—It is obvious that with the increased emphasis on safeguards in the nuclear industry the Institute must devote more resources in time and money to the development of new programs. It is with those goals in mind that your Officers have asked the Safeguards Committee under the leadership of Dennis Wilson to review the possibility of developing a public information program to present the facts on safeguards to our national leaders and the public. It is realized that the development of such a program will involve a major effort and in all probability require the use of outside consultants and other forms of assistance.

ANNUAL MEETING—Our 1975 Technical Meeting Committee has worked extremely hard to organize this year's program. As a result of their work, this year's meeting will make a major contribution to research and development in our nation's safeguards program.

The areas upon which I have touched are only the highlights of the activities of the various working committees of the Institute. Although we will never receive the recognition that sensational books and documentaries obtain from the public, we are convinced that the disciplines which we develop will become the basis of a sound national and international safeguards program.

A.R. Soucy

SECRETARY'S CORNER



Mr. DeVito

REPORT ON FEBRUARY INMM EXECUTIVE MEETING

By V.J. DeVito Secretary of INMM

The Executive Committee and several committee chairmen met for the winter meeting in New Orleans, Louisiana, on February 13 and 14, 1975.

Financial statements prepared by **Ralph Jones**, Treasurer, showed that the disbursements totaled \$12,426 for the first half of the fiscal year ending December 31, 1974. The annual budget for the fiscal year is \$23,900. Receipts for the sixmonth period totaled \$18,723. The savings account total was \$12,929. The financial statement for the INMM Journal, which is on a calendar year basis, showed a profit of \$2,053 for the year. The gain was due primarily to an increase in sales of subscriptions and advertising.

The ratification of Larry Dale to the Executive Committee was entered into the records. Larry Dale was appointed to the vacancy created by the resignation of **Curt Chezem**. The appointment was approved by letter prior to the meeting.

John Jaech and Dick Alto reported on N15 Standards activity. Thirteen standards have been approved and fourteen additional standards are in preparation. John Jaech has prepared two discussion papers for presentation at a meeting between the Nuclear Regulatory Commission and participants in industry standards program. One paper deals with the master plan for N15.

Gary Molen was appointed to represent INMM on Ad Hoc Group 5, Nuclear Safety and Security (Safeguards), which will set priorities for international standards.

The report from the Safeguards Committee was read and it was noted that the committee commented on the GESMO report. The committee will continue to participate in other safeguard projects. The primary interest is the preparation of an INMM safeguards brochure.

Harley Toy, Chairman of the Nominating Committee, stated that his committee would include Walt Martin, Bob Delnay, and Bill Donovan.

Manny Kanter and Ralph Jones reported on the success of the INMM safeguards school held at Argonne National Laboratory. The cost of the program was approximately \$12,300, and the tuition received was approximately \$16,000. There were twenty-one who attended the Advanced Concepts-Nuclear Material Control course and nineteen who attended the Statistics course. Nuclear Material Control, Statistics and a Guard Training course are planned for fall 1975. Fall and spring sessions were -authorized by the Executive Committee.

The Executive Committee approved the sending of the INMM Journal to all members of the Joint Committee on Atomic Energy.

The annual meeting for 1976, to be held at the Washington Plaza Hotel in Seattle, was tentatively set for June 23, 24, and 25, 1976.

After evaluating the report from the Site Selection Committee, the committee was instructed to consider Washington, D.C., Toronto, and Philadelphia as sites for the 1977 annual meeting.

Fred Forscher reviewed the status of the old and proposed certification programs. The new certification program will be standard oriented. An ANSI standard will be prepared under N15 writing group INMM 11 with Fred Forscher as Chairman.

The next Executive Committee meeting is scheduled for June 17, 1975, in New Orleans, Louisiana.

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Change Name to NUSAC

Dr. Lumb

Dr. Ralph F. Lumb, President of the Nuclear Surveillance and Auditing Corporation, a company established in 1968 to provide surveillance and auditing services during fuel fabrication and reprocessing, has announced that an official change of name has been effected.

Dr. Lumb said the stockholders voted to change the corporate name to NUSAC, Inc. to eliminate the narrow concept suggested by its former name. He noted that the current scope of NUSAC's activities is substantially broader than those originally contemplated: representation at the ERDA gaseous diffusion plants for UF6 procurement, quality assurance programs during fuel fabrication, and representation during chemical reprocessing.

NUSAC services today also encompass consultation and assistance on licensing matters, including all aspects of physical security for nuclear materials and for nuclear plants, out-of-core computerized fuel management systems, assistance on industrial safety, and material control and accountability systems.

The offices of NUSAC; Inc. are located at 7777 Leesburg Pike, Falls Church, VA 22043. Phone: (703) 893-6004.

BEGINS ACTIVITY ON FOUR FRONTS



By Dennis W. Wilson, Chairman

The INMM Safeguards Committee has been actively engaged in sweeping away the cobwebs of silence and nonparticipation. Since the last report, the Committee has begun activity on four fronts:

1) Panel Discussion at Annual Meeting—Under Bob Keepin's enthusiastic leadership, the 1975 Annual Meeting will be topped off by what is slated to be an exciting discussion on safeguards between information media, nuclear critics, and the nuclear industry. The Safeguards Committee is assisting in collecting material likely to be used during this discussion.

2) Preparing INMM Safeguards Information Medium— With full Executive Committee support, the Safeguards Committee is now compiling ideas and information to be used in preparing an INMM-sponsored safeguards information package. It has not yet been determined what the final form will be, but a printed brochure, slide-tape presentation and video tape are all under active consideration. The project is scheduled for completion by year's end.

3) Formation of INMM "Speaker's Bureau"-Lists are now being compiled of professionals who can and will represent

INMM and the industry in discussions on safeguards. Participation is needed on all levels ranging from congressional hearings or national TV debates to discussions with local groups. The initial list of available participants is expected to be ready by mid-summer.

4) Regulatory Guide Review—The Committee has recently been assigned responsibility for providing comments on Division 5 Regulatory Guides. Provided comments will be transmitted to the NRC via the ANSI nuclear program coordinator.

As is suggested by the above activities, the Committee needs input from capable and willing INMM members. Each Institute member is encouraged—no, solicited—to participate in these activities. This can be done by making input directly to a Committee member (named in the Journal's Winter issue). Better yet, the enthusiastic and aggressive are invited to join the Committee. For these daring and rare creatures, we invite you to contact the Committee Chairman directly. As a minimum, support your profession by keeping current. Active participation in INMM through the Safeguards Committee provides such an opportunity. Do something now before you forget!



N15 REPORT

ACTIVITY OF INMM NOT KEEPING PACE

By John L. Jaech, Chairman

In the last report of the N15 Chairman, it was pointed out that there had been many changes at the subcommittee chairmanship level in recent months. This trend continues but it is hoped that the situation has reached some degree of stability by now. In addition to those individuals named as new chairmen in the last report, we extend welcomes to Gene Miles, Chairman of INMM-1 (Methods of Nuclear Materials Control) and to Laird Hagie, Chairman of INMM-3 (Statistics). Subcommittee INMM-7 (Audit Techniques) is still without a chairman.

After extended communications with the leadership of ASTM Committee C-26, it was decided that N15 Subcommittee INMM-2 (Measurements) be dissolved because of conflicting scopes. The C-26 Committee has agreed to perform the necessary maintenance on the two standards developed by INMM-2. Measurements in the nondestructive assay area are covered by Dennis Bishop's INMM-9 Subcommittee.

After a fast and enthusiastic start in the standards-writing business, our professional organization has seemingly lost some of its enthusiasm, for the activity is not keeping apace of the needs. Although this inactivity is understandable because standards-writing can become a burdensome and often little appreciated activity, it is unfortunate that we as a group are apparently unable or unwilling to make a sustained effort in this area. I am hopeful that by the time this report appears in print, we will have renewed our efforts under the direction of the recently appointed subcommittee chairmen who join our experienced held-over chairmen. You, as individual members, can be a big help in this respect, for standards writing groups are sometimes hampered by having too few members who are willing to write and too many who are willing to comment. If you would like to help, and especially in a writing capacity, please contact the Chairman of N15 or the appropriate Subcommittee Chairman. Also, we always welcome your ideas for new standards.



N. N. C. Announces New Sensitive Water Monitoring System

A continuous discharge water monitoring system about ten times more sensitive than heretofore has been announced by National Nuclear Corporation of Redwood City, Calif. The first of these systems is now being installed in a nuclear plant by a leading electric utility to meet the increasingly stringent NRC limits for radioactive effluents.

The monitor is specially designed to reduce the buildup of radioactive contamination in the monitoring chamber. This side stream instrument can be located in low radiation areas or be shielded against normal nuclear plant radiation levels. This permits the detection and measurement of low concentrations of gamma emitting nuclides in the liquid against a low background.

The unit uses readily replaceable, disposable jars as counting chambers for a highly efficient, well-shielded, sodium iodide detector. Unlike conventional stainless steel counting chambers, these jars have a very low tendency to become contaminated or accumulate fine sludge deposits. Because of the construction of this unit, the disposable jar and top shield can easily be replaced without tools in less than 5 minutes downtime. This jar is replaced whenever the background count rate on a water purge shows an appreciable increase. Ready replacement is possible because the upper shielding is split for easy removal, while the flow enters and leaves the monitor through flexible tubing.

New Book on Thorium

Released by I. S. U. Press

AMES, IOWA—The search for alternative power sources will surely include consideration of a role of thorium. Valuable information about this element, of interest to nuclear reactor technology, is available in **THORIUM: Preparation and Properties**, just published by Iowa State University Press.

The authors, J.F. Smith, O.N. Carlson, D.T. Peterson, and T.E. Scott, professors of metallurgy at Iowa State University and senior metallurgists for the Ames Laboratory of the AEC, have joined forces to update and bring together current knowledge concerning the preparation, properties, and behavior of the metal thorium.

The last book on this subject was **THE METAL THORIUM**, published in 1958 by the American Society for Metals. Since that time a great deal of knowledge has become available. Within the past ten years techniques have been developed for preparing thorium of significantly greater purity than had previously been possible. This change in purity has made notable differences in certain characteristics of thorium behavior. In addition, the available thermodynamic data have increased fivefold.

Chapters include: preparation; purification; annealing; mechanical forming, and other fabrication processes; mechanical properties of thorium and its alloys; diffusion in thorium; physical properties; and alloying behavior. **THORIUM** provides a critical review of published information on the phase diagram, crystallography, and thermodynamic properties of 71 binary systems of thorium.

With the increased interest in breeder reactors thorium takes on greater importance as a "fertile" material. For this reason **THORIUM** is timely and relevant to today's energy crises.

Of special interest to people concerned with nuclear reactor technology as well as materials scientists and engineers, **THORIUM: Preparation and Properties** is available from bookstores or from the Iowa State University Press. (ISBN 0-8138-1635-1, 382 pp., illus., \$9.95.)

Education Report



Three Courses Available Next November

Dr. Kanter

By Manuel A. Kanter, Chairman

As a result of the success of the two courses that the Institute sponsored at Argonne last November, the INMM Executive Committee has decided to continue its education program by the sponsorship of additional sets of courses in the fall of 1975 and in the spring of 1976. The first of these is to be a fundamental set aimed at persons who are new in nuclear material control whereas the latter set will be of an advanced nature for persons who are already working in the field.

The courses for the fall have been scheduled as follows:

- November 3-7, 1975 Introductory Statistics as Applied to Measurement Quality Control. Course Leader, Richard J. Brouns, Pacific Northwest Laboratory.
- November 10-14, 1975 Fundamentals of Nuclear Material Control. Course leader, James E. Lovett, International Atomic Energy Agency.
- November 17-21, 1975 Guard Forces Their Role in Nuclear Materials Security. Course leader, Joseph J. Indusi, Brookhaven National Laboratory.

The courses for spring have not yet been determined but we are considering courses in advanced measurements, advanced statistics, and advanced concepts in nuclear material control. They will probably be given in May. However, I would appreciate suggestions from the membership as to courses which they see to be needed.

There is additional interest in training in nuclear material control. Los Alamos has plans for an additional course in measurements and the Institute is holding discussions with IAEA concerning possible co-sponsorship of courses in nuclear material control and safeguards for the growing staff of countries setting up systems of control for the first time.

INMM Reports 32 New Members

The following 32 individuals have been accepted for INMM membership as of May 27, 1975. To each, the INMM Executive Committee extends its congratulations.

New members not mentioned in this issue of the Journal will be listed in the Summer 1975 (Volume IV, No. 2) issue to be mailed in late July or August.

- Thomas L. Atwell, Staff Member, Group A-1, University of California, Los Alamós Scientific Laboratory, P.O. Box 1663, Los Alamos, NM 87544.
- Harvey C. Austin, SS Control Supervisor, Oak Ridge National Laboratory, P.O. Box X, Oak Ridge, TN 37830.

Roy W. Brown, Manager, Technical Division, Goodyear Atomic Corporation, P.O. Box 628, Piketon, OH 45661.

Milton H. Campbell, Senior Fuel Reprocessing Engineer, Exxon Nuclear Company, Inc., 2955 George Washington Way, Richland, WA 99352.

- Chih Ping Chen, 60-A Van Wyk Road, Lake Hiawatha, NJ 07034.
- Phillip J. Cherico, 46 East Welling Avenue, Pennington, NJ 08534.
- Charles William Emeigh, Babcock & Wilcox Company, Nuclear Materials Division, 609 North Warren Avenue, Apollo, PA 15613.
- Gerald G. Fain, Regional Manager, Licensing & Regulation, General Atomic Company, 2021 K Street, N.W., Suite 709, Washington, D.C. 20006.
- Ralph R. Fullwood, Scientist IV, 742 Torryea Court, Palo Alto, CA 94303.
- Rudolph Gatti, Manager of Accountability Safeguards, Canberra Industries, 45 Gracey Avenue, Meriden, CT 06450.
- Ron L. Hawkins, NDA Specialist, Nuclear Fuel Services, Inc., Carolina Avenue, Erwin, TN 37650.
- **Rupert E. Henry**, Manager of Health Physics, General Electric Company, STGP Division, Building 273, Room 299, Schenectady, NY 12345.
- Steven C. Johnson, Construction Budget Analyst, Arizona Public Service Company, P.O. Box 21666, Station 3003, Phoenix, AZ 85036.
- James L. Karalus, Fuel Engineer, Northern States Power Company, 414 Nicollet Mall, Minneapolis, MN 55401.
- Hugh Kendrick, Division Chairman, Science Applications, Inc., 1600 Anderson Road, McLean, VA 22101.

- Robert A. Kramer, Nuclear Fuel Engineer, 311 West 75th Place, Merrillville, IN 46410.
- James C. McCue, 2555 PGA Boulevard, No. 83, Palm Beach Gardens, FL 33410.
- Charles J. McKenna, Standards Engineer, ITT Grinnell Corporation, 260 West Exchange Street, Providence, RI 02901.
- F. Morgan, Procurement Executive, Ministry of Defence, Atomic Weapons Research Establishment, Building A1.2, Aldermaston, Reading, RG7 4PR, England.
- Nicholas Ovuka, 6215 Waterway Drive, Falls Church, VA 22044.
- **Oscar B. Parker,** National Coordinator, Management Consulting Service, Burns International Security Services, Inc., 1681 J.F. Kennedy Causeway, Miami, FL 33141.
- Charles E. Pietri, Chief, Analytical Chemistry Branch, New Brunswick Laboratory, U.S. ERDA, 13 Joan Street, Kendall Park, NJ 08824.
- Garry L. Quinn, Director of Marketing Reqt's., Boeing Engineering & Construction, P.O. Box 3707, MS 8C-12, Seattle, WA 98124.
- Robert M. Radford, Superintendent, Security & Safeguards Department, E.I. du Pont Nemours, Savannah River Plant, Aiken, SC 29801.
- Mohammed Riaz, Engineering Manager, Nuclear Valves, Weston Hydraulics, 7500 Tyrone Avenue, Van Nuys, CA 91409.
- Charles Herbert Sathrum, Senior Engineer, EDS Nuclear, Inc., 220 Montgomery Street, San Francisco, CA 94104.
- **David B. Sinden**, Scientific Advisor, Atomic Energy Control Board of Canada, P.O. Box 1046, Ottawa, Ont., Canada KIP 559.
- Robert J. Slough, Senior Process Engineer, Catalytic, Inc., 1500 Market Street, Philadelphia, PA 19101.
- Bill R. Teer, Director of Marketing, Transnuclear, Inc., One North Broadway, White Plains, NY 10601.
- John L. Telford, Statistician, Nuclear Fuel Services, Inc., Carolina Avenue, Erwin, TN 37650.
- Charles B. Yulish, President, Charles Yulish Associates, 229 Seventh Avenue, New York, NY 10011.
- Samuel M. Zivi, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439.

MEMBERSHIP REPORT



MANY CONTACTED ABOUT

By James W. Lee INMM Membership Chairman

The years 1974 and 1975 saw a major change take place in the composition of the Membership Committee and in its method of doing business. In an effort to streamline Membership Committee procedure, the Treasurer, Ralph Jones, and Secretary, Vincent DeVito, together with the Membership Committee Chairman, James W. Lee, were delegated as the Membership Committee.

Applications and checks are sent first to the Treasurer, who indicates his approval of the applicant, thence to the Secretary for additional approval and entry into the INMM membership roster, and finally to the Membership Committee chairman who issues the membership card and a letter of welcome. The letter welcoming the applicant into the Institute membership also describes some of the activities of the Institute and suggests how the new member can participate in committees or activities of interest to him.

Routine inquiries are forwarded to the Membership Committee Chairman who responds with information about the Institute and an application form, and follows up the inquiries regularly for a period of time.

The Institute pamphlet and application form had not been revised for several years so the committee, with a very strong assist from Tom Gerdis, Editor of the Journal, and with help and suggestions from the Executive Committee and officers, completely revised the INMM pamphlet and application form in 1975.

Invitations to join the Institute have been mailed to several groups and others are in the process of preparation. The first study courses in nuclear material management sponsored by the Institute, which were conducted by Dr. M.A. Kanter of the Argonne Center for Educational Affairs, produced a fertile field for new memberships, and with Dr. Kanter's cooperation special invitations were forwarded to all students completing the course. A number have responded and submitted applications for membership in the Institute.

The Institute Treasurer, Ralph Jones, was instrumental in obtaining a large listing of individuals in the nuclear industry

who are engaged in activity that would cause them to give consideration to Institute membership and information and invitations to join have been distributed to this listing.

The Institute's dynamic Technical Program Chairman, Dr. G. Robert Keepin of Los Alamos Scientific Laboratory, is conducting a series of technological meetings for representatives of the foreign nuclear industry and through his cooperation a special invitiational letter to join the Institute was tailored for forwarding to these individuals.

The increased emphasis upon physical security and safeguards within the nuclear field, and the resultant direction of Institute attention to these subjects, created a large new class of potential new members. At the present time, the Membership Committee is developing a mailing list of persons who are engaged in security work in the nuclear industry and devising a special letter of invitation to be sent to these individuals.

In the last analysis, however, the real growth of any organization depends upon the continued and enthusiastic interest of its members towards locating and encouraging their friends, colleagues and co-workers to join the Institute and to participate actively in its affairs.

If every member reading this article would stop at this moment and reflect for a minute or two, he surely would think of a minimum of at least two or three individuals who undoubtedly would join the Institute if approached. Do this today.

Then send the Membership Committee the names and addresses of persons you believe would be interested in knowing about the Institute's activities and who might consider joining the Institute. Send your prospects' names to James W. Lee, Chairman, INMM Membership Committee, P.O. Box 14336, No. Palm Beach, FL 33408.

Persons whose names are submitted will promptly receive information about the Institute, a cordial invitation to join it and the initial invitation will be followed up on a regular basis.

CERTIFICATION OF NNM'S



PRE-ANNUAL MEETING REPORT

By Dr. Frederick Forscher, Chairman INMM Certification Committee

Editor's Note: Progress Report of INMM-11, a committee to establish "Criteria and Standards for the Certification of NMM's"

Nuclear fuel has become an object of commerce and international trade. Special Nuclear Materials (SNM) are a commodity of great economic significance and with national security implications.

Individuals who, by virtue of their position in government or industry, make decisions about the disposition and utilization of nuclear materials are not only affecting corporate profitability, but also public safety, environmental quality and national security. Such individuals are in fact members of a new profession.

Public recognition of this profession can be enhanced by (1) criteria and standards that define the professional requirements; (2) an accredited method for certification for such professionals; and (3) regulatory requirements, both national and international, that call for the employment of such professionals.

The fields of knowledge that constitute the core of this new profession are (a) materials control and accounting, often referred to as "Accountability," (b) material and plant protection, often referred to as "Safeguards," and (c) the quality assurance provisions that must be employed to develop public confidence and credibility.

The INMM has for years recognized the significance of this profession. The institute issued "Certifications" of Nuclear Materials Managers since 1963. Seventy-five certificates have been granted up to this time. Any further certification awaits the results of the efforts of the INMM-11 committee.

In mid-1974, the INMM executive committee felt the need to put certification of NMM's on a more formal and accredited basis. Consequently, Dr. Frederick Forscher, the new Chairman of INMM's certification committee applied for an ANSI Charter. The Nuclear Technical Advisory Board of ANSI approved the application.

The revised scope of the Charter reads: "This standard defines the requirements for "Certification" as Nuclear Materials Manager or as Nuclear Materials Specialist. It sets forth the program scope in the fields of (a) material control and accounting, (b) material and plant protection, and (c) quality assurance in which a certified individual must demonstrate proficiency and competency. The certification procedure shall be administered by a certification board that meets ANSI's accreditation requirements."

The certification program must be open to all qualified persons, independent of race, creed, sex or nationality, Acceptance criteria must be based on performance and demonstrated knowledge and understanding of the subject content of this profession. The criteria for certification do not relate to, or consider, an applicant's ability or inability to obtain government clearance or access authorization to nuclear materials. Such matters are a job-related consideration and strictly a matter of concern between the individual applicant and the government agency having jurisdiction in such matters.

The following two sections (Draft No. 1) may be of particular interest to the membership of the INMM. A complete (Draft No. 2) standard will be available to the membership at the Annual Meeting in June 1975. Comments and questions about this standard should be directed to Frederick Forscher (6580 Beacon St., Pittsburgh, PA 15217).

Sect. 3.0 Basic Requirements for Certification.

A candidate for certification as a nuclear materials manager shall have the following prerequisites:

3.1 Degree from a college or university in an appropriate field, such as: Chemistry, Physics, Statistics, Engineering, Accounting, Law Enforcement, Business Administration, Economics, or Mathematics; or a minimum of five years' experience in one or more of the above or other fields which are pertinent to the management of nuclear materials.

The Certification Board reserves the right to request substantiation by completion of the Graduate Record Examination taken within two years before the date of the application.

3.2 A minimum of three years of diversified responsible experience in nuclear materials management.

3.3 Familiarity with the basic concepts of health physics and radiation safety, environmental protection, and criticality prevention unique to the nuclear industry.

3.4 The endorsement of a sponsor, recognized in the nuclear community, who can attest to the candidate's professional or technical capabilities in nuclear materials management. Additional supportive statements as to the candidate's maturity, reliability, integrity and responsibility are optional.

3.5 Satisfactory completion of an examination designed to explore the applicant's qualifications for certification, administered by the Certification Board for Nuclear Materials Managers.

Sect. 6.0 Acceptance Criteria and Administration

6.1 The determination of a candidate's qualifications for, and the issuance of, a "certificate" are the sole responsibility of the Certification Board for Nuclear Materials Managers. The charter and bylaws of this certification board are appended to the standard (Appendix A). This appendix is not yet drafted.

6.2 A "Certified Nuclear Materials Manager" shall have demonstrated acceptable competence in the fields of (a) (Continued on page 17)

GUEST EDITORIAL



Dr. Frederick Forscher (right), a member of the INMM Executive Committee and chairman of the Institute's Certification Committee, is an articulate spokesman for the INMM in his home area of Pittsburgh, Pa. This journal welcomes contributions of members who have letters published in their local newspapers. In fact, the INMM Executive Committee has gone on record as encouraging this sort of activity to make more persons aware of the valuable contributions INMM members are making in the field of Nuclear Materials Management. This photo was taken during the 1974 INMM annual meeting in Atlanta.—Tom Gerdis, Editor.

SHORTAGE OF VISION!

By Dr. Frederick Forscher

The era of shortages has crashed into our consciousness via the energy crisis. But energy is not the only shortage. There are, first of all, the shortages of raw materials such as steel, copper, bauxite, fertilizers and lumber. There are, next, the shortages of food, the scarcity of virgin land, open beaches, fresh water, fishing grounds, etc. Last but not least, there are the shortages of the spirit: credibility, political honesty, religious faith, national goals, personal commitments.

Of all the shortages mentioned, the shortage of the spirit is the most fatal. Societies have survived all other shortages that befell them in the past, all but the lack of vision, of an ethos of their time. "Without vision, peoples perish" goes a well-known phrase.

Shortages are not unique to the United States. The world as a whole is moving from an era of abundance in natural resources into an era of scarcity. Economists talk of the coming 'Spaceship Economy.' The world has never faced such a situation before; certainly never on a global scale. Past experiences may not be the best guide for future actions; past solutions do not apply today.

Perhaps, people are able to perceive only one problem at a time. After the energy crisis there was Watergate, and now we have world-wide inflation and a national recession. But the earlier problems have not gone away; the energy crisis is still with us, and so is the crisis of the spirit that led to Watergate.

We have deluded ouselves too long with the idea that THE solution to all conditions of shortage is money. We have lived under the assumption that money alone could fix nearly everything, as long as there was no shortage of money. But now, with inflation running rampant all over the world, it is obvious that merely printing more money—so that we can buy all the things we want—does not really solve our problem of shortages. In fact, it fans the fires of inflation. The only help for the near future is cutting back on demand. We all will have to become more conservation minded. Conservation must become an integral part of the ethos of survival, the new ethos of our time.

We must begin not only to **understand** the need for conservation but also to **feel** its significance. The idea that energy cannot be recycled must become as much a part of us as the 'feel' of time has become part of our being. We must reach an equilibrium between energy supplied to our spaceship Earth, and the energy that we as its passengers use for all our (social) purposes. "Support your local planet" is an appropriate slogan.

The more advanced, more energy intensive societies are clearly in a transition phase to the steady state economy. This transition may take 100 years or so. It is the period when we move from our present capital intensive economy to an income intensive economy in terms of energy. Energy capital are all non-renewable fuels such as coal, oil, gas and uranium; energy income derives from all renewable energy sources such as wind, tides, hydroelectric power, lumber, food, and of course solar energy itself.

The integration of the many aspects of conservation into our laws and regulations, into our mores and habits, and eventually into our tradition and ethos, will take a long time. Congress will struggle with it for years. We must aim now to reach the consumer and the public at large because, whether we know it or not, we are all involved. Only participatory action will get us there.

Remember, energy is the only 'commodity' that consumers truly consume. Once energy is used, it's gone forever, like the time of day.

Like a person about to go on a diet, the sooner we start, the sooner we feel the benefits. The time to start is long overdue.



Yvonne Ferris Promoted at Dow

Ms. Ferris

Yvonne Ferris, supervisor of the four-member statistical laboratory at the Rocky Flats Division of Dow Chemical U.S.A., Golden, Colo., assumed additional responsibilities for nuclear materials control Jan. 27. She succeeded **Bruce Bowman** who has accepted a position with Dow's Louisiana Division.

Ed Young, manager of production and nuclear materials control, said, "Yvonne has all the skills to get the job done, including the management and technical know-how, and a deep, long-standing involvement in safeguards and nuclear materials management."

An employee of Dow for 18 years, Mrs. Ferris has presented papers at the last four INMM annual meetings. As supervisor of the statistical laboratory, she is responsible for all plant statistical support, and her statistical laboratory staff will eventually number four or five.

Mrs. Ferris holds a bachelor's degree in statistics from Iowa State University, Ames, and upon graduation, joined Dow Rocky Flats as a statistician. While working with Dow, she has advanced through the positions of statistical specialist and senior statistical specialist.

A resident of Wheat Ridge, Colo., Mrs. Ferris is a member of INMM and the American Society of Quality Control. She is also a volunteer with the Gilpin House Project in Denver, for emotionally disturbed adults, a unit chairman for the League of Women Voters, and a member of the Denver Altrusa Club.

Along with J.R. Geoffrey and S.C. Suda, she is co-author of a paper to be delivered at the 1975 annual meeting of INMM. That paper is entitled, "Some Things We Should Have Known About Calculating LEMUF—But Didn't."

New Book on Pressure Vessels

Theory and Design of Modern Pressure Vessels, Second Edition by John F. Harvey. 429 pages plus index; approximately 280 illustrations; 6 x 9; Van Nostrand Reinhold; \$19.95. Publication date: July, 1974.

With the uses and requirements of pressure vessels in industry today growing at an unprecedented rate, this practical book—fully revised and updated—is more than ever an indispensable reference and guide in its field. It presents the actual design of vessels from material selection, stress theory, and design practice to the economics of construction, fabrication, and shipment.

The increased importance of the chemical, nuclear, space, and cryogenic industries has created increased economic and engineering demands for high pressure and large diameter vessels of reduced weight and cost, along with enhanced erection and shipping procedures. The designer must provide a vessel not only of maximum reliability, but also of minimum weight justified on the basis of a careful stress analysis of the entire structure.

Written with a full understanding of the designer's needs, this book details the first steps and practical considerations to be encountered in pressure vessel theory and design. It encompasses the evaluation of primary and secondary stresses in vessels; the significance of these stresses as they arise from pressure, temperature, fatigue, creep, and abrupt changes in section; and the compatibility of these forces with the environmental behavior of the material.

Important design-construction features receiving thorough coverage are the value and meaning of stress concentration factors encountered in vessels at openings, nozzles, supports, and attachments; as well as those to cope with thermal shock and brittle fracture. The author clearly sets forth economic considerations, including basic cost reduction tools available to the designer, with three principal approaches to achieve that goal. Analogies, examples, and models are used in abundance to illustrate the practical application of basic theory to effective vessel design.

John F. Harvey has been associated with the Babcock & Wilcox Company, Barberton, Ohio, in engineering and managerial capacities, and has conducted courses in pressure vessel design at the University of Akron. Mr. Harvey has written numerous technical papers, is a Licensed Professional Engineer in California and Ohio, and holds many patents for developments in pressure vessel constructions and heat exchangers. He is Chairman of the Pressure Vessel Research Committee, and is a member of numerous American Society of Mechanical Engineers committees on nuclear power, pressure vessels, and other code-making bodies.

INMM Certification Report

(Continued from page 14)

material control and accounting, (b) material and plant protection, and (c) quality assurance.

For certification as Nuclear Materials Manager, the applicant's test score in all three fields shall exceed a percentage as fixed by the board.

6.3 A "Certified Nuclear Materials Specialist" shall have demonstrated acceptable competence in at least one of the three fields of (a) material control and accounting, (b) material and plant protection, and (c) quality assurance.

For certification as a Nuclear Materials Specialist, the applicant's test score for the field of his or her specialty shall be in excess of the percentage fixed in Sect. 6.2, and a minimum percentage as fixed by the board in the other two fields.

6.4 A "Certification" becomes void five years after date of issue unless renewed by the board.

Application for renewal of the certification shall be accompanied by evidence that the applicant practiced this profession continuously during the past five-year period. An interruption of more than 12 consecutive months during this five-year period shall be deemed to constitute noncontinuous service.

If the Certified Nuclear Materials Manager has a period of non-continuous service in his professional capacity, the Certification Board shall determine the need for reexamination.



NONDESTRUCTIVE ASSAY TECHNIQUES FOR RECYCLED ²³³U FUEL FOR HIGH-TEMPERATURE GAS-COOLED REACTORS*

BY J.E. RUSHTON, J.D. JENKINS, AND S.R. MCNEANY Reactor Division, Oak Ridge National Laboratory

ABSTRACT

Nondestructive fissile material assay techniques are reviewed for application to the refabrication of recycled ²³³U fuel for High Temperature Gas-Cooled Reactors (HTGR). The refabrication processes and fuel characteristics are identified and explained, and nondestructive assay techniques are evaluated in light of these characteristics and assay requirements. The study concludes that currently developed and implemented gamma-ray assay techniques for ²³⁵U and ²³⁹Pu are not applicable to ²³³U fuel because of its high radioactivity. Calorimetry has potential for ²³³U fuel assay if appropriate corrections for isotopic composition can be applied. For most nondestructive assay requirements, active interrogation methods must be employed. Neutron interrogation systems using either ²⁵²Cf or ¹²⁴Sb-Be neutron sources and delayed or prompt fission neutron detection offer the greatest potential for assay of HTGR recycled ²³³U fuel. The presence of hydrogen in some of the fuel forms during processing and the particulate nature of the HTGR fuel complicate the direct application of these neutron interrogation techinques. Photofission techniques, which offer enhanced penetrability for the assay of bulk fuel quantities or fuel elements, cannot be evaluated for this application because of a lack of basic data.

A. INTRODUCTION

The High Temperature Gas Cooled Reactor (HTGR) operates on a combined ²³⁵U-Th and ²³³U-Th fuel cycle. HTGR's are initially fueled with highly enriched (93% ²³⁵U) uranium and thorium. The ²³³U which is bred from the thorium is recycled to the reactors during later refuelings. In the equilibrium fuel cycle ²³³U accounts for 30 to 40% of the makeup fuel. This high conversion efficiency decreases the requirements for uranium ore, substantially reduces separative work requirements, and utilizes thorium resources. These advantages tend to reduce overall fuel cycle costs and enhance the long term price stability of the fuel cycle.

The Oak Ridge National Laboratory (ORNL) is currently participating in the National HTGR Fuel Recycle Development Program¹ with the objective of developing and demonstrating the recycle technology necessary for the construction, licensing, and operation of commercial reprocessing and refabrication facilities for HTGR fuels. The refabrication portion of the development program is focused on the construction and operation of an HTGR Fuel Refabrication Pilot Plant at ORNL. Refabrication is defined as the production of HTGR fuel elements containing ²³³U as the primary fissile nuclide. The ²³³U can be obtained from the reprocessing of fuel or blanket materials from HTGR's, Gas Cooled Fast Reactors (GCFR), or Light Water Breeder Reactors (LWBR) in other facilities.

This paper addresses the selection of nondestructive fissile material assay techniques for use in HTGR fuel refabrication facilities. This work represents one portion of the development efforts associated with nuclear material management and safeguards for the refabrication portion of the program. Many of the parameters concerning nondestructive assay (NDA) of ²³³U fuels are also applicable to the refabrication of ²³³U-Th fuel for the LWBR; however, the particular fuel characteristics and refabrication processes described here are representative of the HTGR system only. The fuel forms and processes encountered in HTGR fuel refabrication differ dramatically from LWR

^{*}Research sponsored by the U.S. Energy Research and Development Administration under contract with the Union Carbide Corporation.

or LMFBR fuels; and, in addition, the nuclear characteristics of the ²³³U recycled HTGR fuel are different from the fresh or recycled ²³⁵U fuel for the HTGR. The unique characteristic of ²³³U fuel which separates it from other reactor recycle fuels is its high gamma radiation due to the inclusion of trace amounts of ²³²U and its daughter nuclides. The intensity and great penetrability of this gamma radiation require that the entire refabrication be conducted remotely behind thick shielding (e.g., 2-3 ft of concrete). The remote nature of the process and the consequent difficulty in achieving access to the special nuclear material adds to the level of security for safeguards; however, it complicates the requirements for material accountability and nondestructive assay during processing.

In view of the unique fuel characteristics, the physical properties of the fuel and the proposed refabrication process are described in Sections B and C, respectively. Then in Section D the characteristics of the fuel and processes which affect the selection of NDA methods are examined in detail. Specific NDA requirements for a refabrication facility are identified in Section E, and NDA techniques are reviewed in Section F in light of these requirements and the ²³³U fuel characteristics. Finally, conclusions and specific recommendations for development needs are enumerated.

B. DESCRIPTION OF HTGR RECYCLED FUEL

The HTGR fuel element, shown in Figure 1 is a hexagonal graphite block with coolant holes and blind fuel holes. The fuel is a blend of three types of particles bonded together by a graphite matrix. The fissile particle has a low density kernel of uranium carbide-uranium dioxide and carbon surrounded by four coating layers. The coatings,



Figure 1. 1160 (MW(e) HTGR Fuel Element (GAC Drawing).

Table 1

Typical Physical Parameters of the HTGR
Fissile and Fertile Particles
(Average Values)

	Fissile	Fertile
Kernel diameter, um	370	500
Kernel density, g/cm ³	3.2	10.0
Kernel composition, wt %		
Uranium	82.0	
Thorium		87.9
Carbon	15.2	
Oxygen	2.8	12.1
Coating thickness, µm		
Buffer	50	85
ILTI ^a	35	
SiC	30	
OLTI ^b	35	75
Coating density, g/cm ³		
Buffer	1.1	1.1
ICTIa	1.95	
SiC	3.2	
OLTI ^b	1.95	1.95

^a ILTI stands for Inner Low Temperature Isotropic and is the abbreviation used when referring to the inner high-density isotropic carbon coating.

^b OLTI stands for Outer Low Temperature Isotropic and is the abbreviation used when referring to the outer high-density isotropic carbon coating.

from the kernel out, are: (1) a low density buffer layer of pyrolytic carbon, (2) a high density isotropic layer of pyrolytic carbon, (3) a silicon carbide layer, and (4) another layer of high density isotropic pyrolytic carbon. The fertile particle has a ThO₂ kernel surrounded by a low density buffer layer and single layer of high-density isotropic pyrolytic carbon. The third particle type is pure graphite used as a shim for varying fuel content. The typical sizes, densities, and compositions of the fissile and fertile particles are shown in Table 1. The values in Table 1 are a current design basis and are subject to change due to reactor physics requirements or thermal and irradiation performance testing. The particles are not inserted directly into the graphite block but are first molded with a graphite flour and coal tar pitch into fuel rods. The molded rods will be either 0.5 in. in diameter and 1.94 in. long or 0.62 in. in diameter and 2.5 in. long for use in either the Fort St. Vrain Reactor or larger sized HTGR's, respectively.

C. DESCRIPTION OF PROPOSED REFABRICATION PROCESS

The fissile material enters the refabrication process in the form of clean uranyl nitrate solution (approx 125 g/ liter) stripped of fission products and recently cleaned of 232 U daughter nuclides. The uranyl ions are loaded onto weak acid, ion-exchange, resin beads by contacting the resin with an acid deficient uranyl nitrate solution. The loaded resin beads are dried and transferred to a furnace where the beads are carbonized by heating to 600°C to decompose the constituent hydrocarbons and drive off the volatile components. The carbonized resin beads which consist of UO_2 suspended in a porous carbon matrix are then passed to a fluidized bed furnace where in sequential operations the carbonized beads are converted at 1750°C to a $UC_2 + UO_2 + C$ kernel and the four layers of coatings are deposited. Following coating, the particles move to the fuel rod fabrication step where they are blended with the coated ThO₂ particles and inert graphite particles and dispensed into a rod mold. The molds are then injected with a matrix of heated pitch and graphite filler which permeates the interstices between particles to form a solid fuel rod.

In the final process step, the rods are loaded into blind fuel holes in the graphite fuel blocks, and the block is heat treated at 1700–1800°C to carbonize the pitch in the fuel rods so that the fuel particles are bonded with a graphite matrix. The last operations include block cleaning, the insertion of indexing dowels, inspection, and canning of the element for shipment.

The processing of the fuel from uranyl nitrate to fuel elements is completed before the uranium achieves an age of 90 days since the daughter nuclides were separated. The 90-day limitation is imposed so that small samples (approx 0.5-1.0 g) of material can be routinely handled in semiremote facilities for quality control and quality assurance inspections operations. Without using the time limitation, sample handling would also need to be done in totally remote facilities. The high sample rates required in a commercial refabrication facility would then require large investments for remote laboratory facilities.

In the pilot plant to be constructed at ORNL, the entire process from resin loading through fuel element carbonization will be housed in hot cells in the Thorium Uranium Recycle Facility (TURF), an existing hot cell complex at ORNL.² Figure 2 is a plan view of the TURF hot cells showing the location of four major process cells (C, D, E, and G), the decontamination cell (B), and a maintenance cell (A). Cells A, B, C, and D are serviced by an interconnected set of cranes and electromechanical manipulators. In addition, a number of the viewing windows are equipped with master slave manipulators. The walls of all shielded cells have shielding equivalent to 5.5 ft of normal concrete (a facility specifically designed for refabrication of HTGR recycle fuel would require only 2 to 3 ft of concrete at the processing level and somewhat less at other cell heights).



Figure 2. Floor Plan of HTGR Fuel Refabrication Pilot Plant.

Quality control and quality assurance samples taken from the process cells are pneumatically transferred to an adjoining building which will contain the sample inspection laboratory.

The purpose of the HTGR Refabrication Pilot Plant is to develop and demonstrate the processes, equipment, and procedures necessary to allow establishment of a commercial sized HTGR fuel refabrication facility. In keeping with this objective, the pilot plant is envisioned as a facility in which all aspects of operation including material handling, process interfacing, and plant control can be demonstrated on prototypic or scalable equipment. The design objective of the pilot plant is the production of 21/2 reactor fuel elements per day. This output corresponds to the successful processing of approximately 25 kg of heavy metal per day (2.5 kg of ²³³U). By comparison, a commercial facility designed to support a 20,000 MW(e) HTGR economy would, at equilibrium, be required to process approximately 400 kg of heavy metal or 40 kg of ²³³U per day assuming a 60% plant availability factor. Despite the disparity in total capacity, the requirement for a demonstration of prototypic equipment can be achieved in the pilot plant because much of the equipment is limited in throughput rate or batch size either by nuclear criticality considerations or for process reasons.

D. RECYCLED FUEL PROPERTIES THAT AFFECT NONDESTRUCTIVE ASSAY

Certain properties of the fissile and fertile materials in the HTGR recycled fuel exert a controlling influence on the selection and implementation of nondestructive assay techniques. Foremost among these is the high gamma radiation level associated with the ²³³U due to the unavoidable inclusion of trace amounts of ²³²U. This single factor vastly complicates the problem of performing all operations with the material including nondestructive assay. All operations must be performed remotely, and this requirement imposes the physical complexities associated with automated operation and remote maintenance and increases by an order of magnitude the problems associated with construction, handling, and use of calibration standards. Other characteristics which influence assay equipment selection include the fissile and fertile loadings of the fuel elements, the uranium isotopics, the spontaneous fission and (α, n) neutron sources, the neutron response characteristics, thermal emissions, and the physical composition of the fuel forms. Each of these is discussed in detail in this section.

D.1 Gamma Radiation Characteristics

The gamma-ray radiation associated with ²³³U fuel is due to the inclusion of trace amounts of ²³²U. The ²³²U isotope is produced in the reactor environment by neutron reactions with ²³²Th, ²³⁰Th, and ²³³U. Figure 3 shows the primary production modes for ²³²U and the ²³²U decay scheme. The range of ²³²U concentrations in the recycled uranium varies from 100 to 1200 ppm of uranium depending on the neutron flux, the neutron fluence, the neutron energy spectrum, and the ²³²Th, ²³⁰Th, and ²³³U concen-



Figure 3. Production and Decay Chains for ²³²U.

trations in the source fuel elements. The content of 230 Th is an important variable because the productions of 232 U from 230 Th requires only two successive neutron captures whereas 232 U production from 233 U or 232 Th requires an (n,2n) reaction. The (n,2n) reaction is a threshold reaction so that only neutrons above 6.37 MeV for 232 Th and above 6.00 MeV for 233 U can contribute to the production of 232 U from these two nuclides whereas the capture reactions have no thresholds. The 230 Th content of natural thorium is dependent on the thorium ore source, specifically the ratio of uranium-to-thorium in the ore bed (230 Th is the second daughter in the 238 U decay chain). Typical 230 Th concentrations are less than 100 ppm of total thorium.

The decay of ²³²U, shown in Figure 3, leads to the formation of several gamma-ray emitting daughters. The major nuclides for gamma-ray production are ²¹²Pb, ²¹²Bi, and ²⁰⁸Tl. Of these, the ²⁰⁸Tl creates most of the radiation problems because 100% of its decays are accompanied by the emission of a 2.61 MeV gamma ray. Due to the 1.9 year half-life of the ²²⁸Th nuclide, the intensity of the gamma radiation is time-dependent and increases rapidly during the first two years after recovery of the uranium from the spent fuel. At a time of 90 days after the ²³³U and ²³²U have been separated from the ²³²U daughter nuclides, the intensity of the gamma radiation from 100 g of ²³³U with 1200 ppm ²³²U is 700 mrad/hr at a distance of 50 cm. Since the gamma radiation of material containing



Figure 4. Time-Dependent Radioactivity of ²³²U Daughters for Initially Pure ²³²U.

²³²U increases with time, it becomes more difficult to handle from a radiation exposure standpoint as it ages. In light of this, uranium that is received at the refabrication facility will have had most of its daughter products recently removed from it. After the separation process, the daughters start to build back towards secular equilibrium with ²³²U. Assuming complete removal of all daughters, their radioactivity will increase as shown in Figure 4. The figure is based on the premise that at zero time 1.0 g of 232 U is the only material present. The ²³²U activity is off scale on this figure but has a constant value of 21.4 curies over the 90-day time scale. About ten years would be required for all the daughters to reach transient equilibrium with ²³²U due to the 1.9 year half-life of ²²⁸Th. At equilibrium, the ²⁰⁸Tl activity will be 36% of the ²³²U activity and the ²¹²Po activity will be 64% of the ²³²U activity because of the branching that occurs at ²¹²Bi (see Figure 3).

Not all separation techniques completely remove the ²³²U daughter products. For instance, an ion exchange system will remove the ²²⁸Th but leave fractions of the ²²⁴Ra, ²²⁰Rn, and ²¹²Pb. Solvent extraction removes only part of the ²²⁸Th but removes most of the other daughters. The effect on the ²⁰⁸Tl activity of these two separation methods is shown in Figure 5.³ For periods less than 16 days after separation, the ²⁰⁸Tl activity is dependent on the separation technique used and, in addition, depends on the specific process variables. Thus, for times less than 16 days after separation, the relationship between ²⁰⁸Tl ac-



Figure 5. The Effect of Postseparation Time and Type of Separation on the Radioactivity of ²⁰⁸Tl per Unit Initial Radioactivity of ²³²U.

tivity and ²³²U content depends on how the separation was made.

During HTGR fuel refabrication, the concentrations (or radioactivities) of the ²³²U daughter products are not only a function of the time after separation of the decay products and the method of separation but are also dependent on some process variables of the refabrication operation. One of these operations is the conversion of the carbonized resin beads to a $UC_2 + UO_2 + C$ kernel. As described previously, the conversion process consists of heating the carbonized resin beads to 1750°C in a fluidized-bed furnace and holding at that temperature for a period of 20-30 min. During this process, the concentrations of ²³²U daughters will be altered in the fuel particles.⁴ For instance, ²²⁰Rn, which is a gas, diffuses out of the particles. In such a case, the approach to transient equilibrium that was described above will be affected. Two example cases of this situation were studied and their effect on ²³²U daughter concentrations are presented in Figures 6 and 7.

Figure 6 shows the effect on the ²¹²Pb number density of continuously removing various quantities of ²²⁰Rn from an equilibrium concentration of daughter products. Figure 7 shows this same effect on the ²⁰⁸Tl activity. It is seen that as soon as the radon removal has ceased, the ²¹²Pb concentration begins to increase. This is because the ²²⁰Rn has a very short half-life compared to the time scale used in Figure 6 and returns to its equilibrium concentration very quickly. Along with this rapid return of ²²⁰Rn to equilibrium, the formation rate of ²¹²Pb increases which in turn increases the ²¹²Pb number density. A similar phenomena occurs with respect to the ²⁰⁸Tl concentration. However,







45 50



Figure 8. Gamma-Ray Spectrum of a ²³²U-²³²U Fuel Sample Measured with a Ge(Li) Detector.

there is a time lag between the end of the radon removal and the beginning of ²⁰⁸Tl increase because the 10.6 hr half-life of ²¹²Pb is long compared to the time scale presented.

Other processes which will affect the concentrations of the gamma emitting nuclides are resin loading, resin carbonization, and buffer coating of the fissile kernel. After the second coating layer has been applied, the daughter nuclides will be contained within the particles so that process operations which follow coating should not alter the daughter nuclide concentrations in the fuel.

A gamma-ray spectrum of ²³³U measured with a Ge(Li) detector is shown in Figure 8. The detector is true coaxial with a total volume of 54 cm^3 , a relative efficiency of 9%. and a resolution of 2.3 keV at 1332 keV. The uranium contained about 250 ppm ²³²U and was near secular equilibrium with its daughter nuclides. The labels of Figure 8 near the peaks indicate the parent nuclide and the gamma-ray energy. A compilation of gamma-ray energies and intensities for ²³²U, ²³³U, ²³²Th, and their daughter nuclides is available in a separate publication.⁵ All the major peaks in Figure 8 are from gamma-rays emitted during the decay of ²³²U daughters. There is no indication of any ²³³U gamma-ray lines. In particular, the relatively intense ²³³U line at 317 keV does not appear in this spectrum. This indicates that the above detector could not determine the ²³³U content of this sample by direct detection of a ²³³U gamma-ray line. An experiment is currently underway to measure this same material with a detector of smaller sensitive volume and with a Compton suppres-

Table 2
Recycle Fuel Block Loading Ranges for the Commercial HTGR
(Kilograms heavy metal per block)

Period	$^{233}\mathrm{U}$	Thorium	Th/233U Ratio
Transition			
Maximum	0.72	12.1	16.8
Minimum	0.31	6.0	19.4
Equilibrium			
Maximum	0.72	11.6	16.1
Minimum	0.32	7.9	24.7

Ta	hle	3
1 a	DIC.	J

Uranium Isotopic Composition
for Commercial HTGR Recycle Fuel

Description	Isotope	Percent of Total (Atom Density Basis)
Beginning of recycle	233U	92.1
0 0 ,	$^{234}\mathrm{U}$	7.35
(Most reactive fuel)	$^{235}\mathrm{U}$	0.568
``````	236U	0.0245
	$^{232}\mathrm{U}$	0.0126
Equilibrium recycle	233U	61.4
- ,	$^{234}\text{U}$	24.3
	235U	8.02
	$^{236}\mathrm{U}$	6.30
	232U	0.0362

sion spectrometer system to determine whether the ²³³U gamma-rays can be resolved.⁶

In addition to complicating or preventing the application of existing gamma-ray assay techniques, the presence of ²³²U and its daughters also complicates the assay problem because of personnel exposure considerations. Personnel shield requirements are dictated by the presence of the highly penetrating 2.6 MeV gamma-ray from the ²⁰⁸Tl decay.

#### **D.2 Fissile and Fertile Loadings and Uranium Isotopics**

The relative quantities of thorium and uranium that will be loaded into an element will vary considerably. Spatial zoning factors and recycle zoning factors will be changing from year to year during a transition period when the first quantities of recycle  233 U are introduced. Data on maximum and minimum loadings that are expected to be encountered have been supplied by General Atomic Company.⁷ Table 2 gives the range of  233 U and thorium loadings that will occur during equilibrium recycle and the transition period leading to equilibrium. The values given are for the 1160 MW(e) HTGR. Table 3 gives the calculated uranium isotopic ratios for the beginning of recycle and at equilibrium recycle for the large commercial reactors. The  232 U contents are calculated based on thorium feed with less than 5 ppm  230 Th.

From this information, it is evident that nondestructive assay techniques developed for the HTGR fuel cycle must be capable of accommodating a wide range of fissile loadings, fissile-to-thorium ratios, and uranium isotopics. Of particular note is that in the equilibrium recycle fuel, 12% of the fissile uranium is  235 U.

#### **D.3 Neutron Sources and Intensities**

The HTGR recycle fuel is a potential source of spontaneous fission neutrons and  $(\alpha,n)$  reaction neutrons that can affect the methods selected for nondestrucive assay. This section describes the neutron source intensities expected from each of these reactions.

Table 4	
Specific Activities and Neutron Emission Rates	
for Spontaneous Fission ^a	

Nuclide	Spontaneous Fission Half-Life (yr)	Specific Activity for Spontaneous Fission (d/s/g)	Neutron Emission $(\nu_{sp}=2)$ (n/sec/g)
²³⁰ Th	$>1.5 \times 10^{20}$	<3.8 ×10-7	$<7.6 \times 10^{-7}$
$^{232}\mathrm{Th}$	>1021	$<$ 5.7 $\times 10^{-8}$	$1.1 \times 10^{-7}$
$^{232}\mathrm{U}$	$8 \times 10^{13}$	$7.1 \times 10^{-1}$	1.4
$^{233}\mathrm{U}$	$1.25 \times 10^{17}$	$4.55 \times 10^{-4}$	$9.1 \times 10^{-4}$
$^{234}\text{U}$	$2 \times 10^{16}$	$2.8 \times 10^{-3}$	$5.6 \times 10^{-3}$
$^{235}\text{U}$	$1.9 \times 10^{17}$	$3.0 \times 10^{-4}$	$6.0 \times 10^{-4}$
236U	2 $\times 10^{16}$	$2.8 \times 10^{-3}$	$5.6 \times 10^{-3}$
$^{238}\mathrm{U}$	7.19×10 ¹⁵	$7.73 \times 10^{-3}$	$1.5  imes 10^{-2}$

^a W. SCHIRMER AND N. WÄCHTER, "Table of Specific Activities of the Nuclides with Z=88 to Z=104," Actinides Rev., 1: 125-34 (1968).

The specific spontaneous fission activities⁸ and neutron emission rates of nuclides of interest in the HTGR recycle fuel are listed in Table 4. The neutron emission rates are extremely low for all nuclides except  232 U; however, the  232 U concentration in recycle  233 U is expected to be in the range of 100 to 1200 ppm so that the spontaneous fission neutron yield of equilibrium recycle fuel (61.4 at. %  233 U, 24.3 at. %  234 U, 8 at. %  235 U, 6 at. %  236 U, and 0.12 at. %  232 U) is only 4 neutrons/sec/kg. This low neutron emission rate excludes the use of spontaneous fission neutrons as a fissionable material signature. In addition, the low neutron yield from spontaneous fission should not interfere with other NDA methods employing neutrons as the interrogating or signature radiation.

Neutron production by alpha particle reaction with light elements is a second source of neutrons from HTGR recycle fuels. The alpha activities of ²³³U, ²³²U, and the decay daughters of ²³²U provide a time-dependent alpha source. The principal light nuclides of interest are ¹³C and ¹⁸O.

The "thick target" neutron yields for the elements carbon and oxygen are 0.11 neutrons/ $10^6$  alphas and 0.07 neutrons/ $10^6$  alphas, respectively, for an alpha particle energy of 5.305 MeV.⁹ The relationship between the neutron yield and the alpha particle energy has been determined from the calculations of Van Tuyl.¹⁰ The energy-dependent thick target yields of carbon and oxygen can be approximated by the expressions

Neutron yield from carbon =  $4.89 \times 10^{-11} E_{\alpha}^{4.65} n/\alpha$  (1)

Neutron yield from oxygen =  $2.18 \times 10^{-11} E_{\alpha}^{4.84} n/\alpha$  (2)

where  $E_{\alpha}$  is the alpha particle energy in MeV.

For compounds containing oxygen, carbon, and heavy metals, the total neutron yield is determined by the expression of Matlock and Metz¹¹

$$\Upsilon = \frac{\sum m_i S_i \Upsilon_i}{\sum m_i S_i},\tag{3}$$

where  $\Upsilon$  is the total neutron yield,  $m_i$  is the mole fraction of the ith element,  $S_i$  is the stopping power of the ith element, and  $\Upsilon_i$  is the thick target yield. The stopping power S is proportional to  $Z(Z+7)^{-1/2}$ , where Z is the atomic number.¹¹

The calculational method based on Eq. (3) provides only an approximation of the neutron yield. For a more precise calculation of the  $(\alpha,n)$  yield on oxygen the methods of Taherzadeh¹² and Taherzadeh and Gingo¹³ should be utilized. To the author's knowledge there have been no reported measurements of the  $(\alpha,n)$  yield of ²³³U-²³²U carbide or oxide. Such measurements are needed to verify the calculated yields particularly in the case of carbide fuel.

The  $(\alpha, n)$  reaction yields calculated with Eqs. (1-3) are listed in Table 5 for three solid fuel compounds that occur in the fuel refabrication process. Table 5 presents the  $(\alpha, n)$ yields of each compound for 1 kg of beginning recycle uranium and 1 kg of equilibrium recycle uranium at two aging times. The uranium ages of 10 and 90 days represent practical minimum and maximum times for the refabrication of the HTGR fuels. The ²³²U content of the uranium is assumed to be 500 ppm in all cases.

The neutron yields of the HTGR recycle fuel are comparable to the  $(\alpha,n)$  yields of ²³⁹PuO₂.¹⁴ In addition, the neutron yield is time dependent and process dependent because, as described in Section D.1., daughter nuclides evolve from the fuel during some steps of the refabrication process. The  $(\alpha,n)$  yields from the fuel compounds can be used as a qualitative indication of fissile material content; however, the dependence of the neutron yield on age and process variables as well as the possibility of limited  $(\alpha,n)$  production from fuel impurities render any  $(\alpha,n)$  based methods impractical for accurate quantitative nondestructive assay. The  $(\alpha,n)$  yield may affect other assay methods which rely on neutron detection methods.

#### **D.4 Neutron Response Characteristics**

An important nuclear characteristic for evaluation of nondestructive assay equipment is the response of the recycled fuel to neutron interrogation. This section enumerates the fission cross sections and the yields of prompt and delayed neutrons from fission. The use of prompt and delayed gamma rays from neutron induced fission are also

		Table 5	999	<u> </u>	
	Calculated Yields of (a	a,n) Neutrons from HTGR F	Lecycle Fuel		
			Neutron Yield, n/sec/kg U		
		Fuel Compounds in Refabrication			
Uranium, Age (Days)	Uranium Type	Loaded Resin $(UC_{15}O_5H_{15})^a$	$\begin{array}{c} Carbonized \ Resin \\ (U_2C_9O_4)^a \end{array}$	Fuel Kernel $(U_2C_6O)^a$	
10	Beginning recycle ^b Equilibrium recycle ^c	$4.5 \times 10^4$ $4.2 \times 10^4$	$3.5 \times 10^4$ 3.3 × 10 ⁴	$2.7 \times 10^{4}$ 2.5 × 10 ⁴	
90	Beginning recycle ^b Equilibrium recycle ^c	7.6×10 ⁴ 7.5×10 ⁴	$5.8 \times 10^4$ $5.8 \times 10^4$	$4.5 \times 10^{4}$ $4.4 \times 10^{4}$	

^aChemical formula indicates element ratios only, does not imply chemical form.

^bAtom percent of beginning recycle uranium (²³²U, 0.05%; ²³³U, 92.1%; ²³⁴U, 7.35%; ²³⁵U, 0.57%; ²³⁶U, 0.025%).

^cAtom percent of equilibrium recycle uranium (²³²U, 0.05%; ²³³U, 61.4%; ²³⁴U, 24.3%; ²³⁵U, 8.02%; ²³⁶U, 6.30%).



Figure 9. Fission Cross Sections of ²³⁵U and ²³³U.



Figure 10. Fast Neutron Fission Cross Sections of the Fertile Nuclides ²³²Th, ²³⁴U, ²³⁶U, and ²³⁸U.

Thermal Cross Sections (Barns) and Resonance Integrals for the Uranium and Thorium Nuclides^a

	$^{232}\mathrm{U}$	²³³ U	234U	²³⁵ U	236U	238U	²³² Th
σ,	73.1	47.7	100.2	98.6	5.2	2.70	7.40
$\sigma_{\rm f}$	75.2	531.1	< 0.65	582.2			$4 \times 10^{-5}$
$\sigma_{\rm a}$	148.3	578.8	100	680.8		2.70	7.4
ō,	14.7	8.2	12	13.8		8.90	12.67
σt	163.0	587.0	112	694.6		11.60	20.07
If	320	764		275			
Īγ	280	140	630	144	365	275	85
,							

^aS.F. MUGHABGHAB AND D.I. GARBER, Neutron Cross Section, Vol. 1: Resonance Parameters, BNL-325, 1(3): (1973).

Table	7
I adde	1

Prompt Fission Neutron Yields for Thermal Fission^{a,b}

Fission Nuclide	ν _p	-
232U	3.15	-
$^{233}\mathrm{U}$	2.485	
235U	2.402	

^aS.F. MUGHABGHAB AND D.I. GARBER, Neutron Cross Sections, Vol. I: Resonance Parameters, BNL-325, 1(3): (1973).

^bG.R. KEEPIN, The Physics of Nuclear Kinetics, Addison-Wesley, 1965.

Table	8
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Delayed Neutron Yields for Thermal and Fast Fission

	Absolute Delayed Neutron Yield per Fission					
	Neutron Energy Inducing Fission					
Fission Nuclide	Thermal ^a	3.1 MeV ^b	14.9 MeV ^b			
232U		$0.060 \pm 0.006$	$0.031 \pm 0.003$			
$^{233}\mathrm{U}$	$0.0066 \pm 0.0003$	$0.0077 \pm 0.0008$	$0.0043 \pm 0.0004$			
$^{234}\mathrm{U}$		$\mathbf{N}\mathbf{A}^{\mathbf{c}}$	NA			
$^{235}\mathrm{U}$	$0.0158 \pm 0.0005$	$0.018 \pm 0.002$	$0.0095 \pm 0.0008$			
$^{236}\mathrm{U}$		NA	NA .			
$^{238}\mathrm{U}$		$0.049 \pm 0.005$	$0.0286 \pm 0.0025$			

^aG.R. KEEPIN, *The Physics of Nuclear Kinetics*, Addison-Wesley, 1965.

^bC.F. MASTERS, M.M. THORPE, AND D.B. SMITH, "The Measurement of Absolute Delayed-Neutron Yields from 3.1- and 14.9-MeV Fission," *Nucl. Sci. Eng.*, **36**: 202-8 (1969).

° NA, not available.

discussed as a potential NDA signature for HTGR recycled fuel. Response characteristics of neutron radiative capture have not been evaluated.

The fission cross sections of the fissile nuclides ²³³U and ²³⁵U are shown in Figure 9.¹⁵ The thermal neutron cross sections and resonance integrals of the fissile and fertile nuclides of interest are listed in Table 6. The fast neutron fission cross sections of the fertile nuclides are shown in Fig. 10.¹⁶ Two important features which affect the nondestructive assay of this fuel are the low magnitude of the ²³²Th fission cross section and the relatively high cross section and low threshold for fission of ²³⁴U which will comprise 25% of the equilibrium recycle uranium. The prompt fission neutron yields from thermal neutron fission are listed in Table 7.¹⁷ The ²³³U and ²³⁵U neutron yields differ by 3.4% and the  $\nu\sigma_f$  values of these isotopes differ by 5.8% for thermal fission. The absolute delayed neutron yields are enumerated in Table 8 for fission induced by thermal,¹⁷ 3.1 MeV, and 14.9 MeV¹⁸ neutrons. It should be noted that the ratio of the delayed neutron yield of ²³⁵U to ²³³U is 2.4. This difference might be exploited as the basis of an isotope discrimination technique.¹⁹ The large difference also means that the net signal from an NDA technique based on thermal neutron irradiation and delayed neutron counting will be a factor of ~2.6 less for ²³³U than for ²³⁵U.

The prompt and delayed gamma yields from neutron induced fission have not been investigated in detail because it can be demonstrated that the gamma signal rates from the prompt and delayed gammas are substantially lower than the background rate from the ²³²U daughter nuclides in the recycle fuel. Forster, et al., have reported results of a Pin and Pellet Assay System (PAPAS) on PWR-type low enriched uranium fuel pins.²⁰ This system uses thermal neutron interrogation and delayed gamma-ray detection. Using their data on signal and background count rate, we have estimated that the ratio of net signal to background would be 0.15 for the assay of HTGR recycle fuel rods that contain ²³³U with 100 ppm ²³²U and 20-day age. This very low net signal to background ratio could be improved by increasing the ²⁵²Cf source strength of the irradiator; however, the design basis of the plant requires that the assay system handle uranium with 1200 ppm ²³²U at a 90-day age. The background activity of this fuel is approximately 50 times higher than the radiation from 100 ppm ²³²U, 20day uranium. Based on the very low net signal to background ratio for this fuel, it is concluded that neutron induced gamma ray signals cannot be utilized in NDA techniques for HTGR recycled fuel. This conclusion has a strong impact on the selection of NDA methods for recycle HTGR fuel because assay methods using neutron induced gamma-ray signals have been one of the most widely used and accepted active NDA techniques.

#### **D.5 Thermal Emission Characteristics**

Sponaneous alpha decay of the fissile ²³³U and its companion ²³²U and the accompanying release of thermal energy are of interest as a potential source of information for nondestructive assay by calorimetric techniques. While the spontaneous decay heat from ²³³U is small relative to the plutonium isotopes ( $\sim$ 270  $\mu$ W/g of ²³³U) it is sufficient to allow assay of concentrated samples by passive calorimetry. The presence of the ²³²U contaminant introduces problems, however, both by virtue of its own spontaneous decay and by the additional time dependent heat

Table 9     Energy Release of ²³³ U, ²³² U, and ²³² U Daughters from Radioactive Decay						
Component of Decay Heat	Reaction	Reaction Q _a Value (MeV) ^a	Recoil Plus Particle Energy per Decay	Gamma Energy per Decay		
²³³ U decay	$^{233}U \xrightarrow{\alpha} ^{229}Th$	4.909	4.901	0.0081		
²³³ U daughters	$\stackrel{229}{\longrightarrow} \stackrel{\alpha}{\longrightarrow} \stackrel{225}{Ra}$	Negligible – see text				
²³² U decay	$^{232}U \xrightarrow{\alpha} ^{228}Th$	5.414	5.395	0.019		
			Recoil plus particle energy per ²²⁸ Th decay at chain equilibrium	Gamma energy per ²²⁸ Th decay at chain equilibrium		
²³² U daughters	228 Th $\xrightarrow{\alpha}$ 224 Ra	5.521	5.496	0.0251		
	224 Ra $\xrightarrow{\alpha}$ 220 Rn	5.787	5.774	0.0134		
	220 Rn $\xrightarrow{\alpha}$ 216 Po	6.405	6.405	0.0004		
	²¹⁶ Po $\xrightarrow{\alpha}$ ²¹² Pb	6.906	6.906	0.00002		
	²¹² Pb $\xrightarrow{\beta}$ ²¹² Bi	0.580	0.366	0.214		
	$\frac{\beta}{64\%}^{212}$ Po	2.246	1.330	0.107		
	$^{212}\text{Bi} \xrightarrow{36\%}_{\alpha} ^{208}\text{Tl}$	6.206	2.220	0.0143		
	²¹² Po $\xrightarrow{\alpha}$ ²⁰⁸ Pb	8.954	5.731	0.0		
²³² U daughter ch	²⁰⁸ Tl $\xrightarrow{\beta}$ ²⁰⁸ Pb ain – total at chain equilibrium	4.994	0.568 34.796	1.229 1.603		

^aG.M. LEDERER, J.M. HOLLANDER, AND I. PERLMAN, Table of Isotopes, 6th ed., John Wiley and Sons, Inc., New York, 1967.

production of its daughter products. On a gram for gram basis ²³²U produces approximately 2500 times as much heat as does ²³³U.²¹ Hence, in ²³³U containing 400 ppm clean ²³²U, half of the decay heat is attributable to the contaminant. At the end of 10 years, the same material will be producing an additional amount of heat equal to 5.8 times that attributable to ²³³U due to the build in of ²³²U daughter products.

The controlling time constant in the ²³²U daughter chain is the decay constant of its first member, ²²⁸Th. Approximately 30 days after uranium cleanup, the remaining daughters have reached dynamic equilibrium with the ²²⁸Th and from that point on (barring physical removal of any of its members) the entire chain follows the growth curve of the 1.9 year half-life of ²²⁸Th. Over the first 90 days, the period of interest for HTGR fuel refabrication, the increase is almost linear and, at the end of that period, the daughter activities have reached approximately onetenth of their final equilibrium values.

The fissile  233 U also has an extensive family of alpha active daughters. However, the 7340 year half-life of  229 Th, the first member of that chain, acts as an effective throttling valve on the growth of the following members. At 100 days after uranium cleanup, the  233 U daughters contribute less than 0.02% of the heat generated by the parent nuclide.

The decay chain for ²³²U is shown in Figure 3. Table 9 below gives the Q values for each reaction and the average energy in each decay which is attributable to gamma ray emission and recoil plus particle emission. The rationale for this breakdown is that while all of the recoil and particle energy from radioactive decay will remain within a calorimeter, the gamma energy may be only partially absorbed. The fraction absorbed will depend on the sample and calorimeter size and composition. It is evident from Table 9 that the heat generation components of ²³³U and ²³²U are only slightly sensitive to this effect. The energy released by the ²³²U daughters, however, is 4.4% gamma. In any calorimetric measurement where ²³²U daughter activity is a significant contributor to total heat output, account would have to be taken of the partial absorption of the emitted gamma energy.

Physical and Cher	T nical Prope (Avera	able 10 rties of the Pa age Values)	rticulate Fuel	Forms
	Dry Loaded Resin Beads	Carbonized Resin Beads	Converted Kernel	Coated Particle
Diameter, μm	550	385	370	670
Density, g/cm ³	1.7	3.3	3.2	2.2
Elemental				
Composition, wt %	76			
Uranium	47	71	82	20
Silicon				19
Oxygen	15	19	3	1
Carbon	35	10	15.	60
Hydrogen	3			



Figure 11. Heat Generation of 1 g of ²³³U Containing 400, 800, and 1200 ppm ²³²U as a Function of Time After Uranium Cleanup.

Figure 11 shows the magnitude of the heat generation components in 1 g of ²³³U containing 400, 800, and 1200 ppm ²³²U as a function of time after uranium cleanup. From the figure, it is evident that measurement of the ²³³U content in a sample requires accurate knowledge of both the ²³²U contamination, the material's age, and some assurance that the daughters of ²³²U are in secular equilibrium with ²²⁸Th. In addition, it can be seen that increasing ²³²U content places correspondingly increasing accuracy requirements on the overall heat generation measurement in order to maintain a fixed accuracy in the ²³³U content determination.

#### **D.6 Physical Characteristics**

The chemical composition and physical form of the special nuclear material changes drastically while passing through the refabrication plant from uranyl nitrate solution on entrance to finished HTGR fuel blocks on exit. At intermediate stages in the process, the product material can be found in the form of loaded resin beads, carbonized beads, converted kernels, coated particles, and uncarbonized fuel rods. This section described those physical characteristics of the materials which influence the selection of specific assay techniques. The nominal size, density, and chemical composition of the dry loaded resin beads, the carbonized beads, the converted kernels, and the coated particles are listed in Table 10. The physical structure of the coated particles is presented in Table 1. The values in Table 10 are nominal only. The standard deviation in the

#### Table 11

Elemental Composition of Gree
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	Elemental Composition (wt %)					
Element	Begin 233U 1	ning of Recycle	Equil 233U ]	ibrium Recycle		
	Minimum U + Th	Maximum U + Th	Minimum U + Th	Maximum U+Th		
Hydrogen	0.95	0.80	0.90	0.80		
Carbon	79.20	65.16	74.11	64.58		
Oxygen	2.28	3.81	2.81	3.70		
Silicon	0.86	1.67	1.24	2.51		
Thorium	15.81	26.83	19.65	25.81		
Uranium	0.90	1.73	1.29	2.60		

distribution of kernel diameters is typically 15  $\mu$ m for the converted kernel. Variations of 5% in uranium content are observed between different batches of loaded resin. The density of the converted kernel is sensitive to the set of process conditions selected. Studies of the expected variations in chemical compositions of the kernels and coated particles are not yet available for the reference processing conditions.

The elemental composition of the green fuel rod is dependent on the uranium and thorium loadings, the uranium isotopic composition, the particle sizes, the packing fraction, and the matrix composition. Table 11 lists the elemental composition of uncarbonized fuel rods for two uranium and thorium loadings and for two isotopic compositions of uranium. The uranium and thorium loadings correspond to the maximum and minimum loadings contemplated for commercial HTGR recycled fuel (Table 2). A particle packing fraction of 0.62 was assumed for all cases. There are significant variations in the uranium content of the fuel rods. For NDA, this means that a fuel rod assay technique must be capable of operating over a wide range of rod loadings with particular emphasis on those rod loadings that would lead to poor signal to background characteristics.

The hydrogen content of the fuel rods could show more variation than indicated in Table 11 because the particle packing fraction exhibits some variation due to the different sizes of fertile, fissile, and graphite particles that are mixed in the fuel rod. The hydrogen is contained in the pitch that is used to bind the particles together. As the packing fraction changes, the amount of pitch within a rod also varies. The extent of this variation is not presently quantified; however, the hydrogen variability is an important factor in the selection of an NDA technique for uncarbonized fuel rods.

The fuel rods are formed in a molding operation and consequently the dimensions of the rods are extremely precise. Diameters are uniform to  $\pm 0.001$  cm and lengths to  $\pm 0.02$  cm. Because the uranium particles occupy only 5 to 21% of the volume of a rod, the uniformity of the particle distribution is an important factor in selecting NDA methods for fuel rods. Variations of 5 to 10% in the uranium loading per cubic centimeter of fuel rod are expected based on current particle blending performance.

Even with fuel rods that are uniform on a macroscopic scale, the rods are still quite heterogeneous. The fissile material is concentrated in the particle kernels rather than being dispersed uniformly throughout the rod. This microscopic heterogeneity can pose significant problems for some NDA methods because an individual particle can exhibit significant neutron self-shielding at low neutron energies.

#### E. NONDESTRUCTIVE ASSAY REQUIREMENTS

In maintaining the required material balance across the refabrication facility, there are four primary areas where NDA instrumentation can provide the only realistic determination of material content, provide a more rapid and less expensive analysis, or provide the assurance that arises from high sampling rates. These four applications are waste material assay, fuel rod assay, fuel element assay, and small sample assay.

A nondestructive assay technique is required for waste because of the inability to sample the heterogeneous waste material. In this regard, the waste that requires assay is solid material such as plastic sheeting, manipulator boots, plastic sample holders, absorbent paper, disposable graphite furnace parts, small metal parts, etc. This material will contain a few grams of fissile material per cubic foot and in the pilot plant will be segregated into combustible and noncombustible materials and packaged in 30-gal drums for retrievable surface storage. A nondestructive assay technique with an accuracy of 10 to 15% is required for these drums. The fissile content of solid waste from a commercial facility may be somewhat lower as more effort would be expanded to internally recycle the uranium.

The second nondestructive assay technique required for HTGR fuel recycle is a fuel rod assay machine capable of measuring either 100% of the fuel rod output or a sampling of the fuel rods. Because a commercial recycle facility would produce approximately 100,000 rods per day, a 1% sample would require 1000 rods per day to be analyzed. Only a nondestructive technique can be expected to handle this sample rate. If 100% measurement is required and it may be necessary if the rod forming step is the last stage in the process where it is practical to measure the fissile content of the product fuel, then nondestructive assay is a necessity.

A fuel element assay system is required to directly measure the product output of the refabrication facility. Because the uranium content of the fuel element is a small fraction of the element mass ( $\sim 1\%$ ) and because the element mass changes during fuel rod carbonization, weight measurements of the loaded element cannot be used for SNM accounting. The fuel element assay instrument would measure the product stream of the refabrication facility; consequently, the instrument would have to exhibit very low systematic errors or biases. Providing adequate fuel element standards for this instrument would be a difficult and expensive operation. It is the authors' opinion that a fuel element NDA instrument cannot be developed to the performance level required within the near future. This opinion is based primarily on the difficulties attendant with NDA methods that attempt to precisely assay extended heterogeneous distributions of fissile material. Accountability information on loaded fuel elements will most likely have to be based on fuel rod assay techniques.

A system for assaying samples of loaded resin beads, fuel kernels, and coated particles is needed for two reasons. First, it would provide a rapid turn-around on fissile content analysis of loaded resin so that process decisions as to the acceptability of a batch can be made without long delay times. Second, it provides a means of measuring small samples of uncoated and coated particles to verify fissile contents of homogeneous particle batches which are amenable to sampling. This sampling would be performed as part of a material inventory and would also enable verification of fissile material flow during processing. The small sample assay system, if sufficiently accurate and rapid, would eliminate a significant number of chemical analyses that would otherwise be required. The chemical analyses and, particularly, the sample preparations required for coated particles are difficult to perform with the radioactive ²³³U fuel. The precision and accuracy requirements of the small sample assay instrument are very stringent. To meet current LEMUF requirements precisions of 0.3 to 0.5% (2 $\sigma$ ) for single measurements are required. Accuracies attainable will depend on the precise characterization of standards and the control of systematic errors. The usefulness of this nondestructive technique decreases rapidly as the achievable precision and accuracy limits increase above the 1% level.

A fifth NDA capability might be useful and more efficient for the measurement of containers of coated particles in various process stages. The containers would hold 500 to 3000 g of ²³³U particles and would serve as storage vessels between process steps. The assay machine would need to measure the total fissile content of these containers. Such an assay instrument would eliminate sampling and sample analysis operations on this type of material. It might also be more reliable for off-specification material where sampling uncertainties are high. In the pilot plant, off-specification material and scrap of high fissile content will be canned and shipped to the Idaho Chemical Processing Plant (ICPP) for recycle. The container tentatively selected for this scrap material is a double-walled aluminum can 3.5 inches in diameter and 9 inches in length. A method of measuring the fissile content of these cans is needed for the pilot plant operation.

These nondestructive assay requirements have been identified in the analysis of the accountability system required for a commercial recycle facility. The accountability information can be measured and collected by destructive means for fuel rods and small samples; however, the gains to system performance achievable by the use of accurate and reliable NDA instruments make the development and demonstration of such instruments an important part of the program to develop an economical and licensable refabrication technology. The following section examines the applicability of currently implemented or developed methods to the NDA requirements of the refabrication facility.

#### F. EVALUATION OF NDA METHODS FOR APPLICATION TO HTGR RECYCLED ²³³U FUEL

Nondestructive fissile material assay methods are typically classified as either passive or active, the difference being that a passive method measures spontaneous or inherent emissions from the fissile nuclides, whereas an active method uses an interrogating radiation source to induce signature radiation from the fissile material.

The major passive and active methods can be categorized as follows:

#### Passive Techniques

Gamma-Ray Measurements Neutron Measurements Spontaneous Fission Neutrons (a,n) Neutrons Calorimetry

#### Active Techniques

Interrogating Radiation	Reaction	Signature Radiation
Neutrons Thermal Epithermal Subthreshold Superthreshold	Fission	Gamma-rays Prompt Delayed Neutrons Prompt Delayed
Bremsstrahlung (4-10 MeV end point)	Photofission	Gamma-rays Delayed Neutrons Prompt Delayed

In most cases, there have been several instruments developed which utilize one of the methods for a particular NDA application. There is no attempt made here to compile a list of NDA instruments as this has been done elsewhere.²²

Each of the NDA methods is discussed in light of the specific NDA requirements and the nuclear and physical characteristics of the recycled ²³³U fuel.

#### F.1 Passive Assay Techniques

Assay techniques using gamma-ray radiation have been developed for direct enrichment measurements of uranium, assay of uranium and plutonium waste materials, and measurements of uranium and plutonium material holdup in fuel processing facilities.¹⁴ To draw upon this considerable development and operating experience, the gammaray radiation of the ²³³U-²³²U should, if possible, be exploited for nondestructive assay.

Gamma-ray assay techniques might be utilized in two ways: (1) to assay for  233 U content by direct measurement of the  233 U gamma-rays, and (2) to assay for  233 U indirectly by measuring the gamma-ray radiation from one or more of the  232 U daughter nuclides. The latter method

requires that the ²³²U to ²³³U isotopic ratio be known. Two difficulties are associated with the first method of gamma-ray assay. Although the intensity of the 317 keV gamma-ray of ²³³U is greater than the intensity of the 186 keV gamma-ray of ²³⁵U or the 414 keV gamma-ray of ²³⁹Pu for equal fissile masses of each nuclide, the gammaray radiation of the ²³²U daughters in HTGR recycle fuel masks the ²³³U gamma rays in common gamma detectors or spectrometers. (See Section D.1) If small volume detectors or detectors with Compton background suppression can resolve the ²³³U gamma-ray without severe count rate or efficiency problems, then this method would have application to the assay of the low fissile content wastes. The second difficulty with ²³³U assay using gamma ray measurements is that the 317 keV gamma-ray might be significantly attenuated by the matrix material in the waste drum. If the ²³³U gamma ray can be resolved, a method of using the low and high energy gamma rays of ²⁰⁸Tl to make an attenuation correction has been proposed.²³

Assay of the waste material by measurement of the gamma-ray radiation from the ²³²U daughters currently exists at ORNL for waste assay of ²³³U with less than 10 ppm ²³²U.³ The method uses NaI detectors to resolve the 2.61 MeV gamma-ray of ²⁰⁸Tl and compares the activity of a 55 gallon waste drum with the activity of a dummy container that has representative matrix material and a known quantity of ²³³U. In order for the techniques to be accurate, the standard is made from the same batch of uranium as the material in the waste can. Corrections must be made for attenuation effects and sufficient time must pass to allow any process dependent transients in the ²⁰⁸Tl activity to die out. Current performance of this technique indicates accuracies of  $\sim 7\%$  (2 $\sigma$ ). This accuracy would be adequate for the quantities of material expected in the waste drums. Several problems remain to be resolved before such a method is applicable to HTGR recycled ²³³U fuel. The first problem is that the present application is used in a situation where there are only small variations in ²³²U content from batch to batch. Secondly, the refabrication process does not lend itself to complete cleanout after each batch as does the current application. And third, the requirement that ²⁰⁸Tl transients must die out could limit the timeliness of the information from this waste assay technique. Because of the relatively low accuracy requirements for waste assay, it may be possible to resolve the above problems and utilize the indirect gamma-ray assay for the waste material. The indirect method would not be suitable for NDA applications that require high accuracy.

Passive neutron detection systems for the coincident measurement of spontaneous fission neutrons are not possible because of the very low spontaneous fission rates of the nuclides in HTGR recycled fuel. Passive neutron detection systems for  $(\alpha,n)$  neutrons could be utilized with 100 g or greater quantities of this fuel; but accurate correlation of the neutron emission to the fissile content would not be possible because of the time-dependence of the alpha activity and because of batch to batch variations in oxygen and carbon contents of the fuel. The application of calorimetry as an assay method for ²³³U fuel was evaluated in Section D.5. and warrants further study for HTGR recycled fuels because of the excellent precision attainable in the heat measurement. The problem of isotopic content is much more important in the case of these fuels than for plutonium fuels. For accurate ²³³U assay, the ²³²U content would have to be known very accurately by other means, and at the present time no nondestructive techniques exist for measuring the ²³²U content of bulk material.

In summary, the indirect gamma-ray measurement method is the only passive technique that has previously been applied to ²³³U fuel; however, this method can be utilized only if the uranium isotopics are known and the age of the uranium is known. Measurement of the direct ²³³U gamma-ray radiation depends on being able to resolve the ²³³U gamma-rays. This has not yet been demonstrated for HTGR recycled ²³³U.

Neutron detection methods are not applicable to HTGR recycled fuel. Calorimetry may prove to be an accurate technique if methods of correcting for the heating component due to  232 U and its daughters can be developed. None of the passive techniques can be applied in the same manner as they are currently used with  235 U or Pu.

In relation to the specific NDA requirements set forth in Section E, only the waste assay and the scrap assay requirements could possibly be satisfied by passive methods. The lack of readily applied passive techniques will also adversely affect the ability to measure the SNM holdup in process equipment. The gamma activity of the ²³²U daughters can serve as a sensitive indicator of possible uranium holdup, but quantitative measurements would be possible only in very limited circumstances.

#### F.2 Active Assay Techniques

The active methods can be divided into photofission methods and neutron fission methods. Photofission assay techniques incorporate a pulsed electron linear accelerator (LINAC) as a source of bremsstrahlung (4-10 MeV end point) to induce fission in the fissile and fertile nuclides within the sample. If the matrix materials have low atomic numbers, then the penetrability of the bremsstrahlung is superior to fast neutron penetrability. Isotopic discrimination is possible for ²³⁵U-²³²Th fuels if the endpoint energy of the bremsstrahlung is adjustable and if the LINAC stability is good. The photofission method might, therefore, be a prime candidate for a whole element assay system or for the assay of bulk fissile material; however, basic data need to be measured before a detailed analysis is possible. The basic data required are the yields of delayed and prompt neutrons from the photofission of ²³³U, ²³⁴U, and ²³⁶U. Also, a technique must be demonstrated to allow discrimination between the fissile nuclides  $(^{233}\text{U} + ^{235}\text{U})$ and the fertile nuclides  $(^{234}\text{U} + ^{236}\text{U} + ^{232}\text{Th})$ .

The remainder of this section discusses applications of neutron interrogation methods for nondestructive assay. Because of the high gamma-ray background, it has been determined that measurement of the delayed gamma-rays from neutron-induced fission is not possible for recycled

²³³U fuel. No attempt has been made to investigate the applicability of prompt fission gamma-rays for the same reason. Assay instruments utilizing neutron interrogation and prompt or delayed neutron detection seem to have no inherent limitation with respect to the gamma-ray radiation of the recycled ²³³U fuel. Neutron detectors can be selected which either have low gamma-ray sensitivity or can be shielded from gamma-ray radiation without substantial neutron attenuation. The  $(\alpha, n)$  neutrons may represent a background problem for those systems that are designed to assay bulk quantities of ²³³U. This background does affect the required source strength of the interrogating neutrons. The primary problems associated with neutron interrogation techniques are the ability to discriminate between fissile and fertile nuclides, the penetrability of the interrogating neutrons, the uniformity of response from fissile material at different positions within a sample, and the neutron source strength required to achieve the necessary counting precision or signal level. Discrimination against the fertile nuclides ²³²Th, ²³⁴U, ²³⁶U, and ²³⁸U can be obtained by requiring that the interrogating neutrons have energies below the fission thresholds of these nuclides.

Because neutron penetrability and uniformity of response are important parameters in the active techniques, the geometry and composition of the material to be analyzed are critical to the selection of appropriate NDA methods. The fuel rod and small sample assay requirements are, therefore, discussed separately from the whole element and bulk material assay requirements.

In the review of active NDA methods for the fuel rod and small sample assay systems, first priority was given to methods being developed or in use for ²³³U fuels or HTGR ²³⁵U fuel. These methods are delayed neutron activation analysis,²⁴ the fission multiplicity method of the Isotopic Source Assay System (ISAS),25 the isotopic source "Random Driver,"26 the Sb-Be source assay system,27 the twoenergy gamma-ray transmission method,²⁸ and ²⁵²Cf source, delayed neutron assay systems.^{29,30} Of these methods only the delayed neutron and Sb-Be assay systems could be applied to the determination of ²³³U and ²³⁵U in uncarbonized fuel rods and particle samples. ISAS and the Random Driver will not function in the high gamma field associated with recycle fuel. The two-energy gamma-ray transmission method determines only the total heavy metal content. The delayed neutron method described in Ref. 24 used a TRIGA reactor as the interrogating source, and, consequently, is a method for assaying samples only. Because of the high cost of a research reactor, reactor-based assay methods have not been considered for routine sample assay requirements.

The ²⁵²Cf source, delayed neutron assay system of Ref. 29 was developed for the assay of ²³³U in LWBR fuel pellets. The 5-mg ²⁵²Cf source in this instrument was moderated by water and the fuel pellet samples were moved automatically from the irradiation position to a delayed neutron counter. A precision of 0.3% (2 $\sigma$ ) in a single 5-min measurement was reported for a single 2.2 wt % ²³³UO₂-ThO₂ pellett containing 0.62 g of ²³³U.

The Sb-Be assay system consisted of a beryllium, titanium, and nickel irradiation chamber surrounded by 8 cm of Pb shielding. Fast neutron detectors were positioned in a nickel reflector around the lead. The Sb-Be source produced 25-keV neutrons, and the prompt fission neutrons produced in the sample are detected by the fast neutron detectors.

Recently, the Los Alamos Scientific Laboratory (LASL) has investigated the use of thermal neutron irradiation with prompt neutron detection as a method for the assay of HTGR fuel rods. The method is based on the operation of the ²⁵²Cf source, LWR fuel rod scanners.³¹ The investigations at LASL have concentrated on ²³⁵U fuels for HTGR's.

The characteristics of the fuel rods and particle types that affect the selection of NDA equipment were described in a previous section. The particular characteristics that impact the active neutron interrogation methods are the hydrogen content and hydrogen variability in the uncarbonized resin beads and fuel rods, the relatively low fissile content per unit volume in HTGR rods, the particulate nature of the fuel, and the presence of multiple fissile nuclides in the fissile particles.

The hydrogen content of the uncarbonized resin beads and fuel rods makes fast neutron irradiation techniques difficult for these fuel forms because the hydrogen moderates the interrogating neutrons and the fast neutron irradiation becomes a partially thermal irradiation. This property makes unfeasible the fast neutron irradiation of a bulk sample of fuel rods or a bulk quantity of loaded resin beads.

The fissile content per unit volume of an HTGR fuel rod is only one-third to one-fifth that of an LWR fuel rod. This means that for assay systems that require close coupling of the neutron source to the fuel rod or particle sample, such as is done in the Sb-Be system, the count rate will be substantially lower for HTGR rods than for LWR rods unless the irradiation source is increased in size and in overall intensity.

The particulate nature of the fuel results in a double self-shielding phenomena within a fuel rod or sample of particles. Individual fuel kernels exhibit some neutron self-shielding (intraparticle) and particle-to-particle selfshielding (interparticle) also occurs in the rods or particle samples. This self-shielding phenomena is important only in the thermal and resonance energy regions where the neutron cross sections of the fissile and fertile materials are high. Interparticle self-shielding can be treated in a manner similar to self-shielding in LWR fuel rod assay systems if the fissile particles are uniformly distributed over the rod or sample. The intraparticle self-shielding is thought to be the more serious problem for thermal neutron irradiation because the extent of the self-shielding is a function of the kernel density and diameter. These two variables may affect the assay results independently of the fissile content. The effects of these variables on assay accuracy for the resin-derived fuel kernel are not precisely known at this time but a program at LASL to define the particle effects is under way. The approximate magnitude of the particle



Figure 12. Neutron Self-Shielding Factors at 0.025 eV for Weak-Acid Resin Derived ²³³U Kernels as a Function of Fissile Kernel Diameter for Three Kernel Densities.

self-shielding effect is in the range of 4% to 15%.^{32,33} The sensitivity of the self-shielding factor to kernel diameter and density is illustrated in Figure 12. The kernel self-shielding factor of Figure 12 is the ratio of the average 0.025 eV flux within the fuel kernel to the average 0.025 eV flux within an infinite medium of fuel rod materials. The technique of Wälti was used to calculate the kernel self-shielding factors.³² Figure 12 indicates that expected variations in diameter ( $\pm 25 \ \mu$ m) and density ( $\pm 0.2 \ \text{g/cm}^3$ ) would have a 0.5 to 1.0% effect on the self-shielding factor and the analysis of SNM.

If a particle sample or fuel rod contains two fissile nuclides, the ability to assay for total fissile content requires (1) that the isotopics be obtained by some independent means, or (2) that theNDA methods be able to distinguish between the two fissile nuclides, ²³³U and ²³⁵U, or (3) that the NDA method respond identically to both ²³³U and ²³⁵U. The uranium isotopics of each batch of fuel rods will be known because the process control requires this information. Nonetheless, a method that either distinguishes between the two fissile nuclides or treats them identically would have safeguards advantages in that the total fissile content could be determined independent of the isotopic analysis.

None of the active assay methods appear to have a decided advantage for fuel rods or particle samples containing hydrogenous material. The Sb-Be system is most applicable to the particle sample; however, the intensity required for adequate precision suggests that 100-Ci Sb sources would be necessary. This is a factor of 10 increase in the source level over the present LASL device.²⁷ The problems associated with bimonthly or quarterly handling of the Sb source must be factored into the analysis of this technique. The thermal neutron irradiations with prompt neutron detection yield higher count rates per source neutron than other methods; however, the intraparticle selfshielding may significantly limit the achievable accuracy of this method.

The nondestructive assay of fuel elements or bulk quantities of fuel particles (nonhydrogenous) requires the use of penetrating radiation. Bremsstrahlung irradiation and delayed neutron detection was mentioned earlier as a possible method for these two applications. Development work at LASL has been reported on the whole block assay of fresh (235U) HTGR fuel elements using neutron irradiation and delayed neutron counting.34,35,36 The method utilized a 14-MeV neutron generator to induce fissions in the fissile and fertile material in the fuel element. By spectrum tailoring the neutron source in a second measurement, the²³⁵U and thorium could be differentiated. The results of the initial tests indicated that the method was promising; however, accuracies of 5 to 7% (1 $\sigma$ ) were quoted for the ²³⁵U content. The accuracy required for safeguards purposes is a factor of 5 to 10 better than the current performance, and the operation of this technique with mixed fissile nuclides and a significant  $(\alpha, n)$  neutron background has to be demonstrated. This basic method should also be applicable to bulk quantities of fissile particles. The lack of thorium in these particle batches would help improve the attainable accuracy. The particle container size could be selected to optimize the assay performance consistent with practical production constraints.

LASL has developed a second method that is also, with modification, applicable to bulk particle samples. This technique referred to as the Californium Shuffler is a subthreshold or thermal neutron irradiation combined with delayed neutron detectors.³⁷ The ²⁵²Cf source is "shuffled" between the irradiation position and a storage position while the fissile material is constrained in a combination irradiator and neutron counter. The modification necessary to apply this instrument to recycled fuel consists of adding sufficient gamma-ray shielding to the sample chamber so that 0.1 to 1 kg quantities of ²³³U can be assayed. The intensity of the neutron source selected for this application would depend primarily on the ( $\alpha$ ,n) neutron background from the sample. Source sizes in the range 0.5 to 2 mg of ²⁵²Cf would be required.

In summary, several active methods that have been developed for ²³⁵U fuels may with further development be applicable to HTGR recycled ²³³U fuels. No NDA instruments are presently developed, however, that would satisfy the NDA requirements of an HTGR recycle facilty. Methods using fast neutron irradiation must account for the hydrogen variability in the fuel rods. Thermal neutron irradiation techniques, which generally require smaller source intensities, are adversely affected by the double self-shielding effect. Solutions to these problems are not yet in hand. The fuel rod, fuel element, and bulk particle assay instruments would by necessity have to be remotely operable and semiremotely or remotely maintainable.

#### **G.** Conclusions and Recommendations

This paper has identified the major properties of recycled ²³³U fuel for HTGR's that affect the selection of NDA methods. The results indicate strong requirements for active analysis techniques to overcome the problems with passive assay techniques that are caused by gammaray radiation from the ²³²U daughter nuclides. Waste materials with low fissile content can possibly be measured with an indirect, passive gamma-ray detection system. Assay instruments for fuel rods, bulk quantities of fuel particles, fuel elements, and particle samples must employ active interrogation methods. Neutron interrogation methods for fuel rods and particle samples must contend with the effects of hydrogen variability and intraparticle selfshielding. Photofission methods cannot yet be evaluated for these applications because of a lack of data for ²³³U, ²³⁴U, and ²³⁶U responses.

Selection of the most appropriate techniques for nondestructive assay of ²³³U fuels would be facilitated if the following information or techniques were available:

1. Photofission response data for ²³³U, ²³⁴U, and ²³⁶U is required in order to evaluate this method for NDA. The excellent penetrability of high energy bremsstrahlung in carbonaceous materials combined with the current availability of commercial, albeit expensive, LINAC's designed for industrial radiography make the photofission techniques of possible interest for this fuel form.

2. Measurements of the  $(\alpha,n)$  neutron production rates for ²³³U-²³²U carbide and oxide fuel forms encountered in the processing of HTGR ²³³U fuels are necessary to determine the background count rates and the required source intensities for active assay instruments.

3. Research should be initiated on methods to determine by nondestructive means the isotopic information necessary to employ calorimetry for accountability measurements. The application of calorimetry to ²³³U fuels deserves increased development attention because of the precision attainable in calorimetric measurements.

The program to develop commercial technology for the recycle of ²³³U fuels in HTGR reactors is in the conceptual design stage. Detailed design of a pilot plant facility is scheduled to begin in mid 1975. The integration of nondestructive assay equipment into the pilot plant demonstration requires that techniques be demonstrated in the laboratory within the next two to three years. Because much of the NDA equipment must be operated and maintained remotely or semiremotely, it is important that the NDA instruments be integrated into the facility. The pilot plant demonstration provides an opportunity to test and refine NDA instrumentation in a realistic operating environment so that *demonstrated* NDA technology is available for the design of a commercial recycle facility.

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# Making Inferences About the Shipper's Variance In a Shipper-Receiver Difference Situation

#### **By John L. Jaech** Exxon Nuclear Company, Inc.

#### Introduction

There are a number of instances in safeguards in which the situation identified in the title as the "shipper-receiver difference situation" will arise. This is the situation in which two parties make independent measurements on the same items. This occurs for example whenever

- The shipper and the receiver make measurements on the same items (hence the label).
- An inspector, or more generally, an audit team makes measurements on samples of items and compares the results with those values assigned by the facility operator.
- An operator compares measurement methods by measuring some items by both methods and compares results.
- Two analytical laboratories are compared by sending the same items to both laboratories for measurement.

In this article, some results are given that will enable the user to determine what statistical procedure to use in certain estimation and hypothesis-testing situations.

#### The Model

The mathematical model that applies is written in its simplest form. Since biases that may exist between the two sets of measurements do not affect inferences made about the variances, they are not included in this discussion. For simplicity in exposition, the model is written in the shipperreceiver notation framework, keeping in mind the more general applications.

Let

- s; = shipper's reported value for item i
- ri = receiver's reported value for item i
- $\mu_i$  = true value for item i
- $\epsilon_j$  = random error of measurement for the shipper for item i
- $\mathsf{n}_j$  = random error of measurement for the receiver for item i

Then, the model is

$$s_j = u_j + \varepsilon_j$$
 (1)

$$r_{i} = \mu_{i} + \eta_{i}$$
 (2)

where  $\epsilon_i$  and  $n_i$  are normally distributed with zero means and variances denoted by  $\sigma_c^2$  and  $\sigma_n^2$  respectively. The index i runs from 1 to n, the sample size.

#### The Estimation Problem

The basic assumption throughout this discussion is that the receiver has some knowledge of  $\sigma_n^2$  and wishes to make

inferences about  $\sigma_\epsilon^2$ . In the estimation problem, the aim is to obtain an estimate of  $\sigma_\epsilon^2$ . There are two possibilities that the receiver wishes to consider, leading to different estimators.

$$\hat{\sigma}_{\varepsilon}^2 = \mathbf{s}_{\mathbf{d}}^2 - \sigma_{\mathbf{n}}^2 \tag{3}$$

Estimator 2. The receiver does not wish to use his knowledge about  $\sigma_n^2$  in obtaining the estimate of  $\sigma_e^2$ . The Grubbs estimation procedure will be used [1]. This involves computing the variances of the  $s_i$  and  $r_i$  values, denoted by  $s_s^2$  and  $s_r^2$  respectively, and the covariance between them, denoted by  $s_{sr}$ . Then, it is known that  $s_s^2$  estimates  $\sigma_\mu^2 + \sigma_e^2$  while  $s_{sr}$ estimates  $\sigma_\mu^2$ . Therefore, the estimate of  $\sigma_e$  is

$$\tilde{\sigma}_{c}^{2} = s_{s}^{2} - s_{sr} \tag{4}$$

where the tilde is used in place of the caret to distinguish this from the first estimator.

#### Variance of Estimator 1.

Assuming that  $\sigma_n^2$  is known without error, then the variance of  $\sigma_\epsilon^2$  is simply the variance of  $s_d^2$ . It is well known that under the assumption of normality that applies in this discussion,

riance 
$$s_d^2 = \frac{2 \sigma_d^4}{(n-1)}$$
 (5)

where  $\sigma_d^2$  is the true variance, i.e.,  $\sigma_d^2 = \sigma_{\varepsilon}^2 + \sigma_{\eta}^2$ . Therefore, (5) becomes

variance 
$$\hat{\sigma}_{\varepsilon}^2 = \frac{2 \left(\sigma_{\varepsilon}^2 + \sigma_{\eta}^2\right)^2}{(n-1)}$$
 (6)

Variance of Estimator 2.

From Grubbs [1], the variance of 
$$\sigma_{\epsilon}^2$$
 is given by

variance 
$$\tilde{\sigma}_{\varepsilon}^2 = \frac{2}{(n-1)} \frac{\sigma_{\varepsilon}^4}{(n-1)} + \frac{(\sigma_{\mu}^2 \sigma_{\varepsilon}^2 + \sigma_{\mu}^2 \sigma_{\eta}^2 + \sigma_{\varepsilon}^2 \sigma_{\eta}^2)}{(n-1)}$$
 (7)

which is a function of  $\sigma_{\mu}^2$  in addition to  $\sigma_{\epsilon}^2$  and  $\sigma_{n}^2$ .

Comparison of Variances

In comparing variance  $\hat{\sigma}_{\epsilon_2}^2$  in (6) with variance  $\hat{\sigma}_{\epsilon}^2$  in (7), express  $\sigma_{\epsilon}^2$  as a function of  $\sigma_{\eta}$  by

$$\sigma_{\varepsilon}^{2} = R \sigma_{\eta}^{2}$$
 (8)

Then (6) becomes

variance  $\hat{\sigma}_{\varepsilon}^2 = \frac{2 \sigma_{\eta}^4 (R+1)^2}{(n-1)}$  (9)

and (7) becomes

variance 
$$\hat{\sigma}_{c}^{2} = \frac{\sigma_{\eta}^{4} R(2R+1) + \sigma_{\mu}^{2} \sigma_{\eta}^{2} (R+1)}{(n-1)}$$
 (10)

The ratio 0 of the two yarjances is formed. This becomes a function of R and of T =  $\sigma_{\mu}/\sigma_{\eta}$ 

$$\theta = \frac{2 (R+1)^2}{R^2 + (R+T)(R+1)}$$
(11)

Find those values of R and T such that  $\theta_{\epsilon}$  to identify the region in which  $\hat{\sigma}_{\epsilon}^2$  is more precise than  $\hat{\sigma}_{\epsilon}^2$ . To do this, solve the inequality

$$\frac{2(R+1)^2}{R^2 + (R+T)(R+1)}$$
 <1

The solution is

$$\frac{3R+2}{R+1}$$
 

Whenever R and T are such that inequality (12) holds,  $\hat{\sigma}_{\epsilon}^2$  is more precise (has smaller variance) than  $\vartheta_{\epsilon}^2$ . The following table gives the solution to (12).

#### TABLE I

Region in Which Estimator I  $(\hat{\sigma}_{\epsilon}^2)$  is Preferred

<u>R</u>	<u>T&gt;</u>
0	2
0.5	2.33
1	2.5
2	2.67
4	2.8
	3

It is clear that the critical parameter is T, the ratio of the product variance to the receiver's measurement error variance. If this ratio exceeds 3,  $\sigma_{\epsilon}^2$  in Equation (3) is always preferred, regardless of the relationship between  $\sigma_{\epsilon}^2$  and  $\sigma_n^2$ . If the ratio, T, is less than 2,  $\sigma_{\epsilon}^2$  in Equation (4) is preferred, again regardless of the relationship between  $\sigma_{\epsilon}^2$  and  $\sigma_n^2$ . If  $\sigma_{\epsilon}^2$  and  $\sigma_n^2$  are of the same order of magnitude, i.e., if R=1, then  $\sigma_{\epsilon}^2$  is preferred when T>2.5, and  $\sigma_{\epsilon}^2$  is preferred.

#### The Hypothesis-Testing Problem

In the hypothesis-testing problem, the shipper has assigned a value to  $\sigma_c^2$ . The receiver wishes to determine if the shipper-receiver data confirm that the shipper's stated value is correct. Two tests of the hypothesis that  $\sigma_c^2 = \sigma_{co}^2$  are considered. These are related to the two estimators previously discussed.

Test 1

Accept the value of  $\sigma_{\eta}^2$  as being known. Form the "d" statistic,  $d_i$  =  $s_i$  -  $r_i$  and calculate the variance of the di values,  $sd^2$ . The hypothesis is  $\sigma d^2 = \sigma_{\varepsilon o}^2 + \sigma_{\eta}^2$  where  $\sigma_{\varepsilon o}^2$  is the value for  $\sigma_{\varepsilon}^2$  stated by the shipper, and  $\sigma_{\eta}$  is presumed known by the receiver. Under this hypothesis, the statistic

$$x_{n-1}^{2} = \frac{(n-1) sd^{2}}{\sigma_{e,0}^{2} + \sigma_{n}^{2}}$$
(13)

is distributed as chi-square with (n-1) degrees of freedom. Rejection of the hypothesis is assymed to be equivalent to a rejection of the hypothesis that  $\sigma_{\epsilon}^{2} = \sigma_{\epsilon_{0}}^{2}$  since the receiver accepts the value of  $\sigma_{\eta}^{2}$  as being known.

Test 2

The receiver does not use his knowledge about  $\sigma_n^2$  in testing the hypothesis that  $\sigma_c = \sigma_{c0}$ . The large-sample test due to Jaech [2] is used. This tests the joint hypothesis:  $\sigma_c^2 = \sigma_{c0}^2$  and  $\sigma_n^2 = \sigma_{n0}^2$ , the alternative being that one or both equalities is invalid. This requires computation of ss², sr², and ssr as with the use of the  $\sigma_c^2$  estimator. To perform the test, calculate

$$\hat{\sigma}_{\mu}^{2} = \frac{s_{s}^{2} \sigma_{\eta 0}^{4} + 2s_{sr} \sigma_{\varepsilon 0}^{2} \sigma_{\eta 0}^{2} + s_{r}^{2} \sigma_{\varepsilon 0}^{4}}{(\sigma_{\varepsilon 0}^{2} + \sigma_{\eta 0}^{2})^{2}} - \frac{\sigma_{\varepsilon 0}^{2} \sigma_{\eta 0}^{2}}{\sigma_{\varepsilon 0}^{2} + \sigma_{\eta 0}^{2}} (14)$$

$$L_1 = -n - 0.5 n \ln (s_S^2 s_r^2 - s_S r^2)$$
(15)

$$L_{2} = -0.5 \text{ n } \ln \left(\hat{\sigma}_{\mu}^{2} \sigma_{\varepsilon o}^{2} + \hat{\sigma}_{\mu}^{2} \sigma_{\eta o}^{2} + \sigma_{\varepsilon o}^{2} \sigma_{\eta o}^{2}\right)$$
$$-n \left[\frac{\left(\hat{\sigma}_{\mu}^{2} + \sigma_{\eta o}^{2}\right) s_{S}^{2} - 2\hat{\sigma}_{\mu}^{2} s_{S}r + \left(\hat{\sigma}_{\mu}^{2} + \sigma_{\varepsilon o}^{2}\right) s_{r}^{2}}{2 \left(\hat{\sigma}_{\mu}^{2} \sigma_{\varepsilon o}^{2} + \hat{\sigma}_{\mu}^{2} \sigma_{\eta o}^{2} + \sigma_{\varepsilon o}^{2} \sigma_{\eta o}^{2}\right)}\right] (16)$$
$$\lambda = 2 (L_{1} - L_{2})$$
(17)

For large samples ("large" not specified further for the moment),  $\lambda$  is distributed as chi-square with 2 degrees of freedom. Thus, for example, at the 5% level of significance, the hypothesis  $\sigma_{\rm E}^{2}$  =  $\sigma_{\rm Eo}^{2}$  and  $\sigma_{\rm n}^{2}$  =  $\sigma_{\rm ho}^{2}$  is rejected if  $\lambda > 5.99$ .

#### Comparison of Tests

The two tests are compared on the basis of their power, i.e., their ability to detect specified departures from the hypothesis. In finding the test powers, a Monte Carlo computer simulation is used. Input parameters include

> $\sigma_{\mu}^{2}$ , the true product variance n, the sample size  $W \approx \sigma_{\epsilon}^{2}/\sigma_{\epsilon_{0}}^{2}$ , where  $\sigma_{\epsilon_{0}}^{2}$  is the hypothesized value

The parameter  $\sigma_n^2$  is held fixed, and is set equal to  $\sigma_{co}^2$ . In the simulation, the data are generated for each trial, where it is assumed that ui is normally distributed with given mean and variance  $\sigma_\mu$ .  $\chi_{(n-1)}^2$  is then computed from (13) and  $\lambda$  from (17). Those instances in which  $\chi_{n-1}^2$  is either less than or greater than the appropriate 5% critical values for (n-1) degrees of freedom are noted, as are those instances in which  $\lambda$  exceeds its critical value of 5.99. Note that the chi-square test is set up as two-sided test of the hypothesis that  $\sigma_c^2 = \sigma_{co}^2$ ; Test 2 is two-sided by its very nature. By counting the number of trials in which critical values are exceeded, and dividing by the total number of trials, the test powers are computed. 5000 trials were run for each case.

The results of the simulation are given in Tables II and III. In Table II, n,  $\sigma_{\mu}^2$ , and W are varied while in Table III, n is held fixed at n=20 and  $\sigma_{\mu}$  is extended beyond the range of Table II. Since the power of Test 1 is independent of  $\sigma_{\mu}^2$ , the results for Test 1 are appropriately combined in the tables.

	ſ	<u>TABLE</u> Test Powers Ver	$\frac{11}{\sigma_{11}}$ sus n, $\sigma_{11}^{2}$ , and W	
			r.	
<u>n</u>	<u>w_</u>	$\sigma_{\mu}^2/\sigma_{\eta}^2$	<u>Test l</u>	<u>Test 2</u>
10 10 10 10	1 1 1 2	0 0.25 1 0	.0542 .0462 .0510 .1318	.0992 .0830 .0892 .2736
10 10 10	2 2 4	0.25 1 0	.1750 .1812 .1868 . .5834 .	.2528 .2298 .7254
10 10	4 4	0.25 1	.5704 } .5745 .5698 }	.6838 .6270
20 20 20 20	1 1 1 2	0 0.25 1	.0476 .0482 .0508 2864	.0672 .0676 .0698 4264
20 20 20	2 2 4	0.25 1 0	.2994 .2899 .2838 .8222	. 3946 . 3158 . 9296
20 20	4 4	0.25 1	.8364 } .8310	.9182
40 40 40 40	1 1 2	0 0.25 1 0	.0466 .0502 .0522 .4820	.0556 .0598 .0616 .6986
40 40 40	2 2 4	0.25 1 0 0.25	.4756 .4783 .4774 .9784 .9784 .777	.6194 .5320 .9982
40 40	4 4	1	.9764	.9898

#### TABLE III

Test Power Versus  $\sigma_u^2$  and W

at	n	=	20	
<b>u</b> 5			20	

<u>w</u>	$\sigma_{\mu}^2/\sigma_{\eta}^2$	<u>Test 1</u>		<u>Test 2</u>
2 2 2 2 2 2 2	0 0.25 1 4 16 64	. 2864 . 2994 . 2838 . 3008 . 2882 . 2836	.2904	.4264 .3946 .3158 .2768 .2430 .2312
4 4 4 4 4 4	0 0.25 1 4 16 64	.8222 .8364 .8344 .8354 .8368 .8262	.8316	.9296 .9182 .8784 .8186 .7908 .7684

Table II shows that for Test 2, the actual significance level is larger than the intended value of 0.05. This is a consequence of the fact that the test is a large sample test, and for the smaller values of n, the test tends to reject the hypothesis more often than it should. This discrepancy is quite large for n = 10, but by the time n = 40, the discrepancy is considered tolerable. Even for n = 20, Test 2 can probably be applied with some caution, perhaps accounting for the larger actual level of significance by increasing the critical value somewhat beyond 5.99.

Table III examines the power of Test 2 as a function of  $\sigma_{\mu}^2$ , the product variance, relative to  $\sigma_n^2$ , the measurement error variance. The sample size is fixed at n = 20, the minimum value for which Test 2 should be applied. Table II shows that when  $\sigma_{\mu}^2$  is less than or equal to around twice  $\sigma_n^2$ , then Test 2 has a higher power than Test 1, and is the preferred test. For large values of  $\sigma_{\mu}^2$ , Test 1 has the higher test power.

#### Conclusions and Recommendations

Two situations have been considered when making inferences about the shipper's random error of measurement. On the one hand, it is assumed that the receiver uses his own value for the measurement variance in making inferences about the shipper's variance. On the other hand, he makes inferences about the shipper's variance independent of his knowledge about his own variance. The corresponding estimation and test procedures are referred to as Estimators 1 and 2 and Tests 1 and 2, respectively.

From an estimation viewpoint, Estimator 1 is preferred whenever the ratio of the product variance  $(\sigma_{2}^{2})$  to the receiver's measurement variance  $(\sigma_{n}^{2})$  exceeds 3 while Estimator 2 is preferred if this ratio is less than 2. For values of the ratio between 2 and 3, the estimator preference depends on the ratio of  $\sigma_{E}^{2}$  to  $\sigma_{n}^{2}$ , where  $\sigma_{E}^{2}$  is the measurement variance for the shipper. Table I provides guidance in the choice of which estimator to use.

From a hypothesis-testing viewpoint, Test 2 should not be used when the sample size is less than 20, although in the range from 10-20, it can be used with some caution, possibly by increasing the size of the critical value slightly. For values of n >20 and when  $\sigma_{co}^2 = \sigma_n^2$ . Test 2 is prefered when  $\sigma_\mu^2/\sigma_n^2$  is less than about 2.5. This result is consistent with the result for estimation (see Table I), and it is deduced that at values of R  $\neq$  1, the Table I criteria will also apply from a hypothesis testing viewpoint as long as n exceeds 20.

#### Acknowledgement

 ${\rm I}$  am indebted to Anton Kraft who produced the Monte Carlo simulation results.

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- [2] Jaech, John L., "Further Tests of Significance for Grubb's Estimators". Biometrics, <u>27</u> (1971).

# Radioactive Waste Management for Nuclear Power Reactors Focus of UCLA Extension Program

A new short-course program titled, "Radioactive Waste Management for Nuclear Power Reactors," will be offered by UCLA Extension's Department of Engineering this fall, Monday to Friday, October 20 to 24, from 8:15 a.m. to 5 p.m. in Room 6266 of UCLA's Boelter Hall.

Designed for engineers working in utility and architect engineering companies and manufacturers of reactors and reactor components, the program will also be useful to staff members of state and federal regulatory agencies who want to learn practical engineering aspects of radioactive waste management.

Subject areas covered in the UCLA Extension

program will include source terms of radioactive waste for BWR, PWR, and HTGR, licensing and regulatory guides, safety and accident analysis, handling of gaseous, liquid and solid waste, transportation and burial, health physics and plant operating experience.

A banquet speech by Willard F. Libby, Nobel Prize Winner and professor of chemistry at UCLA, will highlight the Wednesday, October 22 meeting, according to course coordinator A.A. Moghissi.

For additional information write Department of Engineering, UCLA Extension, P.O. Box 24902, Los Angeles, CA 90024 or call (213) 825-1047.

# DEVELOPMENT OF A COMPLETE SYSTEM DETERMINING ROUTINE INSPECTION EFFORTS AND TIMING FOR FABRICATION PLANTS

By Tohru Haginoya, Manager

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Editor's Note: This article appeared in the Winter 1975 (Volume III, No. 4) issue of NMM Journal. However, this article by Tohru Haginoya et al. was incorrectly titled, listing as the author, Dr. Roger H. Moore, formerly of Los Alamos Scientific Laboratory--Tom Gerdis.

#### 1NTRODUCTION

At the Fourteenth Annual Meeting of the INMM we presented a paper "An approach to determining a system of routine inspection. efforts and timing for fabrication plants" developed in 1971. This System consisting mainly of the analysis of time-series data of MUF originating in each fuel fabrication facility was able to determine reasonable routine inspection efforts for each individual facility. But, as stated in the paragraph 81 of INFCIRC/153 (or The Agreement), actual routine inspection efforts should be determined taking, not only the analysis of MUF and so on, but also various other factors in fuel cycle into consideration. Therefore, in 1972 we evaluated those criteria listed in the said paragraph 81, that is, (a) the form of nuclear material (b) the effectiveness of the State's accounting and control system (c) characteristics of the State's nuclear fuel cycle (d) international interdependence (e) technical developments in the field of safeguards, in view of their influence on routine inspection efforts, and including the System developed in 1971 developed a complete System which can determine actual routine inspection efforts for any facility effectively. The outline of this complete System is shown below.

#### BASIC IDEA

First, "the fuel fabrication facility" (or Facility) studied in this paper shall mean such facility that processes uranium hexafluoride (UF₆) with enrichment less than 5 percent and produces  $UO_2$  powder or fuel assemblies. This corresponds to the facility defined in the paragraph 80 (c) in the Agreement and "the inspection efforts" cited here shall mean annual routine inspection efforts.

Secondly, in regard to the way of thinking about inspection we followed such idea mentioned in PART I of the Agreement as "BASIC UNDERTAKING" and considered "all source or special fissionable material in all peaceful nuclear activities" as the object of safeguards.

#### SYSTEM DEVELOPMENT TO CALCULATE INSPECTION EFFORTS

Structure of the System

Overall structure of the System including correlation with the System developed in 1971 is shown below.

 $F = f_1$  (law factor) x  $f_2$  (plant factor)

= a x MRIE x  $f_2$  (plant factor) ------ (1)

where

F : Annual routine inspection efforts (man-day)

 $a(\leq 1)$ : This factor, being the main object of this

study, is to be derived in consideration of the criteria listed in the paragraph 81 (a)  $\sim$  (e) of the Agreement

- MRIE (Maximum Routine Inspection Effort); Maximum routine inspection effort for the Facility, defined in the paragraph 80 of the Agreement
- f₂ : A function to be derived in consideration of the accountability of each individual Facility and developed by us in 1971

Basic Idea of Quantifying the Criteria

From the standpoint of inspection the distance between any nuclear facility and a nuclear weapon comes into question. In other words, the problem is how long it will take for nuclear material in certain chemical form in any nuclear facility to reach the nuclear weapon. In this study, however, we considered certain quantity of both metallic Pu and metallic  235 U to be equivalent with the nuclear weapon and defined them as Risk Material (RM) and also defined fast critical mass of both metallic Pu and metallic  235 U, namely 8 kg and 25 kg respectively, as Significant Quantity (SQ).

That is:

 $\epsilon$ (8 kg Pu metal) =  $\epsilon$ (25 kg ²³⁵U metal) = 1 SQ --- (2)

where  $\varepsilon$  = SQ function

Now, observing the movement of nuclear material which leads to RM paying special attention to the change of its chemical form it is understood that any nuclear material in certain chemical form will reach RM along possible routes being changed in its chemical form as it passes nuclear facilities. Thus, expressing chemical forms of various nuclear materials and moving directions of nuclear material towards RM with NODEs and ARCs respectively, we can make a network (RM Cycle) consisting of NODEs and ARCs. Of course, each route on the RM Cycle does not always coincide with that on the ordinary fuel cycle (Peaceful Use Cycle).

It is observed that any nuclear facility will give nuclear material in it such working operation which will cause one of the following changes, namely, (a) chemical form ("c" factor) (b) enrichment ("e" factor) (c) compo-sition ("m" factor) (d) burnup ("b" factor). Therefore, it follows that if we can express minimum time (Critical Time : Tc) in which just SQ of any nuclear material starting from any NODE on the RM Cycle reaches the RM NODE being given any one of the said four changes by each nuclear facility and consequently being changed in its chemical form, using such factors contained in the criteria (a)  $\sim$  (e) in the paragraph 81 of the Agreement, then we will be able to obtain a measure equivalent to the abovementioned distance between nuclear material and RM. In the following section we will show our mathematical model which can calculate "a" in Eq. (1) using Risk Degree (RD) concept that shows the relative status of nuclear material or a facility in fuel cycle.

#### Mathematical Model

In this study we introduced a concept of Risk Degree (RD) concerning nuclear material on any (material) NODE in order to formulate "a" and then to calculate reasonable F in Eq. (1). RD at NODE "i" is defined in Eq. (3)

$$RDi = \frac{\frac{1}{Q_i^{RM} \cdot Tc_i}}{\frac{\Sigma}{k \in Ri} \frac{1}{Q_k^{RM} \cdot Tc_k}}$$
(3)

where

 $q_1^{RM}$  equals  $\epsilon$  (Quantity of RM contained in nuclear material at NODE "i") on the condition that  $q_1^{RM}$  becomes SQ at RM NODE. As mentioned before Tc₁ means minimum time in which just SQ of nuclear material intentionally diverted from the NODE "i" reaches RM NODE.

The denominator of Eq. (3) is the sum of all  $1/q^{RM}$ . Tcs from NODE "i" to RM NODE along all the possible routes on RM Cycle.

Thus, RD of any facility can be obtained by replacing the numerator of Eq. (3) with the sum of  $1/Q^{RM}$ . Tc corresponding with all the NODEs which belong to the facility.

Namely,

$$RD = \frac{\sum_{i \in F} \frac{1}{Q_i^{RM} \cdot Tc_i}}{\sum_{k \in R} \frac{1}{Q_k^{RM} \cdot Tc_k}}$$
(4)

Then, let us consider the realtion between "a" and RD. Considering the effect of the NODEs located on the routes from NODE "i" to RM NODE, it is understood that increasing number of the NODEs will also increase the potential possibility that nuclear material will divert from any NODE towards RM NODE along any of the routes. So, we adopted following Eq. (5) taking this effect of the NODE number into consideration.

$$a = \sqrt[n]{RD} \qquad (5)$$

"n" shall mean the number of the unduplicated NODEs which are located along the routes that lead to RM NODE starting from such NODE(s) belonging to any facility. F is calculated by replacing "a" in Eq. (1) with Eq. (5).

Now, Tc₁ in Eq. (3) and Eq. (4) can be obtained by quantifying four factors, "c", "e", "m" and "b", and the basic idea of this quantification will be shown below.

. Quantification of "c" factor

This quantification is to be done by calculating the time in which certain amount of nuclear material corresponding to loss rate "1" to be reported as Design Information is first accumulated every year in a facility until total sum will reach SQ and then processed in the facility containing that nuclear material

#### Quantification of "e" factor

This quantification is done by calculating the time in which uranium with loss rate "1" and enrichment "E" is first accumulated and then enriched to produce SQ of highly enriched uranium ( $\geq 90$  %).

#### Quantification of "m" factor

This is done by calculating both the accumulating time and the processing time of such amount of U-Pu blend equivalent to SQ in the facility which performs blending of U and Pu.

. Quantification of "b" factor

"b" factor is considered about various nuclear reactors. In this case quantification is done by calculating the time in which SQ will be accumulated assuming that nuclear material can be diverted within calculation error  $\alpha_{\rm R}$  to be applied to the discharged fuel from reactors.

#### SIMULATION-CALCULATION OF INSPECTION EFFORTS

Taking, as examples, Facilities with capacities ranging from 100 to 500 TU per year and assuming various cases in the situation of fuel cycle in Japan, we applied our System to the Facilities and calculated inspection efforts for them. We had following results which are naturally to be affected more or less by the situation of those factors contained in the System developed in 1971 as accountability, confidence level and so on of the Facilities.

According to our results annual inspection efforts for any Facility will be a few man-days, that is, one inspection per year in the present fuel cycle of Japan, in which neither an enriching plant nor a reprocessing plant is present. In case of the fuel cycle after five years from now (1972), in which one reprocessing plant will be in operation and quite a few light water reactors will appear, annual inspection efforts for a Facility was calculated to be 15 - 30 man-days. Finally, in the complete fuel cycle, namely, all the nuclear facilities which are needed to provide complete fuel cycle including enriching plants and reprocessing plants will be present in Japan, we obtained annual inspection efforts which range from 20 to 35 mandays.

### New N.T.I.S. Publication

Nuclear Reactors Built, Being Built, or Planned in the United States as of Dec. 31, 1974. This compilation contains current information about facilities built, being built, or planned in the United States for domestic use or export which are capable of sustaining a nuclear chain reaction. Civilian, production, and military reactors are listed, as are reactors for export and critical assembly facilities. Revisions are published twice a year, and the information presented is current as of June 30 or December 31.

The publication (44 pages, 8 x 10 1/2, paperback) is available as TID-8200-R31 for \$4.00 from

National Technical Information Service U.S. Department of Commerce Springfield, Virginia 22161

# Some Thoughts on Constant and Variable Components of Systematic Error

#### By S.C. Suda Brookhaven National Laboratory

Systematic error has been struggling for recognition for almost fifty years. One of the first papers on the topic is due to Student [1] who in 1927 proposed treating some measurement error as semiconstant error. His suggestion appears to have been poorly received and we have no knowledge of Student's later thinking along these lines. We do know Student was ahead of his time and that he wrote during a period when Vigneron [2] and others were still questioning the applicability of the theory of probability and statistics to laboratory measurements. It was thought by some that applying probability theory to laboratory results would be wrong and unjustified.

The treatment of systematic error for the next forty years was the private domain of physicists and those involved in research and development. One approach taken by this group is best expressed by Beers [3] who states that causes of systematic error can generally be removed or controlled it the experiment is designed with care and that the remaining possible causes of systematic error can be listed and an intelligent guess made as to the magnitude of these errors.1) Under these controlled conditions Yardley Beers [3] page 35, suggests that "Since they are usually independent, systematic errors may be combined with random error by the methods of combining independent errors previously described ...".

Other characteristics of the classical approach are: a) systematic error and bias are synonymous; b) systematic error is always constant (i.e. it is not distributed); c) information on the magnitude and direction of these errors is in general unobtainable but if in certain cases this information is available the systematic error is a bias and can be algebraically corrected for; d) schemes that are developed for treating certain types of systematic error are invalid because they cannot be generalized.

More recently, the evaluation of errors associated with calibration of instruments and measurement systems has received much attention and these have expanded our notions of systematic error. For example, Eisenhart [4], page 170, states that "the systematic error of a measurement process will ordinarily have both constant and variable components". The consequence of this concept is that the estimate of the bias can be defined as the "constant component" of systematic error. The "variable component" of systematic error can be used to describe randomly distributed calibration errors which are perpetuated in all determinations for which the measurement process is used. This is the meaning I associate with Jaech's [5] "systematic error variance".

Crow [6] discusses the treatment of errors in a hierarchy of calibrations where each standard or instrument is subject to its own error. The accumulated error in each echelon in the hierarchy includes the error of all higher echelons. Since the calibration error in the standard at each echelon is measured by a standard deviation, the induced systematic error in the lower echelons will have the distribution of those errors.

Mandel [7] , page 281, in consider-ing measurement systems involving calibration lines states, "It is the nature of calibration lines that they are used repeatedly, either in relation to a single problem involving several determinations or for a variety of different problems. Any errors in the calibration line itself are thus perpetuated in all determinations for which it is used". My interpretation is that the estimate of the random error in the calibration line based on repeated calibrations is an estimate of the variable component of systematic error associated with all determinations for which it is used. The variable component of systematic error in the determinations can be reduced by repeating the calibration several more times but this error component cannot be decreased by repeated use of the calibration line.

This brings us to today's authors. I think the writings of John Jaech [5] [8][9] and Roger Moore [10] have brought us to the brink of a new and expanded definition of systematic error and I would like to add my observations. I am convinced that the classical treatment of systematic error is too limited and that it does not reflect today's needs or technology. I have spent many hours searching for a nice generalized theory of systematic error analogous to that on random error. I am convinced it doesn't exist. Now, if a generalized theory doesn't exist, does that imply that systematic error, as an entity separate from bias, does not exist? The Vignerons among us would say yes. I disagree. I think systematic error exists as bias and as the estimates of the variance of bias, standards, reference values, calibration equations and recovery factors.

I suggest the following are characteristics of systematic error:

- There does not exist a single, general model for estimating systematic error. There are several models, each specific to a measurement process and calibration technique.
- 2. Systematic error of a measurement process will ordinarily have both constant and variable components.
  - a) Bias is an example of the constant component²)
  - b) The estimate of the variance of bias and the random error associated with a calibration equation are examples of the variable component
- In a hierarchy of calibrations and measurements, the random error in the measured values in one stage may have the effect of the variable component of systematic error for a lower stage.
- Estimates of the systematic error components are obtained during calibration and in a measurement assurance program and do not depend on the particular measurement. Repeated measurements of a given item will not reduce the systematic error.
- 5. The probability distribution of the variable component of systematic error based on repeated calibrations with the reference value  $\overline{X}$  can be assumed to be normally distributed about  $\overline{X}$ . In some cases such as those involving severe rounding error an assumption can be made that the variable component follows a rectangular distribution. In other cases an interval in which the error lies may be given on the basis of the range of practical experience. These intervals may not necessarily be symmetrical about the mean.

Methods for the empirical estimation of the variable component of systematic error are specific to a measurement process and calibration technique. In this context, I observe at least four types of measurements for which separate computational procedures can be formulated. The four types of measurements are:³⁾

- Direct measurements. Length, time, weighing systems and some chemical determinations are examples of direct measurements.
- 2. Secondary measurements. These measurements involve calibration curves or recovery factors and include in-tank measurements of volume and many chemical determinations.
- Counting measurements. The measurements of nuclear materials are based on the observation of spontaneous or stimulated nuclear radiations. The measurement process is characterized by a calibration equation and counting statistics.
- 4. Relative measurements. A characteristic of these measurements is an internal standard. The measured values are expressed as ratios in which one term of the ratio is generally a standard or defined in terms of a standard through a chain of calculations. Mass spectrometry is an example of relative measurements.

Calorimetry may represent a fifth type of measurement but for the present I have not defined it as a fundamental measurement type but one which is a derived measurement based on types 2 and 4 above.

The estimation techniques for determining the constant and variable components of systematic error for measurement type 1 is the usual method involving the measurement of a standard or reference (T) a number of times during the period of interest and calculating  $\overline{X}$  and  $\sigma_{\overline{X}}$ . The estimate of the bias is  $(\overline{X} - T)$  and the random component is the variance of the bias,  $(\sigma_{\overline{X}}^2 + \sigma_T^2)$ .  $\sigma_T^2$  is the variable component of systematic error associated with the reference value T.

In the case of secondary measurements the random error associated with the calibration equation is the variable component (see Section 3.2 in Reference 11). It is not possible to routinely obtain an estimate of the bias for this type of measurement.

Counting measurements in general involve both a calibration curve and an estimate of bias obtained by periodically remeasuring the standard or reference material. Care must be exercised in combining the data from these two sources because there is a covariance factor. The variance of the standard or reference value is a component of both the estimates. Since the measurement errors are obtained using empirical data, the error associated with counting statistics is not included as a separate component as it is already included in the calibration and measurement quality control data.

I have not worked out estimation techniques for the treatment of relative and calorimetry measurements to my satisfaction at this time. If any one of the readers has this worked out I would like to hear from him.

I do not believe that the results obtained by the technique suggested in this article are different from those determined in [5]. The approach, how-ever, is different. The approach herein is empirical; it is based on the question of what can I say about measurements given a measurement process and calibration technique. Jaech in [5] developes a mathematical model and shows by example how it can be applied given a set of conditions. I agree with Jaech that in propagating the limit of error of measurements whether to treat some error as random error or systematic error variance (the variable component of systematic error) is a choice that should be made on the basis of what is meaningful in a particular application. The intent of this article is to provide additional insight into factors that enter into making this choice.

One last word on terminology. I have no problem using Jaech's term "systematic error variance" for the variable component of systematic error. On the other hand I am confused by "short-term" and "long-term" systematic error and how the latter is a bias but the former is not (Jaech [5], page 81). I suggest that the term bias be reserved to denote a fixed systematic error whose direction and magnitude are known and that we drop the "long-term", "short-term" terminology.

- The results of this approach are usually referred to as "synthetic estimates of systematic error" as opposed to "empirical estimates" which are based on statistical analysis of calibration and measurement quality control data.
- 2) Bias is defined in the usual way as the difference between the mean value of a sample statistic and a standard or reference value.

3) In this context, sampling of material is not considered a measurement as this process does not generate a measured value. Sampling, like many other processes associated with measuring does contribute to the measurement error. These can be estimated using analysis of variance techniques.

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# SOME THOUGHTS ON 'SOME THOUGHTS ON RANDOM ERRORS, SYSTEMATIC ERRORS, AND BIASES' BY JOHN L. JAECH

#### By Roger H. Moore

Editor's Note: This article was prepared for the Winter 1975 (Volume III, No. 4) issue of NMM Journal. It was inadvertently left out of the issue. This article is a companion article to "Some Thoughts on Random Errors, and Biases" by Mr. John L. Jaech which appeared in the Winter issue of this publication--Tom Gerdis.

#### I. OVERVIEW

The "thoughts" in this essay arose from my being asked to comment on a paper by John L. Jaech, "Some Thoughts on Random Errors, Systematic Errors, and Biases," prepared for the Journal of the Institute of Nuclear Materials Management. Tempted as I was to broaden the points raised to encompass a wider spectrum of literature and practice, I decided that the focus of my thoughts would be sharper by limiting them to the symbols and concepts appearing in Jaech's paper and its references. The opinions expressed herein are my own; they do not necessarily reflect those of the management or staff of the Los Alamos Scientific Laboratory or those individuals credited in Section XI with helping me clarify my thinking.

My primary concern is that certain statistical practices and definitions in the nuclear materials industry do not match up with statistical practices and definitions encountered in other industries. Such mismatches inhibit inter-industrial communication, and clarification within our own industry is difficult to achieve. Innovative statistical methods are necessary and must be encouraged, but their advocates should be certain of their internal and external consistency and that the language in which they are presented does not conflict with "established" terminology.

#### II. SOME QUOTATIONS AND INTERPRETATIONS

In my opinion, it is imperative that the nuclear industry determine whether systematic error and bias are synonyms. We can look at three of Jacch's references in an attempt to get our bearings.

In [2, p. 81], Jaech writes: "... opinions differ on the meanings of such terms as <u>bias</u>, <u>random errors</u>, and <u>systematic errors</u>. The definition presented here describes how these terms are used in this book and, by implication, indicates <u>the only reasonable way the error effects can be treated statistically</u> (italics added).... Basic to the definition is the idea of a reference set of data, i.e., the definition is meaningful only with regard to this data set.

* This work was performed under the auspices of the U. S. Atomic Energy Commission

"Definition: An error that affects only a single member of a given data set is called a <u>random error</u>. If the error affects some, but not all, members of the data set, it is called a <u>short-term systematic error</u>. If it affects all members of the data set, it is a <u>long-term systematic error</u> or <u>bias</u>.

"In this definition no distinction is made between a long-term systematic error and a bias because these quantiities differ with respect to how they may be treated statistically but not with respect to their basic meanings...." My interpretation: Jaech is saying that a <u>long-term</u> systematic error and a <u>bias</u> are not the same because they must be treated differently as statistical entities.

In [3, pp. 104-105], Mandel says: "To define accuracy, we refer, (sic) once more to a <u>well-described measuring</u> <u>process</u>, as <u>applied to a given system</u>.... [We]consider the statistical population of measurements generated by repeated application of the process of that system. We must now introduce a new concept: the <u>reference value</u> of the measured property for the system under consideration....

"Whichever way we have defined the reference value, let us denote it, for the particular property of the particular system, by the symbol R and let  $\mu$  denote the mean of the population of repeated measurements of the system. We now define the <u>bias</u> or <u>systematic error</u> of the process of measurement for that system by  $\mu$  - R, i.e., as the difference between the population mean of repeated measurements  $\mu$  and the reference value R."

My interpretation: Mandel is saying that bias and <u>systematic error</u> are indeed synonymous. Moreover, the quantity is a constant; it is not a random variable.

In[3, p. 30], Eisenhart states: "When the limiting mean  $\mu$  associated with measurement of the magnitude of a quantity by a particular process does not agree with the true value  $\tau$  of the magnitude concerned, the measurement process is said to have a systematic error, or bias, of magnitude  $\mu = \tau$ ."

My interpretation: Eisenhart agrees with Mandel's general view that <u>systematic error</u> and <u>bias</u> are synonyms and neither term refers to a random variable.

#### III. NOW WHERE ARE WE?

Based upon the short survey reported in Section 11, we are left with two choices:

(1) We can conclude that - - whatever they are -  $\underline{bi}$ as and systematic error are synonyms and that nuclear materials management literature must be carefully examined before being applied to safeguards problems.

(2) We can conclude that the distinction between bias and <u>systematic error</u> is valid and examine the consequences of making that distinction.

My own predilection is the first choice, for then we have a fairly logical connection to the concepts of accuracy (as measured by systematic error) and precision (as measured by random error).

However, for our present purposes, let us take the second choice and see where it leads us.

#### IV. JAECH'S MODEL I

Remembering that "an error of measurement may be defined as the 'magnitude and the sign of the difference between the measured value and the "true" value," we look at Jaech's Model I:

#### $x_i = \mu + \varepsilon_i$ .

We are told that  $\epsilon_1$  is called a random error. This fits the definition quoted in Section II because it appears that  $\epsilon_1$  affects only the i-th observation. But Model [ provides  $\epsilon_1$  with many more features: It is a random variable with mean 0 and variance  $\sigma_{\epsilon}^2$  and is uncorrelated with any  $\epsilon_j.$  Hence,  $x_j$  is a random variable with mean  $\mu$  and variance  $-\sigma_{\epsilon}^2.$ 

The reason for this detail is that we must go beyond the definition in Jaech's book [2] to make  $\varepsilon_{\rm i}$  a random error. It must do more than affect "only a single member of a given data set." It must be a random variable. Furthermore, according to Model I, it must have mean 0, some specified variance, and be uncorrelated with any other random errors.

#### V. ON DEFINING RANDOM ERROR

In [1, p. 7], we find: "Random Error. An error which behaves as if it were chosen at random from a population of such errors having a given frequency distribution."

Note that this definition requires nothing be stated about the mean, the variance, or the degree of correlation among the errors. Nothing is said about the "true" value being the same as the expected value of the population of measurements.

Thus, the phrase "random error" defines a particular subset of the set of entities which we call "errors." The subset of random errors may itself be further subdivided according to arbitrary criteria but all random errors most certainly are random variables. Furthermore, the definition given here tells us that any error behaving like a random variable is necessarily a random error.

VI. JAECH'S MODEL II

Turning now to Jaech's Model II:

$$x_i = \mu + \varepsilon_i + \eta_i$$
,

we are told that both  $\varepsilon_i$  and  $\eta_i$  are uncorrelated random errors with zero means and variances  $\sigma_{\epsilon}^2$  and  $\sigma_{\eta}^2$ . But, since the sum of two random variables is also a random variable, we can write  $\gamma_i = \varepsilon_i + \eta_i$  and return to a form of Model I:

 $x_i = \mu + \gamma_i$ 

where  $\gamma_{1}\ is\ a\ random\ variable\ with\ mean\ 0$  and variance

 $\sigma_{\rm X}^2 = \sigma_{\rm E}^2 + \sigma_{\rm D}^2.$ 

This conclusion follows directly from the fact that the mean of a sum of two random variables is the sum of their  $m_{eans}$  and, if they are uncorrelated, the variance of their sum is the sum of their variances.

#### VII. ERROR PROPAGATION FORMULAS

I think some value will be obtained by giving a quotation from [3, pp. 314-315], in which Ku writes:

"The results of a measurement process can usually be expressed by a number of averages  $\overline{x}$ ,  $\overline{y}$ , ..., and the standard errors of these averages  $s_{\overline{x}} = s_{\overline{x}}/\sqrt{n}$ ,  $s_{\overline{y}} = s_{\overline{y}}/\sqrt{k}$ , etc. These results, however, may not be of direct interest; the quantity of interest is in the functional relationship  $m_{\overline{w}} = f(m_x, m_y)^*$ . It is desired to estimate  $m_{\overline{w}}$  by  $\overline{w} = f(\overline{x}, \overline{y})$ 

 ${}^{\star}\mathbf{m}_{_{\mathbf{X}}}$  and  $\mathbf{m}_{_{\mathbf{Y}}}$  are the expected values of the variables x and y.

and to compute  $s_{\overline{w}}$  as an estimate of  $\sigma_{\overline{w}}$ .

"If the errors of measurements of these quantities are small in comparison with the values measured, the propagation of error formulas usually work surprisingly well. The  $\sigma_{\overline{w}}^2$ ,  $\sigma_{\overline{x}}^2$ , and  $\sigma_{\overline{y}}^2$ ... [in the following formula] will often be replaced in practice by the computed values  $s_{\overline{w}}^2$ ,  $s_{\overline{x}}^2$ , and  $s_{\overline{y}}^2$ .

"The general formula for  $\sigma_{\bar{w}}^2$  is given by

# $\sigma_{\bar{\mathbf{w}}}^2 \doteq \left[\frac{\partial f}{\partial x}\right]^2 \sigma_{\bar{\mathbf{x}}}^2 + \left[\frac{\partial f}{\partial y}\right]^2 \sigma_{\bar{\mathbf{y}}}^2 + \left[\frac{\partial f}{\partial x}\right] \left[\frac{\partial f}{\partial y}\right] \rho_{\overline{\mathbf{xy}}} \sigma_{\bar{\mathbf{x}}} \sigma_{\bar{\mathbf{y}}}$

where the partial derivatives in square brackets are to be evaluated at the averages of x and y. If X and Y are independent,  $\rho = 0$  and therefore the last term equals zero. If X and Y are measured in pairs, s  $\frac{1}{xy}$  [the covariance of  $\overline{x}$  and  $\overline{y}$ ] can be used as an estimate of  $\rho_{\overline{xy}} \sigma_{\overline{x}} \sigma_{\overline{y}}$ ."

(<u>Note</u>: This quoted material comes from the first "propagation of error" reference in the index of [3].)

As part of his discussion of his Model II, Jaech states that "the variance of x_i is the sum  $\sigma_{x_i}^2 = \sigma_{\epsilon}^2 + \sigma_{\eta}^2$ " and then states that this formula "is called an error propagation formula."

Two things are worth noting:

(1) The variance of a sum of two uncorrelated random variables is equal to the sum of the variances of the variables. This result can be obtained from fundamental statistical theory without resorting to propagation of error methods, even though the general formula reduces to this simple result for linear functions. Furthermore, the result is exact for this situation and does not require the "approximately equal" ( $\doteq$ ) symbol used by Ku.

(2) The "propagation of error" terminology, borrowed from numerical analysis, is somewhat misleading because it is not errors that are propagated in statistical endeavors; rather, the <u>variances</u> of the random variables are propagated.

Extension of these concepts to more than two variables is straightforward.

VIII. JAECH'S MODEL III

The crux of the issues surrounding bias and systematic error may be found by examining Jaech's Model III:

 $x_i = \mu + \Theta + \varepsilon_i$ .

Jaech divides his discussion into two cases, and we follow his lead.

#### Case (1)

 $\Theta$  is a constant whose value is not known, leading Jaech to call  $\Theta$  a "measurement <u>bias</u>." Since  $\varepsilon_i$  appears to play the same role it did in Models I and II, we conclude that  $x_i$  has a mean (expected value) of ( $\mu + \Theta$ ). In his discussion of Model I, Jaech gives  $\mu$  an additional characterization: " $\mu$  is the true value of the item characteristic in question."

A short table permits easy identification of the elements of Jaech's Model III with the symbols used by Mandel and quoted in Section II.

Element Description	Jaech's <u>Model III</u>	Mandel's Notation
True (reference) value	μ	R
Mean (expected) value	μ + Θ	μ
Bias ≈ (Mean value- True value)	Θ	μ - R

Thus, we see that both authors provide frameworks upon which to base analyses of data arising from the measurement of standards. But Jaech's Model III should be augmented with the information that  $\mu$  denotes the "truth;" for if it does not, then the use of  $\theta$  as a designator of "bias" does not follow.

Jaech sets up the problem of finding "some way of expressing the total uncertainty in  $x_i$ ," and then he suggests that two separate statements are possible, one pertaining to the random error standard deviation  $\varepsilon$  and the other placing a bound on the bias of the form  $\varepsilon = |\Theta| < \Theta_0$ . But then he says: "The two statements must be combined somehow in a total uncertainty statement. This can be of the form: total uncertainty in  $x_i = k \sigma_e + \Theta_o$ ." (No choice of k is given.)

I simply do not see why it is imperative to have a single statement of "uncertainty." Bias is one thing, random error variability is another. Why does it follow that a linear combination of  $\Theta_{0}$  and  $\sigma_{\varepsilon}$  is especially useful?

The venerable concept of a mean square error has been used in statistical considerations of similar questions. As Eisenhart [3, p. 39] says: "Gauss himself proposed ... that the mean square error of a procedure - - that is,  $\sigma^2 + (\mu - \tau)^2$ , where  $\sigma$  is its standard deviation; and  $\mu - \tau$ , its bias - - be used to characterize its accuracy." Eisenhart then goes on to show that mean square error "clearly does not 'tell the whole story.""

Consider the analagous problem of comparing two right triangles when only their hypotenuses are known. Absolutely nothing can be inferred about their respective bases, altitudes, or areas.

I submit that the two-statement presentation is better than trying to find a single statement to summarize uncertainty. Given the two statements, a user then can combine the information as he sees fit for his particular purposes.

#### Case (2)

Jaech writes:

" $\Theta$  is randomly selected from a population that has zero mean and variance denoted by  $\sigma_{\Theta}^2$ . In this case,  $\Theta$  is called a <u>systematic error</u> by the author, and  $\sigma_{\Theta}^2$  is called a <u>systematic error variance</u>. Note that  $\Theta$  differs from a random error only in the sense that the same value of  $\Theta$  applies to all observations in question, whereas  $\varepsilon_i$  is different for all i."

Later, in discussing the distinctions between Case (1) and Case (2), Jaech alludes to fixed- and random- effects analysis of variance models and says: "By analogy, I think of bias as being a fixed effect and a systematic error as representing a random effect."

This is troubling. If systematic errors now represent random effects, why aren't they random errors also? They most certainly qualify under the terms of the definition given in Section V. Surely a random variable affecting two or more expressions of a model is no less random for having such a multiple effect.

More serious, I think, is Jaech's statement that "the expected value of  $x_i$  is  $\mu$  and its variance is  $\sigma^2 + \sigma^2_2$ ." What does Model III (with  $\Theta$  a random variable) buy that Model II did not? Perhaps I am anticipating the estimation problem, which Jaech eschewed for his paper.

#### IX. JAECH'S MODEL IV AND LIMIT OF ERROR

Jaech's Model IV is an extension of his Model III to m pairs of systematic and random errors. Because his Model III leaves a number of questions of technique, especially when compared with "established" methods, I find it difficult to know what to say about Model IV. For example, as soon as he states in the discussion of Model III with  $\Theta$ called a bias that " $\Theta$  is known with high probability to be smaller in absolute value than some value  $\Theta_0$ ," we are inferring that  $\Theta$  is a random variable. Why, then, not deal only with Case (2), in which  $\Theta$  clearly is said to be a random variable? Statements to the effect that, in Model IV, we are forced to assume that biases are uniformly or normally distributed do not seem to me to follow from any logical argument. (And, if we really were in the wonderful world of Bayesian statistics, a considerably different development should be followed.)

In general, incidentally, propagation of mean square errors does not follow from propagation of variances. Squaring a bias is <u>not</u> the same as finding a variance. So that the "root-mean-square" approach to computing limits of error simply isn't good enough.

#### X. CONCLUDING REMARKS

Jaech states "that there is no 'right' way to treat the combined effect of systematic errors or biases." I agree. And, although I admire the directness with which he tackles the problem, I am forced to conclude that he is dealing with a paper tiger. Why are we forced to combine systematic errors and/or biases into a single statement to get an overall uncertainty? Are we not really much better off to have the individual components in hand and display them for the user so that he, too, will understand what's happening?

Finally, I do not understand why Jaech states: "In advocating the root-mean-square approach, I am influenced by the familiar central limit theorem of mathematical statistics that implies to me that the systematic errors ... will tend to cancel out." I offer for your consideration the comment of the famed probability, William Feller, which appears in <u>An Introduction to Probability Theory and Its Applications</u>, Volume II, John Wiley and Sons, New York, 1966, p. 252:

"The central limit theorem establishes conditions under which sums of independent random variables are asymptotically normally distributed." Feller shows even that forms of the theorem can be made to work for random variables "with infinite variances."

#### XI. ACKNOWLEDGEMENTS

John Jaech was kind enough to let me see an early draft of his paper, as well as the revised version. He encouraged the preparation of this paper, so that some of the several issues could be displayed for the readers of this journal. I believe his paper presents the clearest statement we have yet seen of some of the confusing information about the application of statistics to the serious problems in nuclear materials control. I look forward to the readers' responses to these papers and to other papers taking up some of these points from both the theoretical and applications viewpoints.

My own thinking has been greatly clarified by the help of a number of acquantances, particularly Y. M. Ferris of Dow Rocky Flats and R. J. Beckman, G. L. Tietjen, and D. B. Smith, all of the Los Alamos Scientific Laboratory. Having named a number of conferees, I am compelled to remind the reader of the inequities involved in the old guilt-by-association gambit.

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#### Nuclear Materials Management Journal of INMM

#### Volumes I, II, and III

Vol. I, No. 1, April 1972

- Roy G. Cardwell, "Control of Nuclear Materials in Research: A Special Management Problem," 8-10.
- R.A. Bradley and J.D. Sease, "Design and Operation of a Plutonium Laboratory," 11-14.

#### Vol. 1, No. 2, July 1972

- William J. Gallagher, "Isotopic Neutron Source Assay Systems: Their Advantages and Disadvantages," 7-9.
- F.A. Costanzi and R.B. Leachman, "Safeguards at Kansas State University: Part I," 10-12.
- James E. Lovett, "Nuclear Material Cost Accounting," 13.
- John L. Jaech, "A New Approach to Calculating LE-MUF," 14-17.

#### Vol. I, No. 4, January 1973

- W.F. Heine and J.D. Moore, "Rapid Assessment of U-235 from Uranium Fuel Fabrication Facilities," 9-11.
- B.D. Sinclair, S.F. Send, H.J. Fenech, and P.K. Shen, "S.C.E.N.I.C.—Southern California Edison Nuclear Inventory Control," 12-16.
- F.A. Costanzi and R.B. Leachman, "Safeguards at Kansas State University: Part II," 17-24.

#### Vol. II, No. 1, Spring 1973

J.A. Powers, "Materials and Plant Protection Standards," 9-10.

Frederick Forscher, "Perspectives on the Energy Crisis," 11.

L.E. Minnick, "Nuclear Power," 12-13.

- E.R. Johnson, "Plutonium," 14-20.
- D.E. Christensen and D.L. Prezbindowski, "Isotopic Correlation Safeguards Techniques: Reactor Characteristics as Observed from Measured Spent Fuel Data," 21-55.

#### Vol. II, No. 2, Summer 1973

Lester Rogers, "AMERICA'S ENERGY NEEDS," 13-15.

- John T. Caldwell, "New Technique for U-235 Enrichment Determination in UF6 Cylinders," 16-20.
- Herman Miller, "Useful Techniques for Quality Assurance and Material Protection," 21-22.
- J.P. Odom, N.D. Eckhoff, and Walter Meyer, "Optimal Nuclear Material Transportation Route Selection," 23-31.
- John L. Jaech, "Pitfalls in the Analysis of Paired Data," 32-39.

#### Vol. II, No. 4, Winter 1974

- Seymour H. Smiley, "Quality Assurance in the Nuclear Fuel Cycle," 8-12.
- W.F. Heine and J.D. Moore, "Rapid Assessment of U-235 in Used High Efficiency Particulate Air Filters," 13-15.

John L. Jaech, "Control Charts for MUF's," 16-28.

#### Vol. III, No. 1, Spring 1974

K.B. Stewart and R.A. Schneider, "Verification Sampling Techniques in a Safeguards Situation," 12-19.

Frederick Forscher, "Today's Need: Energy Managers," 20.

F.A. Costanzi, "Some Electronic Security Devices," 21-24.

R.L. Delnay, "N15 American National Standards," 25-28.

#### Vol. III, No. 2, Summer 1974

- Hans J. Weber, "Nondestructive Assay of Plutonium and Uranium in Mixed-Oxides," 22-30.
- R.E. Tschiegg, "A Computerized Records and Reports System," 31-36.
- Eugene J. Miles, "The Invisible Man(agers)," 37-38.

Manuel A. Kanter, "Safeguards Back to the Forefront," 39.

- John L. Jaech, "Testing for Normality When the Data Are Grouped Due to Rounding," 40-46.
- E.A. DeVer and W.W. Rodenburg, "Mound Laboratory: A Leader in Nuclear Materials Management," 47-49.

#### Volume III, No. 4, Winter 1975

- Dennis W. Wilson, Chmn., "Comments on G.E.S.M.O. by INMM Safeguards Committee," 20-23.
- James E. Lovett, "Concepts of Real Time and Semi-Real Time Material Control," 24-30.
- James A. Powers and LeRoy R. Norderhaug, "Materials and Plant Protection Standards: Revised," 31-35.
- John L. Jaech, "Some Thoughts on Random Errors, Systematic Errors, and Biases," 37-39.
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